University of Nottingham

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Leaching of Alternative Pavement Materials

By

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To My Family

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Abstract

Road pavements are one of the highest volumetric consumers of raw material in any industry and aggregate is the majority material for such constructions and associated bulk fill. Traditionally, aggregates have been sourced from primary crushed rock and sand and gravel. However, many alternative materials, such as by-products from industry, mining and other activities, have been shown to form mechanically suitable replacements for primary aggregates, either used solely or in combination with conventional or alternative binders. The remaining hindrance to the use of such alternative materials is the possibility that they contain unfavourable contaminants that could leach into the surrounding environment and cause pollution.

In this thesis, the leaching of chemical species from a range of conventional and alternative aggregate materials and mixtures is investigated through a variety of characterisation and leaching tests. The test methods are developed and applied to investigate how the physical conditions of usage, such as material grading, compaction and binder treatment, and the in-situ hydraulic regime, influence the leaching process and any resulting potential to impact on the environment.

The effect of different binder treatments on the leaching properties of the materials and mixtures is assessed using the laboratory procedures developed. A method for selecting appropriate binder treatments to control the leaching of chemical species of concern is proposed.

The in-situ leaching conditions of the road construction are simulated in large-scale lysimeter tests. The relationship observed between the laboratory and field tests is used to develop a method for predicting likely in-situ leaching performance from the results of a simple laboratory test. A generic leaching assessment procedure is defined that should encourage the use of alternative materials in road construction and bulk fill.

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Declaration

The work described in this thesis was conducted at the University of Nottingham, School of Civil Engineering between January 1997 and May 2004. I declare that the work is my own and has not been submitted for a degree of another university.

1 Introduction and Objectives

1.1 Background

An abundant supply of aggregate material is essential to the continued development and maintenance of a modern transport network. Road pavements are one of the highest volumetric consumers of raw material in any industry and aggregate is the majority material for such constructions and associated bulk fill. In 1998, the estimated consumption of primary aggregates in the UK was about 210 million tonnes (Mt) [British Geological Survey, 2000] with approximately a third being used in road construction and maintenance [Sherwood, 1995]. Dawson and Mundy (1999) revealed that across Europe an estimated 750 million tonnes of aggregates per year are consumed in the unbound layers of roads, whilst the total annual consumption of aggregates by roads throughout Europe may be estimated at around 1 billion tonnes.

At present, in the UK, the majority of aggregate materials are obtained from primary rock or sand and gravel resources. Such aggregates are generally extracted from the ground via quarrying operations. The use of primary materials results in the depletion of a valued finite resource, whilst their extraction from the ground, causes many adverse environmental impacts, including noise, dust, traffic, visual intrusion, loss of amenity, damage to biodiversity and the generation of derelict land [DETR, 2000a].

In the UK, industrial and domestic activities generate in excess of 200 million tonnes of waste and by-product materials each year that require management and/or disposal [DETR, 2000b]. In addition, further stockpiles of such wastes are estimated in the region of 4,500 million tonnes [Whitbread et al., 1991]. Many material by-products from industry (power station ash, metalliferous slag, foundry sand), mineral extraction (china clay sand, colliery spoil, slate waste), construction (brick, concrete) and incineration (municipal solid waste incinerator ash, sewage sludge incinerator ash), may be suitable for use as an alternative to natural aggregates in road construction and bulk fill.

Multiple environmental benefits would be gained from the use of alternative aggregates as a substitute for primary materials, including:

- a reduction in primary quarrying activity (reduced noise, dust and land consumption);
- a reduction in development of new waste stockpiles and re-use of material in existing piles;
- clearance and reduction of derelict land generated through waste disposal;
- economical disposal or recycling of marginal materials and,
- a reduction in the utilisation of finite natural resources, thus addressing the aims of sustainable development [Nunes, 1997; Department of the Environment, 1993].

Despite these attractive benefits, there are a number of hindrances to the use of alternative materials in road construction applications. Firstly, the material must possess suitable mechanical properties for the particular utilisation application. Nunes (1997) and Sherwood (1995), among others, have demonstrated that many alternative materials can form mechanically suitable replacements for aggregates in road construction or bulk filling tasks, either, when used solely or in combination with other aggregates and/or binders to modify or improve performance. The second hindrance, which is addressed by this thesis, is the possibility that the use of an alternative aggregate may cause unacceptable degradation of the quality of the hydrological environment surrounding the construction.

Many alternative materials contain constituents that may potentially be a source of contamination for ground and surface water systems. Environmental protection agencies and organisations around the world are setting and enforcing increasingly stringent environmental targets for discharges to the environment. In order to pass judgement on the acceptability, or not, of any aggregate material for use on environmental grounds, an appropriate and reliable approach for assessing the in-situ leaching performance of such materials is required. This is the principal subject of this thesis.

The approach taken in this thesis is to define and verify a logical material assessment procedure focusing on a tiered series of material characterisation and leaching tests. The proposed procedure is used to investigate how the physical in-situ conditions of usage, such as material grading, compaction and binder treatment, and the in-situ hydraulic regime, influence the leaching process and the nature of any resulting impact on the environment. The proposed assessment procedure should thus encourage the use of alternative materials in road construction.

1.2 Aims and Objectives

The principal aims of the research presented in this thesis were:

- To determine an appropriate testing strategy (based on existing methods) for assessing the leaching potential of primary and alternative aggregate materials in a road utilisation scenario;
- To propose, where required, suitable binder treatment methods that will beneficially modify the leaching potential of the aggregate material.

In order to achieve the aims described above, the following objectives were defined:

- To select suitable candidate materials for investigation;
- To review available leaching tests in the context of application to materials for road construction and other bulk engineering uses;
- To perform appropriate leaching tests on primary and alternative aggregate materials at conditions simulative of use;
- To define a logical material assessment procedure using these tests and verify the procedure for reliability and ease of use;
- To use the procedure to assess binder treatments which are likely to reduce the leaching potential;
- To confirm that the assessment procedure is suitably simulative of the in-situ condition via the use of field-scale trials;
- To produce a generic guide to leaching assessment and binder treatment strategy.

1.3 Thesis Content

This thesis is organised into ten chapters, of which this introduction is the first. Chapter 2 provides a review of the academic disciplines of relevance to the research objectives. To put the research into context the road construction and traditional aggregate industry are considered. This is followed by a brief review of candidate alternative materials and binder treatments. The concept of leaching is introduced in terms of the controlling physical and chemical variables and current methods of assessment are reviewed. The chapter is completed with a summary of applicable legislation.

Chapter 3 describes the selection of the candidate primary and alternative aggregate materials and binders used in the research. Preliminary material characterisation is described in terms of the chemical composition of the solid material fraction and leachability is defined using a rapid leaching characterisation test.

In Chapter 4 laboratory tank-leaching tests are used to examine leaching by diffusion processes from the aggregates and binders. The effects of material particle size and compaction on the rate and quantity of leaching are investigated. The chapter defines each leaching test procedure, the design and manufacture of the required laboratory equipment and discusses the results obtained.

Chapter 5 describes the laboratory permeameter tests that are used to investigate leaching by advection processes. Again the leaching test procedures are defined and the results presented and discussed.

In Chapter 6 the laboratory leaching test procedures are used to investigate leaching from a range of realistic material mixtures incorporating binders. Conclusions are drawn regarding the effect of the binder treatments on the leaching properties.

Chapter 7 describes the large-scale field simulations that were performed to provide verification of the applicability of the laboratory testing. The design and manufacture of the tests are described and the results presented and discussed.

The relationship between the leaching observed in the field simulations and laboratory testing is examined in Chapter 8. The patterns of leaching observed from the range of tests are discussed and an empirical relationship between laboratory and in-situ leaching is proposed.

In Chapter 9 a leaching assessment procedure is proposed based on laboratory leaching tests. The chapter includes a guide to the selection of suitable binder treatments for modifying leaching performance. The overall conclusions from the research are summarised in Chapter 10 along with recommendations for future work.

2 Literature Review

2.1 Introduction

The application of alternative aggregate materials in road construction is dependent on being able to demonstrate that such utilisation will not result in any deleterious effects to the road adjacent environment. However, the assessment of this environmental impact raises a multidisciplinary problem. This chapter gives an introduction to these disciplines. Initially the traditional aggregates industry is considered, the alternative materials are introduced in terms of their potential for use and the typical design and construction of a road pavement is presented. The mechanisms for the transfer, or leaching, of contaminants from a solid to a liquid are summarised. Existing methods of assessing potential contaminant release are discussed in relation to the different types of test variable and the applicability of such test methods to the scenario of road construction materials. A review of previous and current research into leaching of alternative aggregate materials is presented including a review of test methods and their interpretation. The literature review is completed with an examination of the water regimes in road pavements that could provide pathways for chemical species into the wider environment, together with a summary of the legislation and regulations of relevance to the use of alternative materials in road construction.

2.2 Aggregate Materials

2.2.1 Traditional Aggregate Materials

Annual aggregate production in Great Britain rose from 110 million tonnes in 1960 to nearly 300 million tonnes in 1990 and in 1997 the annual production was 218 million tonnes [Quarry Products Association, 1998]. Trends in the UK aggregates markets are illustrated in Figure 2-1.

Traditionally, aggregate materials have been sourced from naturally occurring geological formations. A summary of the 1997 sources of these primary aggregates is given in Table 2-1.

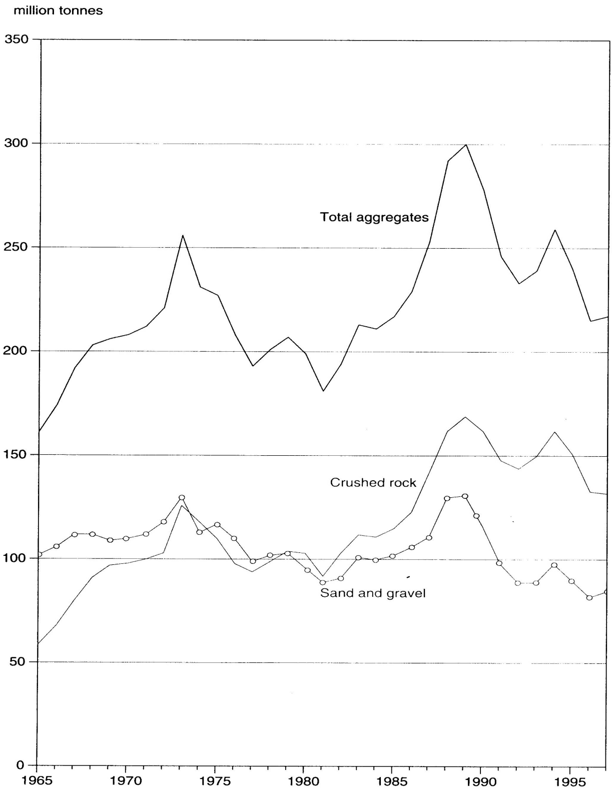


Figure 2-1: Annual Great Britain production of natural aggregates 1965–1997 [Quarry Products Association, 1998]

Table 2-1: Production of aggregates in Great Britain in 1997, by source [Quarry Products Association, 1998]

	Aggregate	Million Tonnes
Crushed Rock	Limestone	77
	Igneous rock	44
	Sandstone	13
	TOTAL	**133**
Sand & Gravel	Sand	43
	Gravel	39
	TOTAL	**85**
TOTAL AGGREGATE PRODUCTION		**218**

In 1997, Great Britain used the 218 million tonnes of crushed rock sand and gravel in uncoated roadstone (19.0%), coated roadstone (13.9%), concrete (32.3%), fill (28.2%), ballast (1.0%) and other uses (5.6%). Of the 135 million tonnes of crushed rock, over 50% was used as roadstone [British Geological Survey, 1998].

The vast majority of primary aggregates are obtained by using opencast mining methods. This process causes disturbances during operation and often results in permanent damage to the landscape. The extent of the environmental impact varies depending upon the mining method utilised, but the majority of public concerns are common to all methods and include:

- The various effects of road traffic, particularly where this is the primary means of transport;
- The effects of blasting, noise and dust, the level of which will vary according to the type of material and the amount of overburden;
- Visual and landscape effects;
- Effects on the water environment including contamination of surface water discharges by solids or by oil and its derivatives and reduction of flow to wells and streams [DETR, 2000a].

Over the past twenty years environmental pressures have resulted in restrictions on extraction planning consents, especially in densely populated areas such as south-east England. These restrictions have encouraged investment in large scale, hard rock Superquarries and marine dredging, backed up by sea and rail transportation networks. Such large scale operations and the transportation of these low-value commodities over such large distances introduces different, but equally challenging, environmental impacts.

2.2.2 Alternative Aggregate Materials

In the UK, industrial and domestic activities generate in excess of 200 million tonnes of waste and by-product materials each year that require management and/or disposal [DETR, 2000b]. In addition, further stockpiles of such materials are estimated in the region of 4500 million tonnes [Whitbred et al., 1991]. Many waste and by-product materials have been shown to form mechanically suitable replacements for primary aggregates in many applications and if utilised would reduce the requirement for primary aggregate extraction.

The term alternative aggregate material is used in this thesis to describe any aggregate that was not directly obtained from a natural geological source or was not mined specifically for the purpose of use as an aggregate material. Alternative aggregate materials may be derived from the wastes, discarded materials or by-products of industrial, mining or domestic processes.

There are a wide range of materials that have potential for use as alternative aggregate materials or as binders or fillers. The materials most commonly referred to in current literature include [Nunes, 1996]:

Municipal and Domestic Materials

- Municipal solid waste incinerator ashes- fly ash and bottom ash;
- Sewage sludge incinerator ashes;
- Tyres;
- Glass;
- Plastic.

Industrial Materials

- Construction and demolition materials- crushed concrete, crushed brick, crushed rubble, unsorted demolition debris, asphalt planings, roofing shingle;
- Power station ashes- pulverised fuel ash, furnace bottom ash;
- Metallurgical processing materials- ferrous slags (blastfurnace and steel slags), non-ferrous slags, foundry materials, steel mill materials;
- By-product gypsum- flue gas desulphurisation gypsum, fluorogypsum, phosphogypsum;
- Cement and lime kiln dust;
- Petroleum contaminated soils;
- Recycled and waste fibres.

Mining and Mineral Materials

- China clay spoil;
- Colliery spoil- unburnt colliery shale (minestone), burnt colliery shale;
- Quarry spoil;
- Slate spoil;
- Spent oil shale;
- Mine refuse;
- Tailings.

2.2.2.1 Utilisation of Alternative Aggregate Materials

Throughout much of Europe, a significant level of material recycling is being achieved. Many countries have introduced systems to encourage this usage. Such systems include taxation on waste disposal or primary aggregates extraction, or setting targets for recycling and for using recycled materials. The introduction of the EC Landfill Directive [99/31/EC, 1999] has set legislative requirements to reduce the amount of waste going to landfill, and as such, should act as a driver for the increased utilisation of alternative aggregate materials. A summary of the utilisation of a range of alternative materials was given by Nunes (1996) and is shown in Table 2-2 for a selection of European countries.

Table 2-2: Summary of the annual production and utilisation of some alternative materials in European Countries [Nunes, 1996]

		Material					
Country		**PFA**	**FBA**	**IFA**	**IBA**	**CS**	**CDW**
Belgium	Production (x10^3 tonnes)	850	106	25	300	0	3000
	Quantity Used (x10^3 tonnes)	723	94	0	0	-	-
	Percentage Used (%)	85	89	0	0	-	--
Germany	Production (x10^3 tonnes)	-	400	270	2600	64800	32600
	Quantity Used (x10^3 tonnes)	-	300	0	1800	13600	3700
	Percentage Used (%)	100	74	0	69	21	11
Denmark	Production (x10^3 tonnes)	890	100	-	415	-	-
	Quantity Used (x10^3 tonnes)	783	88	-	270	-	3670
	Percentage Used (%)	88	88	-	60	-	-
France	Production (x10^3 tonnes)	2535	447	200	1800	4700	20-25000
	Quantity Used (x10^3 tonnes)	1175	>224	-	-	7990*	-
	Percentage Used (%)	70	>50	-	-	170*	-
Britain	Production (x10^3 tonnes)	10500	2600	-	160	45000	24000
	Quantity Used (x10^3 tonnes)	4600	1140	-	-	2800	11000
	Percentage Used (%)	44	44	-	-	6	46
Netherlands	Production (x10^3 tonnes)	970	100	85	650	-	8000
	Quantity Used (x10^3 tonnes)	970	70	35	46	-	>4000
	Percentage Used (%)	100	70	41	7	-	>50

Key:				
	PFA	Pulverised Fuel Ash	CS	Colliery Spoils
	FBA	Furnace Bottom Ash	CDW	Construction and Demolition Waste
	IFA	Incinerator Fly Ash	-	No information
	IBA	Incinerator Bottom Ash	*	Including use of stockpiled materials

Alternative materials have the potential to be used in many different applications as a substitute for the primary aggregates that have traditionally been used. Some of the most significant uses include:

- embankments, as bulk and selected fill;
- unbound roadbases, sub-bases and capping layers;
- cement bound material for sub-bases and roadbases;
- bitumen bound material for sub-bases and roadbases;
- as a filler in bituminous materials;
- drainage layers and as a lightweight drainage material behind structures;
- gabions;
- pipe bedding;
- lightweight block making;
- production of lightweight aggregate;
- structural concrete and building mortar;
- cement manufacture;
- as a partial replacement for cement;
- as a partial replacement for sand in grading correction.

This thesis focuses on the application of alternative aggregate materials in road construction and bulk fill.

2.2.3 Aggregate Material Binders

Binder treatments, such as cement, lime and bitumen, have traditionally been used to modify the mechanical properties of aggregate materials to make them suitable for use in a variety of applications. Binders have also been shown to be a useful addition to many alternative aggregate materials to enable their use in higher specification applications [Nunes, 1997].

The most commonly used binders can generally be categorised as hydraulic, pozzolanic or bituminous. In addition to the conventional binders, a number of alternative materials exhibit binding properties. The three categories of binder are described below.

2.2.3.1 Hydraulic Binders

Hydraulic binders react with water to produce a cementitious material. Hydrated calcium silicates are responsible for most of the gain in strength as they form the strong part of the hard material. Hydrated calcium aluminates can also contribute to strength gain [Scott, 1991]. The hydration of hydraulic binders often generates heat that increases the rate of hardening.

Cement is the most commonly used hydraulic binder. It is produced in a high temperature process (about 1450°C), where calcarium, clay and other materials rich in silica, alumina and iron, produce Portland Clinker. The clinker is ground and mixed with other additives to produce different types of cement.

Cement kiln dust is a by-product of the cement manufacturing process. It does however exhibit some hydraulic binding properties and has been used successfully in mixtures incorporating cement and pozzolanic binders.

2.2.3.2 Pozzolanic Binders

Pozzolanic materials contain silica (SiO_2) and siliceous or aluminous minerals. The hardening reactions that form calcium silicates and/or calcium aluminates require a source of calcium oxide and water to act as activators for the reactions. Pozzolanic reactions are very sensitive to curing temperature. At temperatures below 10°C reactions are very slow and at 40°C, they occur 10 to 20 times faster than at 20°C. The presence of organic matter or nitrates can inhibit the reactions, sulphate or calcium carbonate can accelerate reactions and sulphates and sulphides can increase swelling.

Lime is commonly used as an activator for pozzolanic activity but does not have any cementitious properties by itself. Used solely, lime modification reactions with clay minerals, can reduce material plasticity and shrinkage and increase workability, traffickability, compactability and bearing capacity [Sherwood, 1993].

Cement can also be used as an activator for pozzolanic reactions. The calcium hydroxide produced as a reaction product in the hydration of the cement, can provide the calcium source for the pozzolan [Sherwood, 1995].

A number of alternative materials possess pozzolanic properties. The most studied examples are pulverized-fuel ash and ground and ground granulated blastfurnace slag. Alternative activators include cement kiln dust and flue-gas desulphurisation gypsum.

2.2.3.3 Bituminous Binders

Bitumen is a mixture of high molecular weight hydrocarbons, obtained as a residue from petroleum or coal-tar refining. Bitumen is a viscous fluid which softens gradually when heated. It is black or brown in colour and possesses waterproofing and adhesive properties. Bitumen is commonly used as a binder in road construction, especially in road surface dressing [BS 3690: Part 2, 1989], wearing course and, in some cases, base course and road base.

This thesis focuses on the utilisation of alternative aggregate materials in road construction and bulk fill applications both solely and in combination with a range of binder treatments such as those described above.

2.3 The Road Construction

In the UK, pavement design conforms to the guidance and specifications described in the Design Manual for Roads and Bridges [Department of Transport, 1994] The design is focused on achieving an adequate mechanical performance for the predicted quantity of traffic over the lifetime of the road. Materials with high strength and resistance to permanent deformation reduce the need for expensive maintenance.

In the UK, roads are constructed from a number of pavement layers. Flexible and rigid constructions are the most common and are illustrated schematically in Figure 2-2 and Figure 2-3 respectively. Flexible pavements have bitumen bound surface layers whilst rigid pavements use high quality concrete surfacing. A third method of construction, known as the flexible composite design, consists of a bituminous upper layer supported on a base of cement-bound material.

Each layer in the pavement requires higher specifications, in terms of bearing capacity and durability, than the layer immediately below it. Building in layers generally reduces overall costs and enables a range of construction materials to be used, particularly for lower specification fill and capping applications.

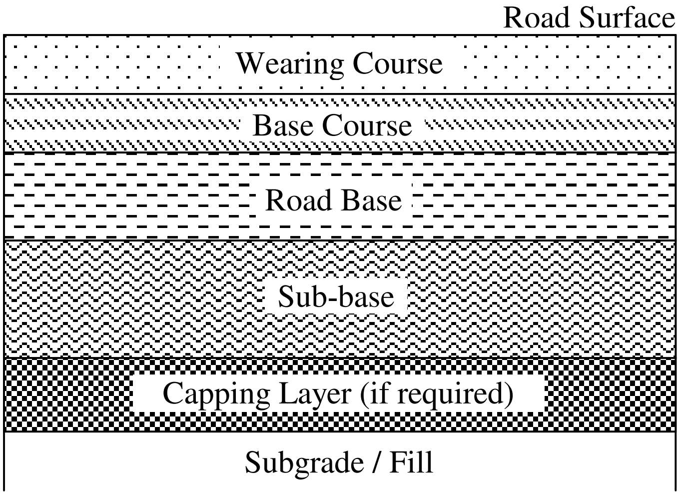


Figure 2-2: Typical flexible pavement construction (not to scale) [Sherwood, 1994b]

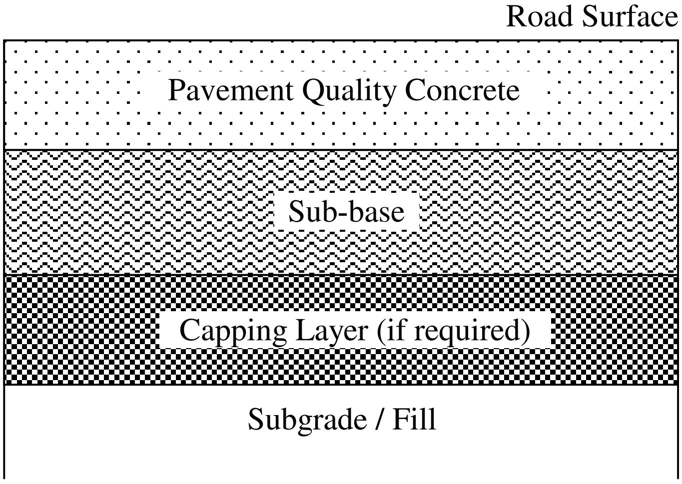


Figure 2-3: Typical rigid pavement construction layers (not to scale) [Sherwood. 1994b]

Bulk fill material is required where the existing ground level needs to be raised or lowered. Although the design engineers aim to balance the amount of ‘cut’ and ‘fill’ on any particular scheme, it is sometimes necessary to import large volumes of fill material. Bulk fill material must be inexpensive, readily available, relatively easy to transport, place and compact and once compacted, it must be strong and stable enough to support the layer above it. A wide range of materials are considered to be acceptable, including some alternative materials.

The capping layer is used to support the sub-base when constructing on a weak subgrade. The capping layer also protects the subgrade from water and frost penetration and maximises the load spreading capabilities of the sub-base. The capping layer is usually made from low cost granular or stabilised material [Sherwood, 1994b], thereby providing opportunities for the use of many alternative materials in an unbound or bound form [Sherwood, 1995].

The sub-base layer must:

- provide a platform on which to transport, lay and compact the paving materials;
- perform as a structural layer that spreads the wheel loads;
- act as an insulating layer to protect the subgrade;
- contribute to road drainage.

In flexible pavements the sub-base may be constructed from unbound granular material, which is generally categorised as either Type 1 or Type 2. Type 1 materials are the highest quality and include crushed rock, slag or concrete, and well-burnt non-plastic shale. Type 2 materials are of lower quality and have less demanding specifications (especially in terms of particle size distribution). Type 2 materials include those permitted for Type 1 and also natural sand and gravel. In rigid (and heavily trafficked flexible) constructions stabilised materials are often used in the sub-base.

The road-base is the most important structural layer in a flexible pavement and is designed to distribute the surface loads so that the bearing capacity of the subgrade is not exceeded. Road-base material needs to be of a high quality to provide the pavement with added stiffness and resistance to fatigue. Unbound road-bases are not commonly used in the UK but elsewhere they are widespread. Road-base bound with cement or bitumen is more common. In rigid pavements no road-base is required since the concrete paving is laid directly on top of the sub-base.

The surface layer of the pavement is subject to the most stringent specifications. Flexible pavements have a bitumen-bound surfacing whilst rigid pavements use pavement quality concrete. It is possible to use some alternative materials as aggregate or filler in the concrete or rolled asphalt.

The physical and mechanical properties of the materials used in each layer of the pavement are subject to material specifications. For unbound granular materials, the majority of specifications relate to the physical or mechanical properties of the aggregate such as particle size distribution, particle strength, density, and shape, frost heave, water adsorption and durability [BS 812, 1975, 1985, 1989, 1990]. These traditional methods of assessment have been developed from years of experience using conventional primary aggregates. Promoters of alternative aggregate materials have criticised this approach since it excludes the use of many alternative materials that may provide satisfactory in-situ performance but have non-conventional particle properties. Nunes (1997) amongst many others have developed performance based specifications and have shown that many alternative materials could perform in a similar manner to conventional aggregate material, either solely or when treated with the addition of binding materials.

Sherwood (1995) summarised the potential for use of a range of alternative materials in different road construction applications. This summary is given in Table 2-3.

Table 2-3: Summary of potential uses of alternative materials in road construction [Sherwood, 1995].

Material	**Bulk Fill**	**Unbound Capping**	**Unbound Sub-base**	**Cement-Bound Material**	**Concrete Aggregate or Additive**	**Bitumen Bound Material**	**Surface Dressing Aggregate**
Crushed Concrete	High*	High	High	High	High	Some	None
Asphalt Planings	High*	High	High	Low	None	High	None
Demolition Wastes	High	Some	Some	Low	Low	Low	None
Blastfurnace Slag	High*	High	High	High	High	High	High
Steel Slag	Low	Low	Low	Low	Low	Some	High
Burnt Colliery Spoil	High	High	Some	High	Low	Low	None
Unburnt Colliery Spoil	High	Low	None	Some	None	None	None
Spent Oil Shale	High	High	Some	High	Low	Low	None
Pulverised Fuel Ash	High	Low	Low	High	High	None#	None
Furnace Bottom Ash	High	Some	Some	High	Some	Low	None
China Clay Sand	High	High	Some	High	High	Some	Low
Slate Waste	High	High	High	Some	Some	Low#	None
Incinerator Ash	High	Some	Some	None	Some	None	None

*Suitable but inappropriate (wasteful) use

PFA and slate dust can be used as filler

Due to the less stringent specification requirements of the sub-base and capping layers, it is in these layers that alternative materials have most potential for use. These layers, together with imported fill, also consume the largest quantities of aggregate in pavement constructions.

The current UK road design specifications contain little reference to the required chemical properties of the aggregate materials. The exceptions to this are those chemical properties that have a detrimental affect on the mechanical performance of the material, such as sulphate that may cause swelling, rather than for environmental protection.

It is not the objective of this thesis to set specifications for permissible concentrations of elemental species in aggregate materials or their leachates. Any such limits should be set at the discretion of the relevant environmental protection authorities. However, the aim is to recommend a suitable protocol by which any material can be assessed for its environmental suitability in a similar way to that required for mechanical performance.

2.4 Leaching in the Environment

Leaching may be defined as "the extraction of a soluble material from an insoluble solid by dissolution in a suitable solvent" [Sharp, 1990]. The process of leaching encompasses the physical, chemical and biological reactions that mobilise a contaminant as well as the transport mechanisms that carry the contaminant away from the matrix. The solvent or fluid that initiates the leaching process is called the leachant, and the resulting fluid containing the soluble material is termed the leachate.

In the road construction scenario, the alternative aggregate materials forming the structural layers of the pavement contain a variety of soluble constituents. The solvent that will extract the soluble constituents may take the form of rainwater percolating through the road structure, surface runoff seeping in from the road margins or groundwater entering the structure from the surrounding environment. The resulting leachate will move into the wider environment where the acceptability of contaminant loading will be assessed by the relevant environmental protection authority.

The mechanisms by which the soluble material enters and/or leaves the solvent are a complex combination of physical, chemical and biological processes. For further details the reader is referred to water chemistry texts such as van der Sloot, Heasman and Quevauviller (1997), Dojlido and Best (1993) and Conner (1990) amongst many others. A summary of the factors influencing leaching of waste products is given in Table 2-4 and the majority of factors could equally be applied to the leaching of alternative aggregate materials. The understanding of leaching behaviour is still a relatively new field of research and much of the reported literature has been carried out since the commencement of the research presented in this thesis. The processes are complex and interrelated and vary between different materials and are currently not fully understood.

Table 2-4: Factors influencing leaching [adapted from van der Sloot, de Groot and Wijkstra (1989) and van der Sloot, Heasman and Quevauviller (1997)]

Chemical Factors	Physical Factors	Biological Factors
• pH of the material and/or leachant; • Redox condition; • Leachability of the chemical species; • Chemical speciation in the material matrix; • Chemical interactions in the pores and at the surface; • Changes in the chemical environment (pH, redox) in the material with time; • Surface dissolution; • Chemical speciation in the pore water; • Reaction kinetics; • Chemical composition of the leachant; • Complexation with inorganic or organic compounds;	• Particle size and therefore surface-to-volume ratio of the material; • Particle shape; • Porosity; • Matrix and/or particle permeability; • Pore structure; • Continuous or intermittent contact with water; • Temperature in relation to diffusion rate and with respect to durability (freeze/thaw); • Density differences in the material matrix (e.g. gravel in concrete); • Homogeneity or heterogeneity of the solid matrix in terms of mineral phases; • Time; • Hydrogeological conditions.	• Colonisation; • Material degradation by boring organisms; • Pore clogging by biological substances; • Changes in the chemical environment due to biological activity (redox).

2.4.1 Leaching Processes

The processes that occur at the interface between the solid and liquid phases and within the bulk solid and the bulk liquid during the leaching process are illustrated in Figure 2-4. The process of leaching is driven by concentration gradients. These gradients may occur between or within the individual solid or aqueous phases. Theoretically, molecular movement will occur along the concentration gradient until equilibrium conditions are established [Alexander et al., 1994; Conner 1990].

On initial contact with a leachant, rapid leaching of the solid matrix may occur as a result of surface phenomena. The surface of each material particle may contain deposits of salts derived by sublimation, condensation or the expulsion of elements from sample particles. These surface components can be readily leached upon contact with a leachant in a process known as *surface wash-off*. This process results in rapid initial leaching followed by much slower release. *Surface dissolution* is another surface process where elements from the surface layer of material particles are rapidly dissolved. Theoretically, surface dissolution will continue until a *leached layer,* deficient in any leachable fraction, is formed and thereafter the rate of leaching becomes limited by diffusion processes [Baldwin et al., 1997]. Surface effects will theoretically be more significant in the leaching of finely graded materials than coarse-graded materials. This is because the finely graded materials have a substantially higher surface area to volume ratio than the coarse graded materials and also shorter diffusion pathway lengths. This relationship was noted by Bialucha (2000) when she performed a series of tests on materials at different gradings and also by Fällman (1997) and Stegemann and Schneider (1991). Surface leaching processes will also be affected by the physical properties of the material, since connected porosity and permeability will influence the depth of the leached layer.

Advective leaching is driven by a pressure gradient across the leachant which causes it to flow through or over a solid matrix. Leaching by advection processes predominantly result from surface effects and the rate of transportation of leached species away from the solid matrix is rapid. Conner (1990) stated that advection is the dominant leaching and transport mechanism in permeable systems with high flow rates.

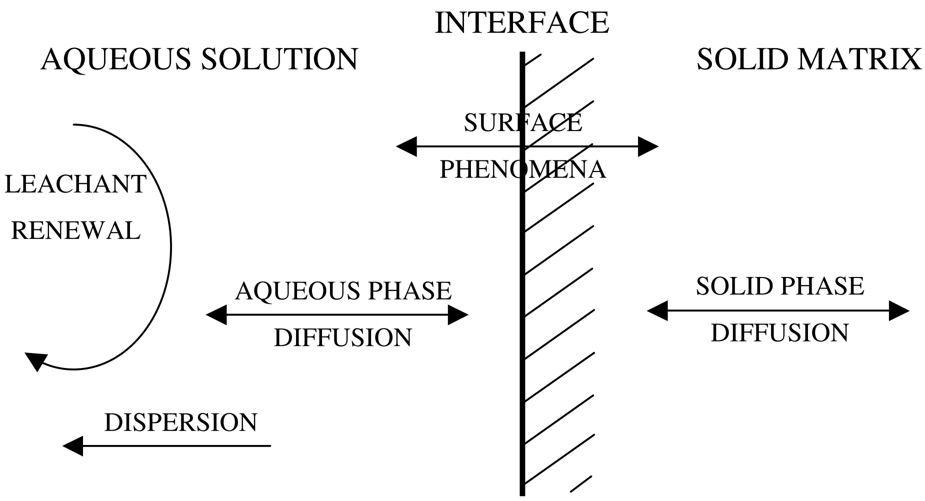


Figure 2-4: Leaching processes [adapted from Cötê et al. from Conner 1990]

Leaching by diffusion occurs at a molecular level by the random motion of individual molecules or ions moving down the concentration gradient to establish equilibrium conditions. Diffusion can occur within the solid phase, the aqueous phase or across the solid-aqueous phase interface, and is generally considered to be a much slower phenomenon than the surface leaching processes described above. Solid phase diffusion can occur as a result of non-homogeneous chemical makeup or leaching of the solid matrix such as that occurring in the leached layer. The availability of further leachable species at the surface of the solid matrix will, in theory, be rate limited by the solid phase diffusion and this process is very slow. Aqueous phase diffusion may occur within the bulk leachant or that contained within the pores of the solid matrix. In general the highest species concentrations will occur in the leachate adjacent to the solid matrix, as a result of leaching. Aqueous phase diffusion will therefore be the dominant mechanism for the transportation of leached species away from the solid matrix in low permeability or quasi-static aqueous systems [Conner, 1990]. Diffusion across the solid-aqueous phase interface occurs when the two phases are not in equilibrium. Under low hydraulic flow conditions the rate of diffusion across the interface is largely dependant on diffusion through the separate solid and aqueous phases. The concentration gradients across the interface will also depend upon the rate of dispersion and/or dilution of the leached species into the aqueous phase. In a high flow scenario, where the leachant is constantly renewed, the concentration gradient across the interface will be maintained and diffusion through the solid matrix is likely to govern the availability of chemical species for leaching.

In the road construction scenario, the processes of greatest importance in the leaching of alternative aggregate materials are likely to be governed by the hydraulic regime occurring within the material in the application under consideration. The hydraulic regime is likely to be a function of the methods of ingress and egress of leachant into the structure (see Section 2.6), the permeability and porosity of the aggregate material, the hydraulic gradient across the structural layer and the pore water chemistry, among others.

2.4.2 Mechanisms Controlling Leaching

Table 2-4 listed many of the factors influencing the leaching of chemical species from solid matrices.

Leaching mechanisms are the properties that control the quantity of a chemical species leached from a solid matrix (mg/kg), the concentration of that chemical species in the leachate (mg/l) and the rate at which leaching occurs. The leaching mechanisms are discussed below in terms of solubility, availability and release controlled leaching.

2.4.2.1 Solubility Controlled Leaching

Solubility can be defined as 'the maximum quantity of one phase dissolved by another under specified conditions' [Sharp, 1990]. In terms of leaching, the solubility of a chemical species will control the maximum concentration (mg/l) of that species that can occur in the leachate. Under solubility controlled conditions, the leached quantity (mg/kg) is proportional to the ratio of the volume of leachant to the mass of solid matrix (liquid to solid ratio, L/S).

The solubility of different chemical species varies considerably and the local leaching conditions can cause changes in solubility over many orders of magnitude. The most commonly referenced of the factors effecting solubility is the pH of the leaching environment.

The pH of a solvent or leachant has a significant influence over the solubility, and hence the leaching, of most metals and some other chemical species. Some typical examples of the pH dependence of leaching are given in Figure 2-5 for calcium, sulphur, silica, barium, molybdenum, cadmium, copper, lead and zinc in cement-stabilized municipal solid waste incinerator (MSWI) fly ash [van der Sloot, 1996]. Many metals, for example lead and zinc, exhibit a marked increase in solubility at both low and high pH values, typically as shown in Figure 2-5. Other chemical constituents may exhibit maximum solubility in the neutral pH range, such as oxyanions of vanadium and molybdenum as that shown in Figure 2-5, or show no dependence on pH, such as sodium and chloride [van der Sloot, 1991; van der Sloot, Heasman and Quevauviller, 1997].

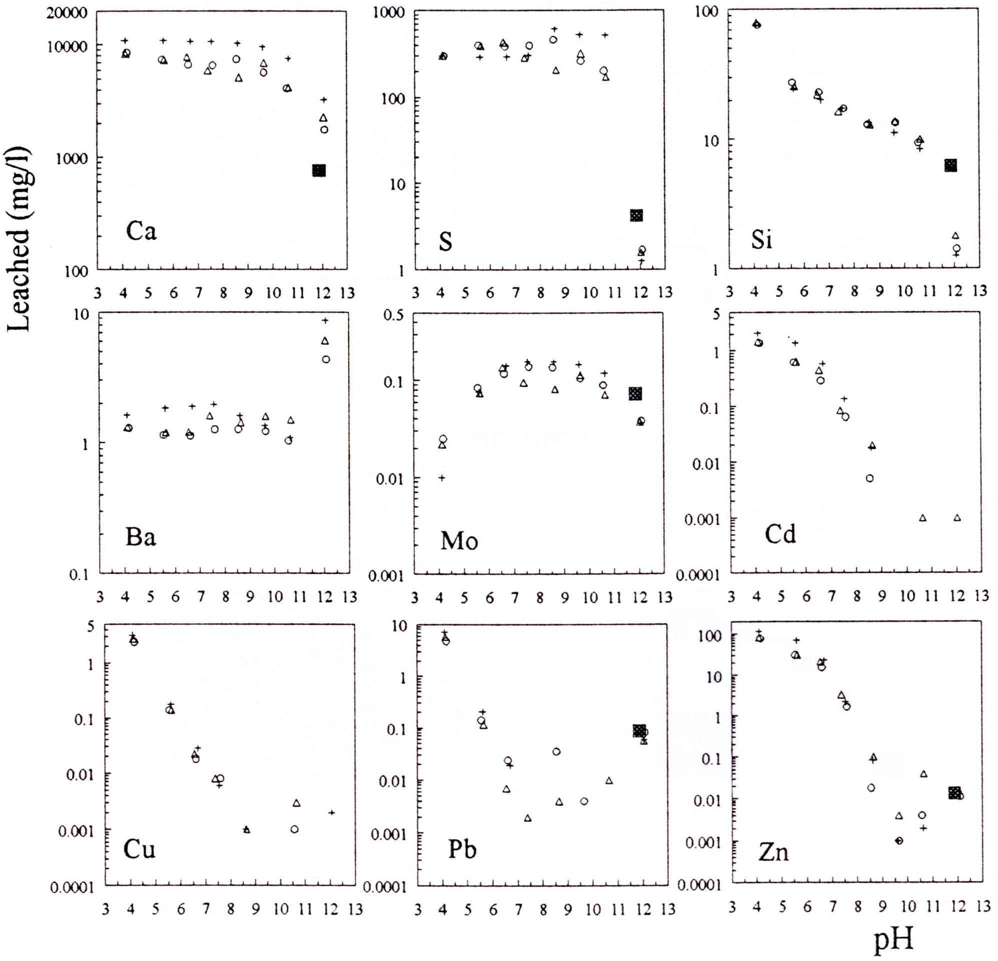


Figure 2-5: pH controlled leaching test data for a number of elements in cement-stabilized municipal solid waste incinerator (MSWI) fly ash [van der Sloot, Heasman and Quevauviller, 1997]

The initial pH of a leachant may differ widely from the pH of the resulting leachate or in-situ pore water, with a concurrent change in solubility. This pH modification may be particularly apparent in systems where the liquid to solid ratio (L/S), or the ratio of mass of dry material being leached to the mass of leachant, is low and the solid phase dominates the system. At high L/S ratios the initial pH of the solution may become more important. According to van der Sloot, Heasman and Quevauviller (1997), the pH in leaching systems such as alternative aggregate materials in road construction is usually governed by the dissolution of the major mineral phases from the solid or by uptake of atmospheric or biologically generated carbon dioxide (CO_2).

The general trends of release of a particular chemical species as a function of pH are similar for different materials. However, absolute concentrations obtained in the leachates often vary over several orders of magnitude between different materials. This variation is shown in Figure 2-6 for the leaching of zinc as a function of pH for 12 different bulk materials [van der Sloot, 1996]. It would therefore be apparent that factors other than the pH are having an effect on the leachability of the chemical species. Many of these variations result from other factors that affect solubility such as redox, temperature, complexation and sorption. Van der Sloot (1996) illustrated the effect of such factors on the leaching of cadmium and zinc, as shown Figure 2-7.

Reduction-oxidation (redox) potential

The redox potential of a system is important in leaching as it can lead to the formation of different chemical phases which can have solubilities that differ by orders of magnitude. The redox conditions can be a function of the solid matrix or the environment of leaching and can be accelerated if micro-organisms act as a catalyst. Biologically-active environments usually have reducing conditions, whereas contact with air or surface waters results in oxidising conditions [van der Sloot, 1991; Alexander and Ford, 1994]. The presence of strong oxidants or reductants can alter the valance state of leached species which in turn can affect speciation, mobility and toxicity [Conner, 1990; Dojlido and Best, 1993]. Under reducing conditions, metal leachability drops significantly, while the leachability of barium, manganese, iron and sometimes arsenic, may increase substantially.

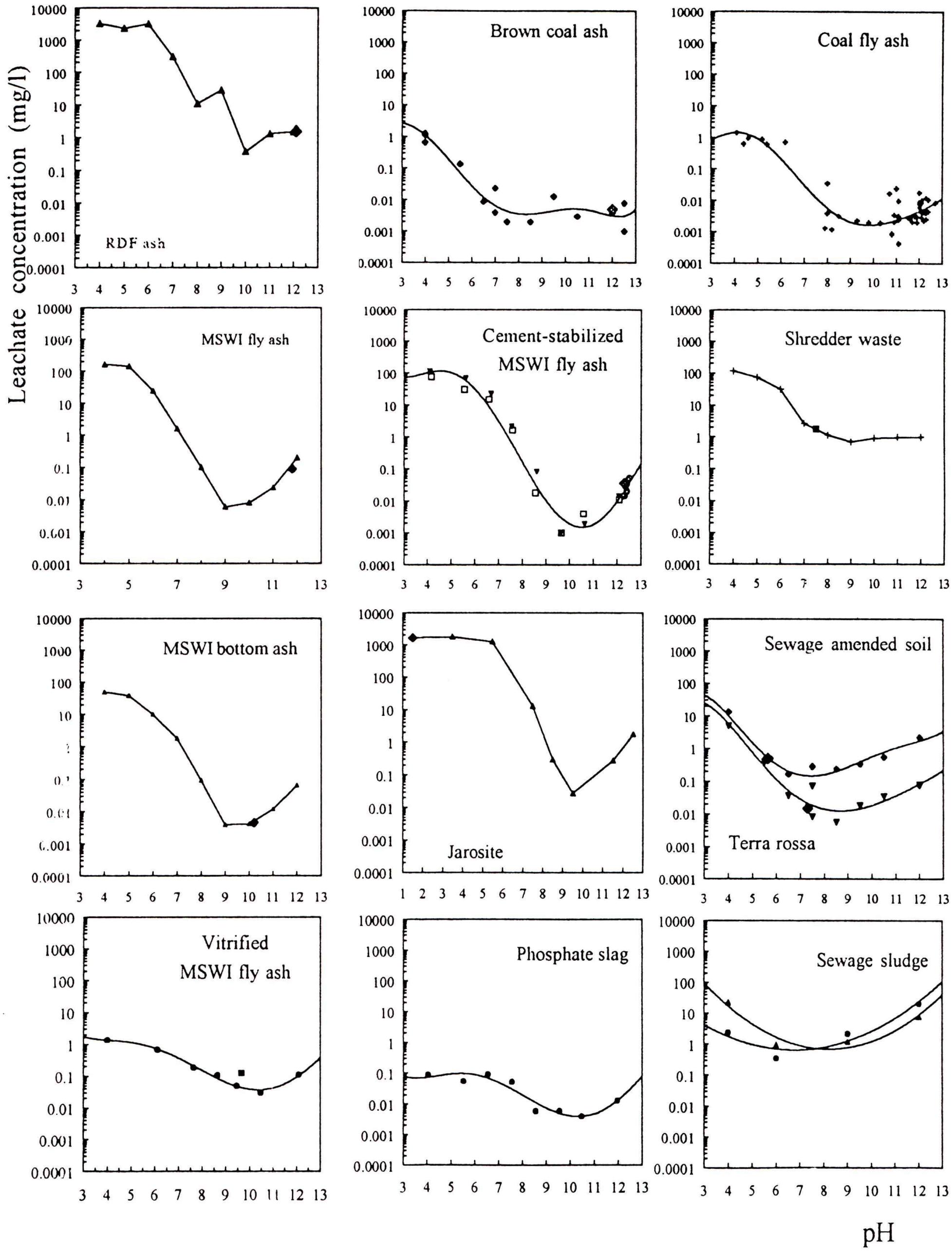


Figure 2-6: Zinc leachability from 12 different bulk materials as a function of pH [van der Sloot, 1996]

<u>*Sorption*</u>

Many solid phase minerals have sorptive properties which can bind dissolved constituents onto the surface of a material via sorption reactions. The bonds that form during sorptive reactions vary in strength and dictate the ease with which the sorbed species may be desorbed again. Both anionic and cationic species can be sorbed depending on the surface charge of the sorption sites, which is a function of pH. Many sorption/desorption processes are strongly dependant on the leachate pH, redox or complexation conditions [van der Sloot, Heasman and Quevauviller, 1997].

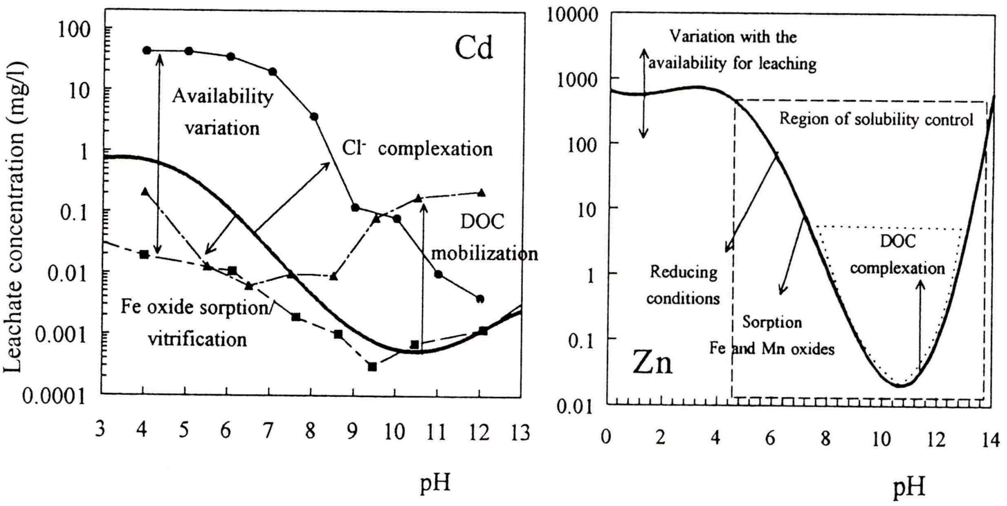


Figure 2-7: Main factors influencing the leaching of cadmium and zinc in specific pH domains [van der Sloot, 1996]

<u>Liquid-solid ratio (L/S)</u>

The ratio of the mass of leachant to the mass of solid matrix being leached exerts significant control over the quantity of a species (mg/kg) that is leached. In a saturated solution the maximum concentration (mg per litre of leachant) of individual chemical species is controlled by the factors considered above. Theoretically, under solubility controlled conditions the greater the L/S the greater quantity (mg/kg) of a species that is leached from the solid matrix.

2.4.2.2 Availability Controlled Leaching

The maximum availability of an individual chemical species can be defined as the quantity (milligrams of chemical species per kilogram of solid matrix, mg/kg) that will leach from a given solid matrix under the most intense leaching conditions. Once the available proportion of the chemical species has been leached, no further release will occur. Availability controlled leaching is most likely to occur in systems with a high L/S ratio, where solubility limits are not reached. For availability controlled leaching the total leached quantity (mg/kg) reaches a maximum and the higher the L/S ratio the lower the resulting concentration (mg/l) of chemical species in solution.

The available quantity of a chemical species may be up to 100% of the total quantity (mg/kg) of that species present in the solid matrix of a given material. The availability is, however, usually significantly less than 100% and just because a particular species is present in a solid matrix does not mean that it will leach even under the most intense leaching conditions [van der Sloot, 1991]. A number of graphs showing availability in comparison to the total matrix composition are shown in Figure 2-8. Availability and the relationship to the quantity of the chemical species within the solid matrix, varies considerably for different chemical species and solid matrices.

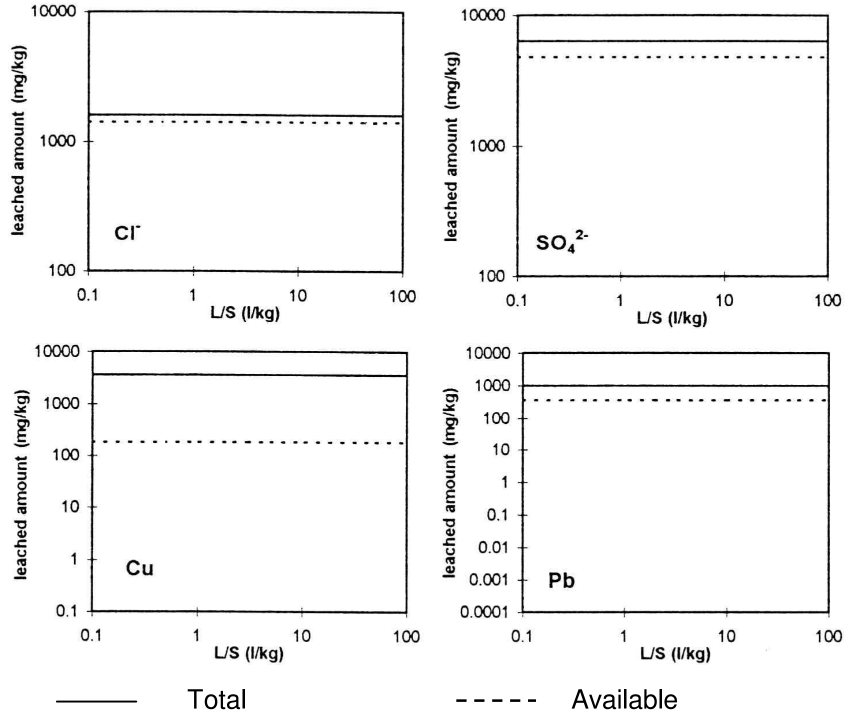


Figure 2-8: Typical graphs of total species composition verses available species [adapted from van der Sloot, Heasman and Quevauviller, 1997]

Total availability can be affected by the physical properties of the solid matrix such as the surface area to volume ratio, porosity and permeability (which could be equated to an effective or available surface area), and properties relating to the physical and chemical bonds within each particle and the mineral phases present.

The chemical form of chemical species in the solid matrix (speciation) and the distribution over different solid phases in the material (fractionation) largely dictates availability for leaching and the potential for leaching. For example, elements tied up in silicate phases or poorly soluble mineral phases are only released after complete destruction or dissolution of the matrix, which is unlikely to occur in environmental systems even in the long term [van der Sloot, 1991].

Liquid-solid ratio (L/S)

Availability controlled leaching is most likely to occur in systems with a high L/S, where the leached species are diluted and solubility limits are not reached. For availability controlled leaching the total leached quantity (mg/kg) is fixed and the higher the L/S the lower the resulting concentration (mg/l) of chemical species in solution.

2.4.2.3 Release Controlled Leaching

Release controlled leaching is leaching that is limited by the rate at which the chemical species can be leached from the solid matrix. Leaching may be rate limited by solid state diffusion or by species transfer across the solid-aqueous phase boundary. The leaching of species with high mobility is most likely to become availability or solubility controlled whereas species with low mobility are most likely to exhibit release controlled leaching. Species mobility varies significantly between different chemical species and between materials and is also dependant on the properties of the solid matrix, such as porosity and permeability, and the leaching environment, such as pH, complexation, redox and temperature.

Matrix Properties

Physical retardation or tortuosity is a relationship defining the path length over which species leached by diffusion controlled processes must travel to leave the boundaries of the material in question and is effectively defining the connected porosity of a material. The tortuosity of a material has a significant effect on the leaching of monolithic materials. Table 2-5 summarises the tortuosity of a number of bound and unbound materials. Lightweight concrete has a relatively high tortuosity despite the high porosity of the material as a result of low connected porosity. Bituminous bound materials have high tortuosity because of the hydrophobic nature of the binder which causes a resistance to wetting [van der Sloot, 1991].

Table 2-5: Physical retardation or tortuosity for a range of materials [van der Sloot, 1991]

Material	**Tortuosity or Physical Retardation Factor**
Unconsolidated granular material	2.5
Stabilised coal fly ash	10 – 30
Stabilised incinerator bottom ash	40
Lightweight concrete	220
Concrete	340
Fly ash concrete	400 – 900
Bituminous concrete	2000 – 10000

2.4.2.4 Relationship between Leaching Mechanisms

The governing mechanisms of leaching may switch between release, availability and solubility controlled depending on the properties of the solid matrix and the specific local environmental conditions. For example a shift in pH may change the solubility from low to high and therefore switch leaching from being solubility controlled to being availability or release controlled. Such processes could equally occur in reverse. Figure 2-9 illustrates the general trends that can be expected to be found in leaching data that is either solubility or availability controlled when plotting leached quantity (mg/kg) and leached concentration (mg/l) against the pH of the leaching environment. The schematic diagrams shown in Figure 2-10 illustrate the relationship between leached quantity (mg/kg) and leached concentration (mg/l) against the L/S of the leaching environment, assuming constant pH.

2.4.3 Effect of Binding on Leaching

Solidification and stabilisation technology has been widely used in hazardous waste management. Many techniques have been developed, although the most important binders / activators at present appear to be:

- Portland cement
- Lime/PFA
- Kiln dust (lime and cement)
- Portland cement/PFA
- Portland cement/lime
- Portland cement/sodium silicate [Conner, 1990].

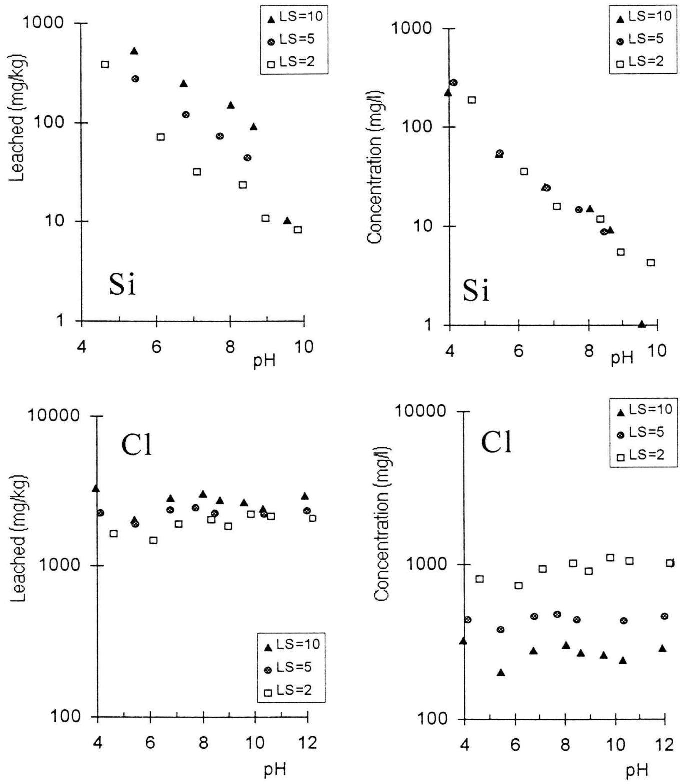


Figure 2-9: Illustration of solubility controlled (silica) verses availability controlled (chloride) leaching [van der Sloot, Heasman and Quevauviller, 1997]

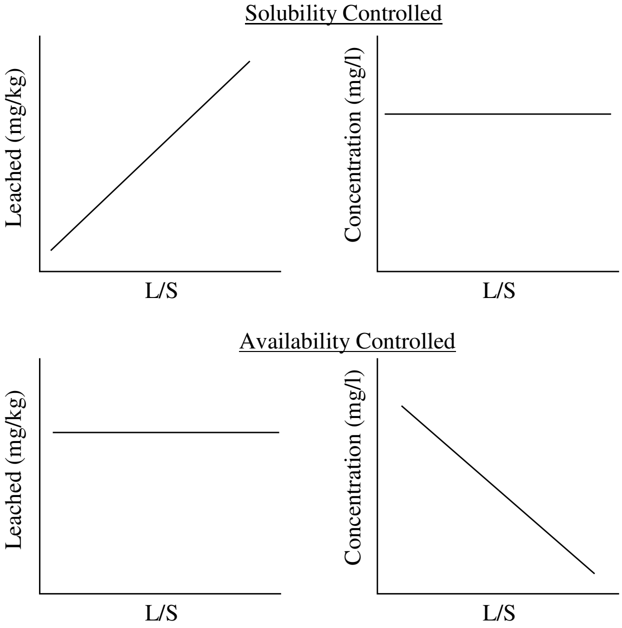


Figure 2-10: Schematic diagram of solubility and availability controlled leaching as a function of L/S (assuming constant pH)

Binders can reduce leaching though both physical and chemical containment mechanisms. Physically, binders can coat the surface of the contaminated material resulting in a reduction in the direct contact between the material and the leachant. If the binder is hydrophobic, such as bitumen, the binder forms a water resistant layer and any contact can be almost eliminated. Binders are often very finely graded and can therefore reduce the permeability and effective porosity of the material.

The mechanisms involved in chemical stabilisation are very complex and interrelated. However, controlling the pH and/or redox potential within a bound matrix are the most commonly utilised containment methods. Cement and pozzolan-based binders rely heavily on pH control [Conner, 1990]. The pH and redox potential of a system should be geared towards controlling the contaminants of concern in a particular material. When considering using binders to control potential contamination it must be remembered that the binders themselves contain a degree of contamination. At present, there is only limited understanding of how binders can control leaching of contaminants from aggregates, in the road utilisation scenario.

2.5 Assessment of Leaching

Leaching is generally assessed by using some form of laboratory leaching test. Leaching tests involve contacting a leachant with a sample of the solid material to produce a leachate or eluate that is then analysed to determine its chemical composition. Numerous tests have been developed by researchers around the world to test various materials for a wide range of utilisation and disposal applications [Environment Canada, 1990]. Many of the test methods originate from the fields of hazardous and radioactive waste management and may not therefore be relevant to apply to the leaching of road construction materials. Most tests can be classified as either batch or flow-through tests and are a variation of the same basic principles with slight modifications in the specific test conditions. Some examples of such tests are listed in Table 2-6.

Table 2-6: Examples of leaching tests from around the world [adapted from van der Sloot, Heasman and Quevauviller, 1997]

<table>
<tr><th colspan="30">LEACHING TESTS FOR GRANULAR MATERIALS</th></tr>
<tr><td colspan="30"><i>SINGLE BATCH LEACHING TESTS (equilibrium based)</i></td></tr>
<tr><td colspan="6">pH Domain 4 - 5</td><td colspan="6">pH 5 - 6</td><td colspan="6">Material Dictated</td><td colspan="6">Complexation</td><td colspan="6">Low L/S</td></tr>
<tr><td colspan="6">TCLP</td><td colspan="6">Swiss TVA</td><td colspan="6">DIN 38414 S4</td><td colspan="6">MBLP (Synth)</td><td colspan="6">MBLP</td></tr>
<tr><td colspan="6">EPtox</td><td colspan="6"></td><td colspan="6">AFNOR X-31-210</td><td colspan="6">(California WET test)</td><td colspan="6">CEN TC 292 compliance test</td></tr>
<tr><td colspan="6">Availability test (NEN 7341)</td><td colspan="6"></td><td colspan="6">Ö-norm S2072</td><td colspan="6"></td><td colspan="6">Wisconsin SLT</td></tr>
<tr><td colspan="6">California WET</td><td colspan="6"></td><td colspan="6">CEN TC 292 compliance test</td><td colspan="6"></td><td colspan="6"></td></tr>
<tr><td colspan="6">Ontario LEP</td><td colspan="6"></td><td colspan="6">Canada EE MCC-3C (Canada)</td><td colspan="6"></td><td colspan="6"></td></tr>
<tr><td colspan="6">Quebec QRsQ</td><td colspan="6"></td><td colspan="6">ASTM D 3987</td><td colspan="6"></td><td colspan="6"></td></tr>
<tr><td colspan="6">Soil HAc</td><td colspan="6"></td><td colspan="6">Soil – $NaNO_3$</td><td colspan="6"></td><td colspan="6"></td></tr>
<tr><td colspan="6"></td><td colspan="6"></td><td colspan="6">Soil – $CaCl_2$</td><td colspan="6"></td><td colspan="6"></td></tr>
<tr><td colspan="30"><i>MULTIPLE BATCH AND PERCOLATION TESTS (mostly based on local equilibrium – LEA)</i></td></tr>
<tr><td colspan="10">Serial Batch (low L/S)</td><td colspan="10">Serial Batch L/S>10</td><td colspan="10">Percolation or Flow Through Tests</td></tr>
<tr><td colspan="10">UHHamburg</td><td colspan="10">NF-X31-210</td><td colspan="10">NEN 7343 (NVN 25008) column up</td></tr>
<tr><td colspan="10">WRU</td><td colspan="10">WRU</td><td colspan="10">ASTM column up</td></tr>
<tr><td colspan="10"></td><td colspan="10">ASTM D4793-88</td><td colspan="10">Column German (pH static)</td></tr>
<tr><td colspan="10"></td><td colspan="10">NEN 7349 (NVN 2508)</td><td colspan="10"></td></tr>
<tr><td colspan="10"></td><td colspan="10">MEP method 1320</td><td colspan="10"></td></tr>
<tr><td colspan="10"></td><td colspan="10">Sweden ENA</td><td colspan="10"></td></tr>
<tr><td colspan="10"></td><td colspan="10">MWEP</td><td colspan="10"></td></tr>
<tr><td colspan="15"><i>STATIC METHODS</i></td><td colspan="15"><i>SPECIATION METHODS</i></td></tr>
<tr><td colspan="15">mcc-1</td><td colspan="15">Sequential chemical extraction</td></tr>
<tr><td colspan="15">mcc-2</td><td colspan="15">pH static test procedures</td></tr>
<tr><td colspan="15">Compacted granular tank leaching test (Rutgers/ECN)</td><td colspan="15"></td></tr>
<tr><th colspan="30">LEACHING TESTS FOR MONOLITHIC MATERIALS</th></tr>
<tr><td colspan="30"><i>DYNAMIC LEACHING TESTS</i></td></tr>
<tr><td colspan="30">ANS 16.1</td></tr>
<tr><td colspan="30">Tank leaching test NEN 7345</td></tr>
<tr><td colspan="30">Spray test (impregnated wood)</td></tr>
<tr><td colspan="30">Swedish MULP</td></tr>
</table>

2.5.1 European Leaching Tests

The European Committee for Standardisation (CEN) has many technical committees (TC) involved in developing leaching test standards for various applications. These technical committees include:

- CEN TC 38 Durability of Wood;
- CEN TC 51 Concrete & Building Limes;

- CEN TC 104 Concrete;
- CEN TC 154 Aggregates;
- CEN TC 164 Water Supply;
- CEN TC 292 Characterisation of Wastes;
- CEN TC 308 Characterisation of Sludges.

The leaching tests discussed here are those under development by CEN TC 154 and 292.

On 5 March 2002 the leaching test for aggregates developed by CEN TC 154 was approved by CEN. The same test became a British Standard on 9 August 2002. The test is entitled BS EN 1744-3 'Tests for chemical properties of aggregates – Part 3: Preparation of eluates by leaching of aggregates' [BS EN 1744-3, 2002]. The test consists of a single batch tank-leaching test over a duration of 24 hours. The L/S ratio is 10 by mass with a material sample mass of 2kg and particle size of less than 32mm. The leachant is distilled deionised water that is agitated using a dip stirrer. The pH of the leachate is controlled by the material sample.

The BS EN1744-3 test is designed to test unbound aggregates for the purpose of compliance testing. It is therefore a test to be used regularly to confirm that the batch of material is typical of that material as a whole and that it is suitable for the proposed use. Although the test is now a certified standard there is no guidance to accompany it with regards to interpretation of the data. This limits the current application and usability of this standard.

CEN TC 292 Characterisation of Wastes committee has been developing leaching tests for many years. The CEN TC 292 approach has been to develop tests within three categories depending upon the purpose. These categories are:

1. *Characterisation / leaching behaviour tests* – used to obtain information on the short and long term leaching behaviour and characteristic properties of materials;
2. *Compliance tests* – used to determine whether the material complies with specific reference values and is similar to the leaching behaviour identified by the characterisation tests;

3. *On-site verification tests* – used as a rapid check to confirm that the material is the same as that which was subject to the compliance test [BS EN 12457, 2002].

The CEN TC 292 compliance tests became a British and European Standard in 2002 (BS EN 12457 Leaching: Compliance test for leaching of granular waste materials and sludges: Part 1, 2, 3 and 4). The tests are one or two batch tests with a duration of 6 or 24 hours. The L/S ratio is 2 or 10 l/kg by mass for the single batch tests or 2 and 8 for the two batch test. The material sample has a mass of 175g and a particle size less than either 4 or 10mm. The leachant is distilled deionised water and the leaching vessel is agitated. The pH of the leachate is controlled by the material sample [BS EN 12457, 2002]. The test is generally for use as a compliance test, although, the two batch test does enable limited identification of the leaching mechanisms in operation. A compliance leaching test for monolithic materials is also under development by CEN TC 292.

CEN TC 292 are developing a suite of characterisation or leaching behaviour tests that are designed to address different aspects of leaching behaviour. The tests cover availability, pH static, acid neutralisation capacity, percolation, diffusion and in-situ simulation tests [CEN website]. Table 2-7 lists these tests with their working titles, project reference numbers, current status and type of test.

Table 2-7: Summary of leaching behaviour tests under development by CEN TC 292 [CEN website]

Test Title	**Project Reference**	**Current Status**	**Type of Test**
Digestion for subsequent determination of aqua regia soluble portion of elements in waste	PrEN 13657	Under approval	Availability
Influence of pH on leaching with initial acid/base addition	PrEN 14429	Under approval	Acid neutralisation capacity
Influence of pH on leaching with continuous pH control	-	Under development	pH static at a range of pH values
Up-flow percolation test	PrEN 14405	Under approval	Column test – accelerated leaching
Simulation of the leaching behaviour of a waste material under specific conditions – Down-flow percolation test	-	Under development	In-situ simulation
Dynamic leaching test for monolithic waste	-	Under development	Diffusion test

2.5.2 Leaching Test Conditions

There are many different types of leaching test and the test conditions tend to vary between each. Some of the most common variables are discussed below [Environment Canada, 1990; van der Sloot, Heasman and Quevauviller, 1997].

2.5.2.1 Sample Pre-treatment/Preparation

Depending on the type of test and the material to be tested pre-treatment may include liquid/solid separation, drying, subsampling, particle size reduction, surface washing, compaction, preservation, curing and aging.

2.5.2.2 Leachant Composition

Commonly used leachants include distilled-deionised water, site leachate (real or synthetic) or a chemical solution, such as pH modified water. During the leaching test certain properties of the leachant, such as pH, can either be artificially controlled or be controlled by the leaching of chemical species from the solid material. Where the leachant properties are controlled by the latter case and the test is performed at low to moderate L/S, the pH and composition of the leachate will largely be controlled by leaching of the solid phase and will be relatively independent of the initial composition of the leachant. At higher L/S, or where the properties of the leachant are artificially adjusted during the test, the leaching conditions may be more influenced by the leachant.

2.5.2.3 Mode and Method of Waste-Leachant Contact

Conditions that may affect the transfer of chemical species from the solid to aqueous phase include agitation (for example by stirring the leachant or moving the leaching vessel), the degree of saturation and exchange between other components of the system, such as the leaching vessel and the atmosphere.

2.5.2.4 Liquid-solid Ratio (L/S)

The L/S ratio is usually chosen depending on the objectives of the leaching test, the solubility of the chemical species of interest and the analytical detection limits. The ratio should not be so large as to dilute the concentration of the chemical constituents below their analytical detection limit, however, they should be high enough to prevent solubility constraints from dominating the leaching process. In practical terms, the L/S must also be large enough to enable a sufficient quantity of leachate to be separated from the solid matrix for analytical purposes. In practice, L/S commonly varies between 2:1 and 100:1.

2.5.2.5 Contact Time

The amount of time during which the liquid phase is in contact with the solid phase will influence the quantity of chemical species leached until steady-state conditions are achieved. In batch tests, the contact time is equal to the test duration, whereas in dynamic tests, such as column flow-through tests, the contact time is a function of the flow rate of the leachant. Larger particle sizes will generally require a longer contact time to establish steady-state conditions.

2.5.2.6 Temperature

The properties or mechanisms relevant to leaching vary exponentially with temperature. For convenience, most leaching tests are carried out at room temperature. Higher temperatures can be used to accelerate the rate of leaching, although this should be treated with caution as it may also change the properties of the material being leached and promote biological activity.

2.5.2.7 Separation of Leachate from the Solid Phase

Leachates are usually separated from the solid material by filtration through a 0.45µm filter membrane, which is a convention used to define soluble species. A filter medium with low species adsorption should always be used.

2.6 Leaching in the Road Construction Environment

2.6.1 Water Movement in the Road Pavement

Roads are designed to exclude or inhibit the infiltration of water into the pavement and the underlying subgrade. The primary reason for this exclusion is that the presence of water increases pore pressures and reduces the effective stress of the layer resulting in a material with low stiffness and resistance to deformation [Brown, 1985].

Water also provides the principle route for the movement of the chemical constituents of the construction materials into the adjacent environment. Restricting movement of water into and through the road construction will inhibit this pathway.

Inevitably, some water will enter the pavement construction. Konrad and Roy (1998) and Gschwendt and Staño (1998), among others, have monitored in-situ pavement moisture content and have identified seasonal variations that indicate water must enter and leave the pavement. To prevent saturation of the pavement and subgrade, drainage systems are usually included in the design. Numerous designs of highway and pavement drainage systems exist [e.g. Hoppe, 1998; Luker and Montague, 1994], the choice of which depends upon the road design and the geographic and environmental location [Baldwin et al., 1997]. Many sub-base drainage systems rely on the sub-base materials having adequate permeability (suggested by McEnroe (1994) and Jones and Jones (1989a) to be greater than approximately 10^{-4} to 10^{-3}m/s) and being constructed with a cross-fall (usually between 2.5% and 7.5%).

A typical layered road construction has many routes for the movement of water into, through and out of the structure. These routes vary throughout the construction, service life and reconstruction (or maintenance) of the road. Routes for water ingress and egress were reviewed by, Roy (1981), Dawson (1985) and Baldwin et al. (1997) among others. Figure 2-11 illustrates the routes and mechanisms identified.

It can be seen from Figure 2-11 that water can enter or leave a pavement from all physical directions: through the pavement surface, from the subgrade below and from the road margins. Not all mechanisms would be present in every situation and some are mutually exclusive, e.g. it is not possible to drain water into the subgrade by soakaway action when water is entering the pavement as a result of an artesian head.

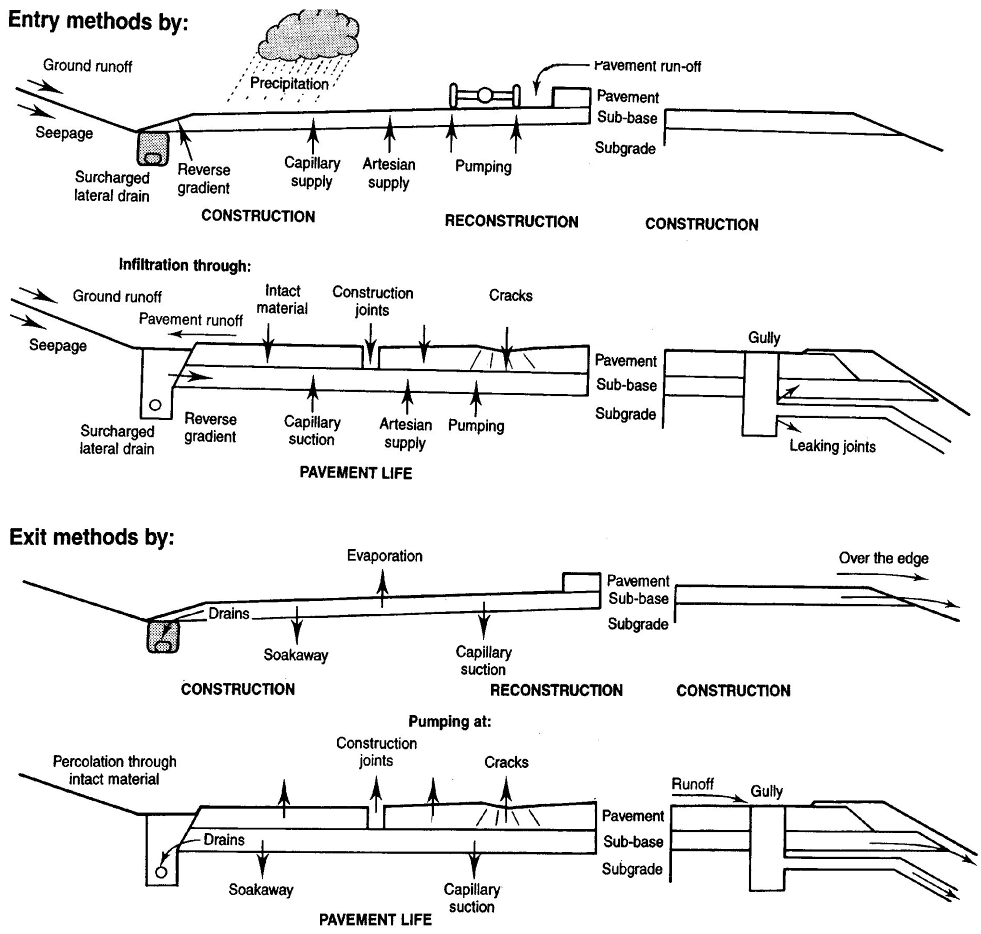


Figure 2-11: Methods by which water may enter or leave a road construction during construction, life and reconstruction [Baldwin et al., 1997]

2.6.1.1 Routes of Water Ingress

Baldwin et al. (1997) suggested that during construction and reconstruction exposed subgrade and sub-base materials can receive water via runoff from the adjacent pavement (which, by design, has a cross-fall to guide water to the drain at the edge of the pavement), as well as directly from precipitation. Once surfaced, with bitumen or cement bound material, the infiltration of precipitation is almost completely eliminated, as the permeability of such surfacing materials is usually very low. Ridgeway (1976) studied infiltration through Portland cement concrete and dense-graded bituminous concrete road surfaces. The study showed that, when intact, the materials were virtually impermeable. In contrast, Raimbault (1999) claimed that it might be possible for 8.6mm of precipitation per day to permeate a typical bituminous surfacing. Raimbault's figures are however based purely on the permeability of the surfacing material (taken as 10^{-7}m/s) which in practice may be significantly inhibited by the sporadic pattern of rainfall and the relatively rapid rate of highway surface runoff. Quibel (1998) used an empirical drainage model to compute flow within a pavement using realistic meteorological data for different regions in France. The model predicted that for a surface permeability of 10^{-8}m/s infiltration into the road would be very low. However, when the surface permeability was $5x10^{-7}$m/s, the road base was greater then 90% saturated for between about 200 and 300 days in the year depending on climate. This model was simplistic but it does highlight the importance of pavement surface permeability in controlling ingress to the structure. Similarly, work by Van Ganse (reported by Raimbault, 1999) determined the proportion of precipitation that would infiltrate through road surfaces of various permeabilities. The results are shown in Table 2-8 and are generally in agreement with Quibel (1998) and Raimbault (1999).

Table 2-8: Proportion of precipitation to infiltrate through pavement surfaces with various permeability [from Van Ganse, reported in Raimbault, 1999]

Permeability, k (m/s)	10^{-9}	10^{-8}	10^{-7}	10^{-5}
Infiltration/Precipitation, I/P (%)	0	5	50	~100

A low permeability road surface often contains construction joints and surface cracks that result from material shrinkage and distress due to loading as the pavement ages, which may allow a more significant passage of water. The flow rate through cracks or joints depends on the water carrying capacity, length of and area that drains to each crack and the duration and, to a lesser extent, the intensity of the rainfall. Ridgeway (1976) estimated an infiltration rate of 100cm^3/hour/cm of crack. If the degree of cracking can be quantified from field observations the rate of water inflow can be estimated. Baldwin et al. (1997) estimated that Ridgeway's figures could be reworked as 2 litres/hour/m^2 of pavement that has just reached the maintenance intervention level. To calculate inflow, Jones and Jones (1989b) developed the following equation:

$$q = \frac{2.8 \times 10^{-6}}{w} \left(N + 1 + \frac{W_c}{C_s}\right) + k_p$$

where,

q is the inflow rate (m^3/sec/m^2)

2.8×10^{-6} is the average infiltration rate into cracks (m^3/sec/m)

w is the carriageway width (m)
N is the number of lanes
W_c is the total length of transverse cracks (m)
C_s is the transverse joint spacing or average distance between cracks (m)
and k_p is the infiltration rate ($m^3/sec/m^2 = m/s$) through intact surfacing,

and for a 'mean' pavement condition estimated the inflow rate at 6 litres/hour/m^2 of pavement.

Water may also enter the structure from the subgrade via artesian flow, capillary action of the lower pavement layers or by pumping action under traffic loading. All three mechanisms may occur during the construction or service life of the pavement although Dawson and Hill (1998) concluded that significant contributions from such sources were likely to occur in only a few cases. If artesian flow does occur, it is likely to be seasonal and occur if the groundwater table rises significantly during wetter months. Although sub-base materials may be designed to be free-draining, some compacted granular materials exhibit a capillarity that enables them to draw in water, vertically against gravity, as a result of the surface tension properties of water. Material capillarity is a function of particle size and shape and the degree of compaction [Biczysho, 1985] with small particles and void spaces creating the largest capillary fringe.

Finally, water may enter the pavement from the road margins. The most significant route is likely to be precipitation falling on unsurfaced road margins, seeping into the ground and then being drawn into the road by capillary action (although, typically, drainage would be designed to intercept such water). In super-elevated pavements (or ones improperly designed) reverse gradients on permeable layers above formation level may result in water flowing back into the pavement instead of into the drains. Poorly designed or maintained drains can potentially increase the amount of water in the road and if surcharging of the drainage system occurs or any of the pipes or gullies are leaking, then water is available to enter the pavement structure and the mechanisms designed for removing this water are not functioning.

2.6.1.2 Routes of Water Egress

There are fewer distinguishable routes for water to leave the pavement, many of which are the ingress mechanisms acting in reverse. The routes of egress from the pavement are of particular importance for understanding contaminant migration into the wider environment. Potential routes of egress through the pavement surface include evaporation through the intact pavement surface or cracks or by traffic loading pumping water through cracks and joints. These methods are unlikely to contribute significantly to the removal of moisture. Water is most likely to leave the pavement to the subgrade or road margins. On permeable subgrades (such as sandstone, chalk and many limestones) with low water tables, water in the pavement is most likely to soakaway into the subgrade under gravity or with the assistance of the capillary action (or greater suction) of the subgrade. However, if water moving vertically through the pavement reaches a layer of lower permeability material, then an alternative preferential flow path will be sought. The designed cross-fall of the road structure and the anisotropic (greater horizontal than vertical) permeability of compacted aggregates [Head, 1981; Abbott, 1990] will result in the water flowing under gravity across the road to lateral or median drains and providing a significant degree of drainage [Gerke (1987); Baldwin et al. (1997); Dawson and Hill (1998)].

2.6.2 Pavement Water Quality

Water that enters a pavement structure will typically be sourced from either groundwater or precipitation and contain a variety of dissolved chemical constituents [Freeze and Cherry, 1979]. Groundwater often contains cations such as calcium, magnesium, sodium and potassium and the most common anions are chloride, sulphate, carbonate and bicarbonate. Silica may also be a major constituent whilst minor constituents may include iron, manganese, fluoride, nitrate, strontium and boron [Fetter, 1994]. Precipitation also contains many particulate and gaseous substances that are adsorbed as it falls through the atmosphere. Road traffic, maintenance and accidental discharges can contaminate the road surface and be mobilised by precipitation. Luker and Montague (1994) listed typical pollutants found in highway drainage discharges that are summarised in Table 2-9.

Table 2-9: Sources and classifications of pollution found in highway drainage [Luker and Montague, 1994]

	Sources			
Classification	***Traffic***	***Maintenance***	***Accidents & Disposal***	***Other***
Sediment	Carbon Rubber Organic solids Litter Grit Asbestos	De-icing grit Rust Resurfacing grit Metal filings Plastic Organic solids	Soil Cement Sand Gravel Plastic Litter	Atmospheric dust Organic detritus Soil
Metal	Lead Zinc Nickel & chromium Iron & rust Cadmium Copper Vanadium	Iron Copper Chromium Nickel Cadmium Arsenic	Lead Zinc Copper Chromium Iron	Iron Manganese
Hydrocarbon	Petrol Oil PAHs & MTBE Hydraulic fluid Grease Antifreeze Olefins	Tar & bitumen Asphalt PAHs Oils Grease Solvents	Petrol Solvents Oil Grease	
Salt & Nutrient	Nitrates Bromide Sulphates Ammonia	Chlorides Sulphates Phosphates Urea Bromide Cyanide	Fertiliser Organo-compounds	Acid rain
Microbial	Livestock Excrement	Organic solids	Bacteria Viruses Organic solids Excrement	Bacteria Viruses Flesh, blood & bone
Other		PCBs Herbicides Pesticides	Herbicides Pesticides	

2.7 Legislation and Regulations

The legislation and regulations of relevance to the leaching of alternative aggregate materials in road construction and bulk fill applications can principally be drawn from the fields of waste management and water quality. UK legislation has largely originated from the need to implement the various European Directives.

2.7.1 Waste Management Legislation and Regulation

2.7.1.1 The Definition of Waste

In 1975 the European Waste Framework Directive 75/442/EEC (1975) set a minimal framework for waste management and resource use. The definition of waste given in this Directive is that:

- *''Waste' shall mean any substance or object in the categories set out in Annex 1* (of the Directive) *which the holder discards or intends or is required to discard.*
- *'Holder' shall mean the producer of the waste or the natural or legal person who is in possession of it;*
- *'Producer' shall mean anyone whose activities produce waste and/or anyone who carries out pre-processing, mixing or other operations resulting in a change in the nature or composition of this waste'.*

The Waste Framework Directive [75/442/EEC, 1975] also introduced a list a materials that should be classified as waste.

The Environmental Protection Act 1990 partly transposed the European Directive into national legislation, and defined waste as:

- *'Any substance which constitutes a scrap material or an effluent or other unwanted surplus substance arising from the application of any process" or,*
- *"Any substance or article which requires to be disposed of as being broken, worn out, contaminated or otherwise spoiled'.*

The Waste Management Licensing Regulations (1994) revised the definition given in the Environmental Protection Act by stating that any reference to waste *'shall include a reference to Directive Waste'* referring to the Waste Framework Directive.

The list of wastes given in the Waste Framework Directive [75/442/EEC, 1975] has since been changed and expanded a number of times and is now presented in the European Waste Catalogue [2000/532/EC, 2000].

The definition of waste is an issue that has become legally contentious. Case law has focused on the definition of the term 'discard'. The outcome of the case law means that discarded materials only cease to be waste when they have been fully recovered and put to final use. It does not cease to be waste just because someone intends to use it, it has an economic value, it has been processed ready for recycling or it is not polluting. This creates a real hindrance for the use of alternative aggregates in road construction because of the additional requirements and costs associated with handling, transporting and processing a material that is classed as a waste. This also generates problems in the actual utilisation of the material where the use of a waste in such applications would require a waste management license and the associated permissions, assessments, monitoring and after use issues that would severely hinder progress to utilising alternative aggregates. The point at which one person's waste becomes another person's resource is often not easy to define and the interpretation can be inconsistent. This is an issue that needs further addressing in order to encourage the use of alternative aggregates in road construction and bulk fill.

2.7.1.2 The Landfill Tax

In October 1996 the UK government introduced a tax on all waste going to landfill. The 'Landfill Tax' was initially set at two rates: A lower rate of £2 per tonne for inactive (or inert) wastes and a standard rate of £7 per tonne for all other taxable wastes [HMCE, 2000]. The standard rate was set to increase at increments and in 2004 is at a level of £15 per tonne. The Landfill Tax has increased the cost of disposing of waste materials, and thus provides an economic driver for the increased reuse and recycling of industrial wastes and by-product materials as alternative aggregates.

2.7.1.3 The Landfill Directive

Council Directive 1999/31/EC on the landfill of waste, 'The Landfill Directive' [99/31/EC, 1999] is implemented in England and Wales through the Landfill (England and Wales) Regulations 2002 [SI 2002/1559, 2002] and minor modifications to the Pollution Prevention and Control (PPC) Regulations 2000. The requirements of the landfill directive that impact on the use of alternative aggregates in road construction include the following:

- The reclassification of landfill sites into those accepting inert, non-hazardous and hazardous wastes and the end of 'co-disposal' of non-hazardous and hazardous wastes;
- The requirements for the pre-treatment of wastes;

- The revised definitions of inert, non-hazardous and hazardous wastes that make some previously inert waste into non-hazardous wastes and some previously non-hazardous wastes into hazardous wastes;
- The banning of whole and shredded tyres from landfill (except when used in engineering applications).

The stricter requirements on the landfill of wastes are likely to increase the costs of waste disposal to landfill and thus provide an incentive to minimise waste and increase the reuse and recycling of materials, including alternative aggregate materials.

2.7.2 The Aggregates Levy

The Aggregates Levy was introduced in April 2002 at a rate of £1.60 per tonne of aggregate produced. This levy was designed to reflect the true social and environmental costs associated with material extraction and is a tax on the commercial exploitation in the UK of rock, sand and gravel. The resultant increase in the price of primary aggregates may encourage the use of recycled or alternative sources of aggregate materials although there are concerns because not all sources of alternative aggregate materials are exempt from the levy.

2.7.3 Water Quality Legislation and Regulation

European Directive 76/464/EEC on ‘Pollution Caused by Certain Substances Discharged into the Aquatic Environment of the Community’ established a framework for the elimination or reduction of pollution of inland, coastal and territorial waters by dangerous substances. This Directive introduced two lists of potentially polluting substances: List I or ‘black list’ substances that are considered to be the most harmful and are generally characterised as being toxic and/or persistent and/or bioaccumulative; and, List II or ‘grey list’ substances are considered less harmful.

The Water Framework Directive [2000/60/EC, 2000] includes new requirements for protecting and restoring surface water, groundwater and dependent ecological systems. The Water Framework Directive sets out requirements for the integrated management of surface and groundwater, both in terms of water quality and resources. This Directive requires the protection of water quality and restoration of the status of water bodies where they fail to achieve ‘good’ status. More specifically the directive requires that direct discharges of pollutants are prevented from entering groundwater, and that controls are placed on activities to prevent and limit (indirect) entry of pollutants to groundwater.

2.7.3.1 Protection of Groundwater

EC Directive 80/68/EEC on the 'Protection of Groundwater against Pollution Caused by Certain Dangerous Substances' has been implemented in the UK through the Groundwater Regulations 1998. The Groundwater Directive [80/68/EEC, 1980] requires the management of activities so as to prevent groundwater pollution and the general purpose of the Groundwater Directive is:

'To prevent the pollution of groundwater by the substances listed in the annex to the directive. More specifically, Member States are obliged to take necessary steps to:

a) *Prevent substances in List I from entering the groundwater, and*
b) *Limit the introduction of List II substances into groundwater so as to avoid pollution.'*

In December 1992, the National Rivers Authority, the environmental regulator in England and Wales at this time, published its 'Policy and Practice for the Protection of Groundwater' [NRA, 1992]. The policy resulted from the duty to monitor and protect the quality and quantity of the groundwater and includes the principles and implementation of resource and source protection, which relies on vulnerability and protection zone maps. This guidance has since been superseded by the Environment Agency, the current environmental regulator in England and Wales, with guidance incorporating groundwater source protection zones.

2.7.3.2 Protection of Surface Water

The protection of the quality of surface waters is mainly based around concentrations of substances that are acceptable to aquatic species. Environmental Quality Standards (EQS) for freshwater are set depending on the hardness of the water.

2.7.3.3 Quality of Water for Human Consumption

A number of EC Directives have been developed in relation to standards for drinking water, including:

- Directive 75/440/EEC on the quality required of surface water intended for the abstraction of drinking water, and

- Directive 80/778/EEC on the quality of water intended for human consumption.

Directive 80/778/EEC sets standards for the quality of drinking water sources both directly and after treatment and includes Maximum Admissible Concentrations and Guide Levels for 62 determinants and Minimum Required Concentrations for four determinants. The World Health Organisation also sets guideline values for drinking water quality [WHO, 1984].

In the UK drinking water quality standards are defined in:

- Water Supply (Water Quality) Regulations 1989 (SI 1989/1147) (as amended), and
- Water Supply (Water Quality) Regulations 2000 (SI 2000/3184) (as amended).

3 Material Selection and Characterisation

3.1 Introduction

A large number of alternative materials have potential for use in road construction and bulk fill applications. This chapter describes the selection and sampling of a range of materials, binders and binder activators for examination in the research. The selected materials are then characterised by determining the chemical composition of the sold fraction of the materials and by carrying out a simple test for the determination of leachability. The results of the material characterisation are discussed and chemical constituents of potential concern are identified.

It must be noted here that this research was not aimed at approving, for use in road construction applications, any of the materials that were selected for testing. Instead, the materials were used as a tool for investigating the effect of the in-situ material condition on leaching and thereby determining a suitable testing regime for the assessment of any candidate material.

3.2 Material Selection, Sampling and Storage

Numerous materials have the potential to be used as aggregate materials and/or binders in road construction and bulk fill. The selection of suitable candidate materials for investigation within this thesis was based, principally, on the following factors:

a) Recommendations from the projects' industrial collaborators and sponsors [See acknowledgements];
b) Mechanically proven materials and mixtures or reported successful usage, predominantly as determined by Nunes (1997) and recommended by Sherwood (1995) and Baldwin et al. (1997);
c) Significant consumer potential, including the magnitude and location of resources in the UK [Sherwood, 1995; British Geological Survey, 2000];
d) Availability of data on chemical composition and leaching under different testing regimes, and in particular following on from the work of Baldwin et al. (1997);
e) The physical and chemical properties of the materials, including:
    - Particle size (75mm to <75µm) and grading (single size, continuous or gap graded);

- Chemical constituents and leaching potential;
- Material source type (processed natural materials, recycled materials and by-products from high temperature processes).

Using the selection procedure described above, 20 materials were chosen for investigation, of which 14 were principally aggregate materials and 6 were binders or binder activators. The aggregates and binders are listed in Table 3-1 and Table 3-2 respectively, alongside details of the material supplier and a brief description of the material.

Table 3-1: Aggregate materials selected for investigation

Material	Code	Supplier	Particle Size /Grading	Source
Asphalt planings	AP	Nottingham City Council	Coarse-graded	Aggregate with bituminous binder from road resurface
Air-cooled blast furnace slag	BFS	British Steel / East Coast Slag Products	Coarse (DoT Type1)	Slag from high temperature (1300-1600°C) iron ore processing
Basic oxygen steel slag	BOS	British Steel/ East Coast Slag Products	Coarse (DoT Type1)	Slag from high temperature steel making
China clay sand	CCS	CAMAS/ Bardon Aggregates	Medium	Washed, weathered granite by-product of china clay extraction
Crumbed rubber	CR	Rushden Granulating Company	Medium	Crumbed rubber from vehicle tyres
Foundry sand (alkaline phenolic binder, brass casting)	FSB	Castings Development Centre	Fine	Alkaline phenolic bound sand from a brass foundry
Furnace bottom ash	FBA	PowerGen	Medium	Bottom ash from coal-fired power station
Granite	GR	Tarmac Quarry Products	Coarse (DoT Type1)	Primary aggregate
Limestone	LS	Tarmac Quarry Products	Coarse (DoT Type1)	Primary aggregate
Minestone	MS	R.J.B. Mining	Coarse	Spoil from coal mining
MSW incinerator ash (New incinerator)	MSW(N)	Ballast Phoenix	Coarse	Bottom ash from municipal waste incineration
MSW incinerator ash (Old incinerator)	MSW(O)	Ballast Phoenix	Coarse	Bottom ash and fly ash from municipal waste incineration
Pulverised fuel ash (Conditioned)	PFA	PowerGen	Fine	Fly ash from coal-fired power station. Pozzolanic material
Sewage sludge incinerator ash	SSA	Ballast Phoenix	Fine	Bottom ash from sewage sludge incineration

Table 3-2: Binders and activators selected for investigation

Material	Code	Supplier	Particle Size /Grading	Source
Bitumen	bi	Shell Bitumen	Viscous fluid	Petroleum Product
Cement	ce	Blue Circle Cement	Fine	Ordinary Portland Cement 1450°C manufacture Hydraulic binder / activator
Cement kiln dust	ckd	Blue Circle Cement	Fine	Fly ash from cement kiln 1450°C process Hydraulic binder / activator
Flue-gas desulphurisation gypsum	fgd	PowerGen	Fine	Air pollution control residue for coal-fired power station Activator for pozzolanic binder
Granulated blast furnace slag	gbfs	British Steel/ East Coast Slag Products	Medium	Slag from iron ore processing (1300-1600°C) - cooled by water quenching Hydraulic binder
Lime	li	Buxton Lime Industries	Fine	Manufactured from limestone at 1150-1300°C Activator for pozzolanic binder

To enable a comparison to be made between the leaching of alternative materials and that from widely used and accepted sources, conventional granite and limestone aggregates were included in the selection. The selected binders included both conventional and alternative sources. All have been shown to perform as binders either solely or in conjunction with other binders, materials or activators [Nunes, 1997]. A number of the materials (including pulverized-fuel ash and blastfurnace slag) may be classified as both an aggregate and a binder. These materials exhibit pozzolanic or hydraulic binding properties that can aid material stabilisation.

For each of the materials listed in Table 3-1 and Table 3-2 a representative sample of the commercially available product was sourced. The products generally fall into one of three categories:

1. Material is available as a commercial product that is sold in pre-packaged units;
2. Material is available as a commercial product that is sold from a regulated/managed stockpile;
3. The material is not currently marketed as a commercial product or has an unpredictable or non site-specific source.

Table 3-3 provides a summary of the market source of each material sample. Commercially available material products were used wherever available since the raw material will generally have undergone some processing that may affect the leaching properties of the material. For example raw blastfurnace slag is weathered outside for at least one year, before being crushed, screened and mixed to the required grading of the commercially available product. For the materials that are currently not available as a product, the material sample was obtained from the "fresh" product stream to minimise any potential for cross-contamination or changes in composition resulting from uncontrolled weathering.

Table 3-3: Summary of material sample source

Material	Code	Source of Laboratory Sample
AGGREGATES		
Asphalt planings	AP	Process waste stream
Blastfurnace slag	BFS	Commercial - stockpile
Basic oxygen steel slag	BOS	Commercial- stockpile
China clay sand	CCS	Commercial- stockpile
Crumbed rubber	CR	Commercial- pre-packed
Foundry sand (phenolic, brass)	FSB	Process waste stream
Furnace bottom ash	FBA	Commercial- stockpile
Granite	GR	Commercial- stockpile
Limestone	LS	Commercial- stockpile
Minestone	MS	Process waste stream
MSW incinerator ash (new incinerator)	MSW (N)	Process waste stream
MSW incinerator ash (old incinerator)	MSW (O)	Process waste stream
Pulverized-fuel ash	PFA	Commercial- stockpile
Sewage sludge incinerator ash	SSA	Process waste stream
BINDERS		
Bitumen	bi	Commercial- pre-packed
Cement	Ce	Commercial- pre-packed
Cement kiln dust	ckd	Commercial- pre-packed
Flue-gas desulphurisation gypsum	fgd	Commercial- stockpile
Granulated blast furnace slag	gbfs	Commercial- stockpile
Quicklime	li	Commercial- pre-packed

Alternative and conventional materials generally exhibit inherent variability resulting from uncontrollable natural or process based irregularities. In addition, some materials are known to experience changes in their physical and/or chemical properties over time. When deciding on the mass of each material to be sampled initially, a compromise had to be made between these two factors to avoid excess analytical expenditure whilst retaining a consistent material composition. The physical volume of material to store and the distance to be travelled to the supply were also key factors. In general, 60kg of each aggregate and 40kg of each binder were initially sampled. This quantity was sufficient for completing the small-scale laboratory testing, described in Chapters 3 to 6, from a single supply of material. Restocking of a reduced number of the materials was required for the large scale testing described in Chapter 7.

To minimise cross contamination of materials during sampling and storage all materials were stored in clean, sealed plastic bags or drums and all equipment used during the sampling procedure was non-metallic, wherever possible, and cleaned before use.

3.3 Assessment of the Chemical Components of the Solid Fraction

To establish some knowledge of the materials being investigated an initial characterisation of the chemical composition of the solid fraction of each material was carried out. This characterisation was not aimed at becoming the sole or principal assessment of the suitability of materials for use, but was considered important in the overall understanding of each material and would enable:

1. The identification of potentially leachable species;
2. A comparison to published composition data to confirm that the laboratory sample is representative of the material as a whole;
3. The determination of the proportion of each component of the solid fraction that has been leached in the different tests (species mobility).

3.3.1 Methodology

The chemical analysis of the solid fraction of each material was carried out by the British Geological Survey, in Keyworth, Nottingham, England. Typically, the analysis identified the presence (or absence) of 17 major oxides, 17 minor elements and 4 compounds and other material properties as listed in Table 3-4. Where the analytical method allowed, the majority of determinands were measured for each material. Assuming the largely organically-inert nature of many of the materials under test, the total organic carbon (TOC) and polycyclic aromatic hydrocarbon (PAH) contents were only determined for those materials that were thought to contain a significant organic content. The loss on ignition (LOI) was determined for all materials.

Table 3-4: Analytical determinands measured in the analysis of the solid fraction

Test	Determinands
Solids - Major Oxides	SiO_2 TiO_2 Al_2O_3 Fe_2O_3t Mn_3O_4 MgO CaO Na_2O K_2O P_2O_5 SO_3 Cr_2O_3 SrO ZrO_2 BaO ZnO V_2O_3
Solids - Minor Elements	V Cr Co Ni Cu Zn Mo Ag Cd Sn Sb Ba Pb U As Se Hg
Solids- Compounds and material properties	polycyclic aromatic hydrocarbons (PAH) cyanide (CN) total organic carbon (TOC) loss on ignition (LOI)

The choice of analytical method was dependent upon certain properties of the individual material and the elemental fraction being determined. For the majority of the materials, one, or both, of the following multi-element techniques was employed:

1. X-ray fluorescence (XRF) spectrometry - The method involves bombarding a solid material sample with x-rays and detecting the wavelengths of the emitted energy that corresponds to the different elemental species. In general, major elemental composition is determined from a sample of material that has been made into a fused bead whilst the minor elements are determined from a pressed powder pellet. The limits of analytical detection are dependant upon the determinand, the method employed and the material;
2. Sample digesting into solution with quantitative analysis of the eluate - The method for analysing the eluate is dependant on the chemical species but usually consists of one or more of the following:
   - Inductively coupled plasma–atomic emission spectrometry (ICP-AES);
   - Inductively coupled plasma–mass spectrometry (ICP-MS);
   - Atomic absorption spectrometry (AAS).

In addition to the multi-element analyses, a number of determinand specific techniques were employed, including the loss on ignition (LOI) determined from the change in mass of a sample subjected to 1050°C for 1 hour.

A number of problems were encountered in determining the composition of the solid fraction that resulted in deviations from the standard analytical methods.

- The MSW incinerator ashes and the basic oxygen steel slag were found to be magnetic and could not be analysed by XRF spectrometry. The analysis of these three materials and the blastfurnace slag (which also proved difficult to analyse) were sub-contracted, by the British Geological Survey, to an external laboratory;
- The crumbed rubber could not be made into a pressed powder pellet due to the nature of the material. The minor elements of the crumbed rubber were determined following acid digestion and ICP-AES, with the undigested portion of the sample assumed to be carbon;
- In preparing the fused beads problems were encountered with the crumbed rubber and the asphalt planings because the high temperature of the fusion process caused the materials to burn. Both materials were pre-ashed before preparation of the beads and a quantitative back analysis carried out;
- The preparation of fused beads can also result in the loss of some sulphur and therefore due to the very high sulphur content in the flue-gas desulphurisation gypsum, this fused bead was prepared using a method designed to retain sulphur;
- A pressed powder pellet could not be prepared for the quicklime because the pellet binder caused the material to swell and break up the sample.

3.3.2 Results

The total solids composition was determined for the 14 aggregates and 4 of the binders listed in Table 3-1 and Table 3-2. The bitumen was not included in this section of the testing because of the difficulties in analysing such a material and the granulated blast furnace slag was not analysed since it was considered to be similar to the blastfurnace slag, which had also proved difficult to analyse. A summary of the major and minor elemental components of each material, in parts per million (ppm), is given in Table 3-5a for the aggregates and Table 3-5b for the binders and activators.

The concentration of each element generally varies over several orders of magnitude between the different materials. A graphical representation of this variation is shown in Figure 3-1 for sodium, barium and chromium, as illustrative examples.

Table 3-5a: Major and minor elemental composition of the solid materials

	Major and Minor Element Composition (ppm)							
Aggregate	**Below limit of detection**	**<1**	**1-10**	**10-100**	**100-1,000**	**1,000-10,000**	**10,000-100,000**	**>100,000**
AP	Mo Ag Cd Sb U Hg	Se	Sn As	V Co Ni Cu Zn Pb *PAH* Sr Zr	Cr Ba Mn P	Ti Na S	*TOC* Al Fe Mg Ca K	Si *LOI*
BFS	Co Ni Mo Cd Sn Sb Pb *CN* As P Zr *LOI*		Cu Zn Ag Se	V Cr U	Ba Sr	Ti Fe Mn Na K S	Al Mg	Si Ca
BOS	Ni Cu Sb Na Sr Zr	Se	Zn Cd Sn	Ag Ba *CN*	V Cr K S	Ti Al P *LOI*	Si Mn Mg	Fe Ca
CCS	Co Cu Mo Ag Cd Sb Hg S Zr	Se	Cr Ni U As	V Zn Sn Ba Pb Sr	Ti Mn Ca P	Fe Mg Na	Al K *LOI*	Si
CR	Ni Mo *CN* Se Ti Mn Mg K P Sr Zr		V Cr Cd Ba	Cu	Co *PAH* Al Fe Ca	Zn Si Na		*TOC LOI*
FSB	Ag Cd Sb U *CN* S Sr	Se	Co Ni Mo Sn Pb As *PAH*	V Cu Zn Mn P	Cr Ba Ti Mg Ca Na	*TOC* Al Fe K Zr	*LOI*	Si
FBA	Cd Hg Sr Zr	Se	Ag Sn Sb U *PAH*	Co Mo Pb As	V Cr Ni Cu Zn Ba	Ti Mn Na P	*TOC* Mg Ca K *LOI*	Si Al Fe
GR	Mo Ag Cd Sn Sb U Hg S	Se	Ni Pb As	Cr Co Cu Zn Sr Zr	V Ba Mn P	Ti	Al Fe Mg Ca Na K *LOI*	Si
LS	Ni Mo Sn Sb Zr	Se	Cr Co Ag U As	V Cu Cd Pb Ti K	Zn Ba Al Fe Mn Na P Sr	Mg	Si	Ca *LOI*
MS	Ag Sn Sb	Se Hg	Mo Cd U	Co Ni Cu Zn Pb As *PAH*	V Cr Ba Mn P S Sr Zr	Ti Mg Ca Na	Fe K	*TOC* Si Al *LOI*
MSW (N)	Mo CN Sr Zr	Se	Cd	V Ag PAH	Cr Ni Sn Sb Mn	Cu Zn Ba Ti Mg K P S	TOC Al Fe Ca Na LOI	*Si*
MSW (O)	U Sr Zr		Se Hg PAH	V Co Ni Mo Ag Cd As	Cr Cu Sn Sb Ba Mn	Zn Pb Ti Mg P	TOC Al Fe Na K LOI	*Si Ca*
PFA	Sr Zr	Hg	Ag Cd Sn Sb U Se PAH	Co Mo	V Cr Ni Cu Zn Ba Pb As	Ti Mn Na P	Fe Mg Ca K LOI	*Si Al*
SSA	Sr	Hg	Cd U Se PAH	V Co Ni Mo Ag As	Cu Sn Sb Pb Zr	Cr Zn Ba TOC Ti Mn Mg Na	Al Fe Ca K P	*Si LOI*

Table 3-5b: Major and minor elemental composition of the solid binder and activator materials

	Major and Minor Element Composition (ppm)							
Binder	**Below limit of detection**	**<1**	**1-10**	**10-100**	**100-1,000**	**1,000-10,000**	**10,000-100,000**	**>100,000**
Ce	Mo Ag Sn Sb Hg	Se	Co Cd U As	V Cr Ni Cu Zn Pb Zr	Ba Mn P Sr	Ti Mg Na K *LOI*	Si Al Fe S	Ca
CKD	Sn Sb U Zr	Hg	Co Mo Ag Cd As	V Cr Ni Cu Zn Se	Ba Pb Mn P Sr	Ti Mg Na	Si Al Fe K S	Ca *LOI*
FGD	Co Ni Mo Sn Sb Pb U Ti Mn K P	As Hg	Cr Cu Zn Ag Cd Se	V Ba Mg Sr	Al Fe Na	Si		Ca S *LOI*
Lime	Cr Co Ni Cu Mo Cd Pb Hg Ti S Zr	As Se	V Zn Ba	Na K P	Al Fe Mn Sr	Si Mg	*TOC LOI*	Ca

X = from minor element analysis, X = from major oxide analysis, *X* = determinand specific technique

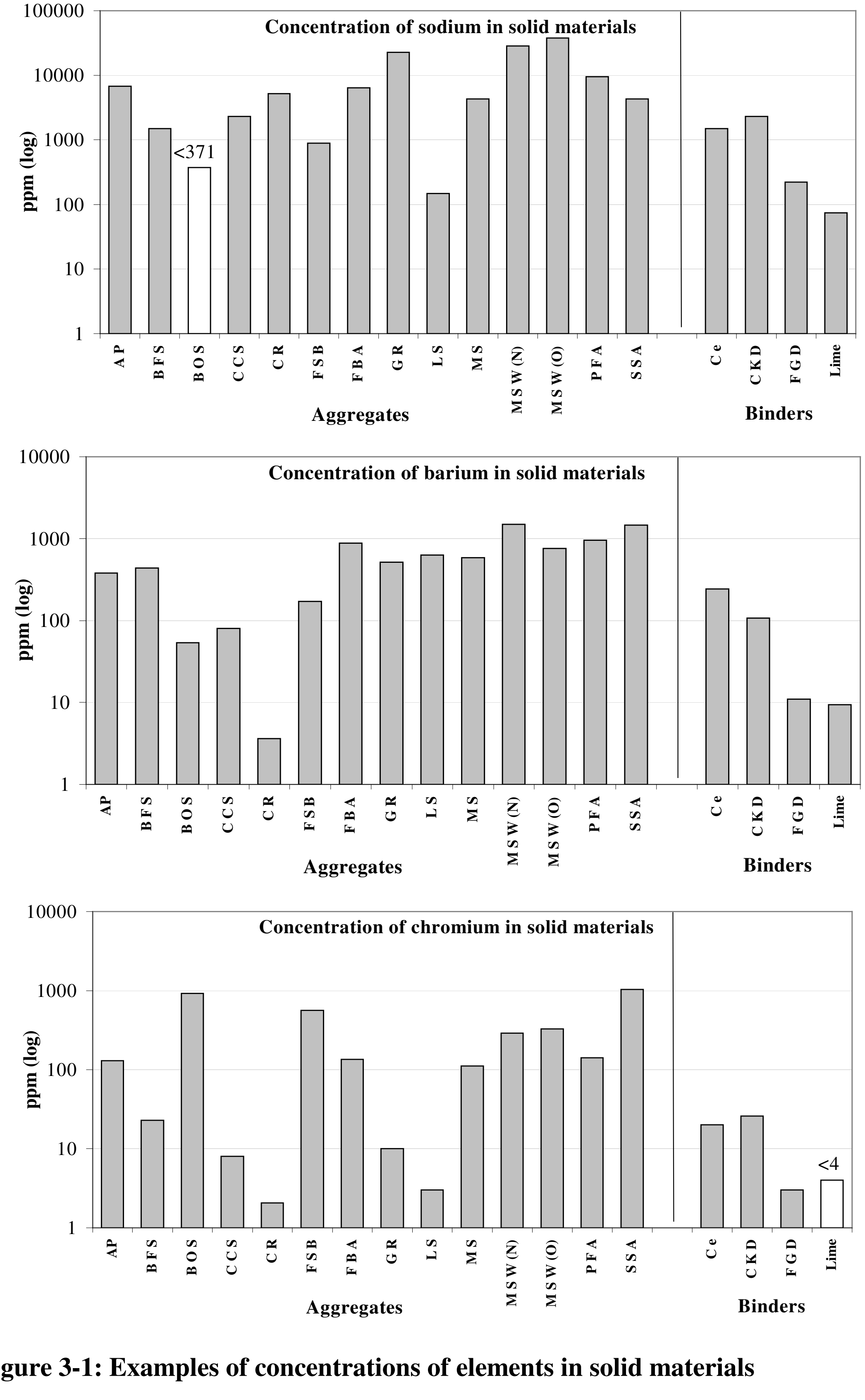


Figure 3-1: Examples of concentrations of elements in solid materials

Figure 3-2 shows graphically the loss on ignition (LOI) shown by each material. In theory, it would be expected that the materials that originate from high temperature processes would have a very low LOI. This is certainly the case for the blastfurnace slag, basic oxygen steel slag and cement, all of which have a LOI of less than 1%. The pulverized-fuel ash and furnace bottom ash both have a LOI of 7.8% which may be higher than expected from such a hot process but does fall within the typical range of 0.5 to 18% quoted by PowerGen (undated). Similarly the LOI from the MSW incinerator ashes and the sewage sludge incinerator ash range between 3.9% (for the new incinerator) to 10.2% (for the sewage sludge ash), which is within the range of 2.6 to 15.2% recorded by Roe (1976). A similar case can be illustrated for the cement kiln dust [Davis and Hooks, 1974] and minestone [Rainbow, 1989]. The crumbed rubber has a very high LOI of 95%.

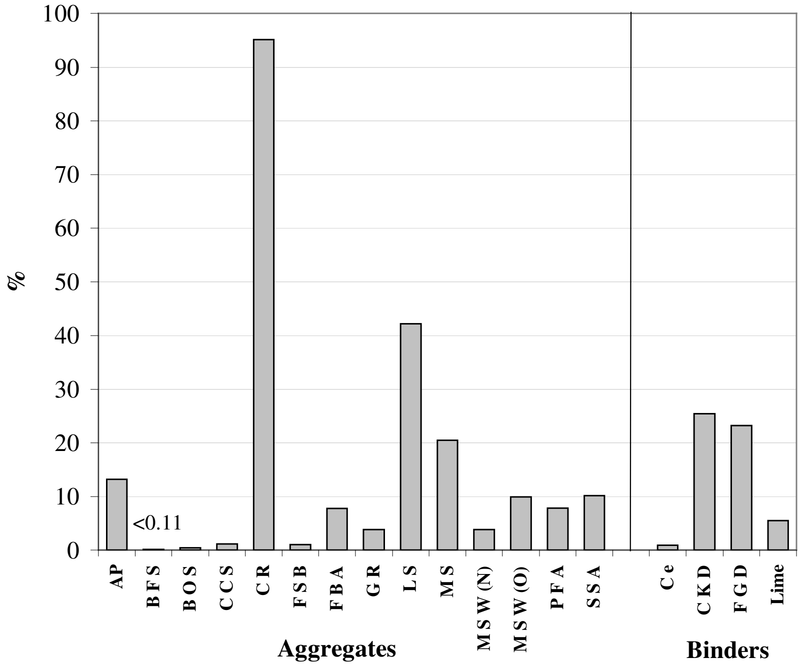


Figure 3-2: Loss on ignition of the solid materials

It can be seen from Table 3-5a and b that, with the exception of the crumbed rubber, (for which the LOI is 95% and total organic carbon content is 83%), all of the materials have either silicon or calcium as their major component. Other major constituents of the materials (greater than 10%) include iron, sulphur and aluminium.

Relative to the other materials, the MSW incinerator ashes and the sewage sludge incinerator ash contain the most significant quantities of many of the minor elements, including nickel, copper, zinc, silver, tin, antimony and barium. The MSW incinerator ashes also contain the highest concentrations of sodium. In addition, the ash from the old incinerator contains significant quantities of cadmium, lead and mercury, relative to the other materials. The sewage sludge incinerator ash also contains relatively high concentrations of chromium, lead and aluminium.

The pulverized-fuel and furnace bottom ashes contain relatively high concentrations of nickel, copper, barium, aluminium and arsenic. Both power station ashes generally have similar chemical compositions, as specified in the literature [Sherwood, 1995] and shown in Figure 3-3. There are also many similarities between the composition of the power station ashes, MSW incinerator ashes and the sewage sludge incinerator ash. With the exception of the sodium and potassium, all the other major elements that were detected were of the same order of magnitude in each of these materials.

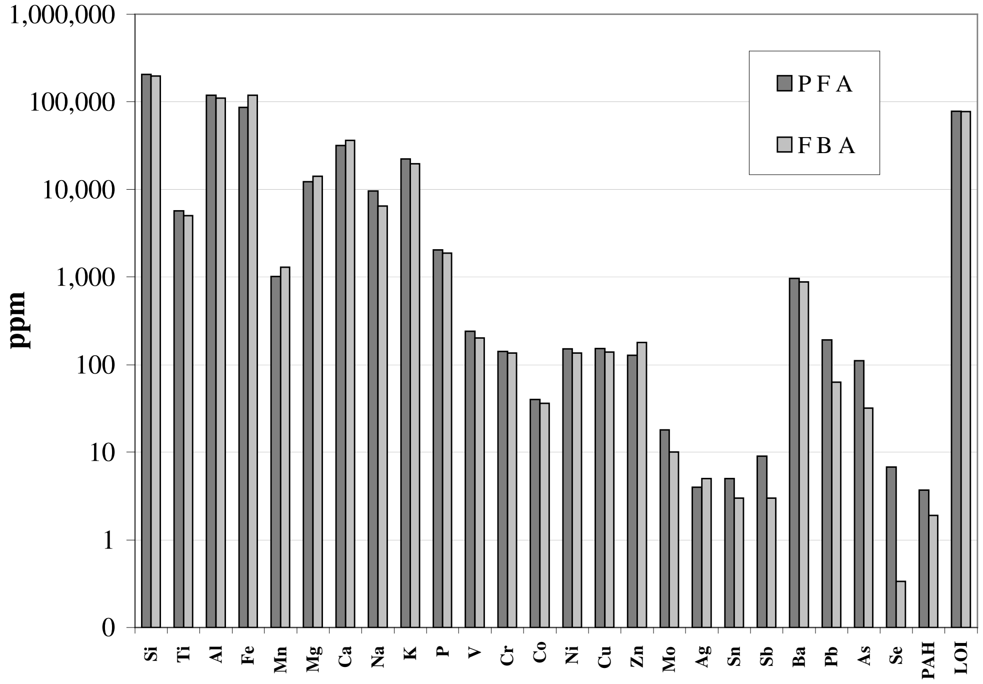


Figure 3-3: Solid composition of pulverized-fuel ash and furnace bottom ash

The basic oxygen steel slag contains relatively high concentrations of manganese, iron, magnesium, vanadium and chromium. The blastfurnace slag also contains high concentrations of magnesium, however, for many species the concentrations in the two slag products differ by more than an order of magnitude and thus it is concluded that they do not have similar chemical compositions.

Relatively high concentrations of cobalt, zinc and PAH were found in the crumbed rubber and chromium and zirconium in the foundry sand (phenolic, brass). In addition, there are relatively high concentrations of aluminium in the minestone, selenium in the cement kiln dust and, not surprisingly, PAH's in the asphalt planings. One surprising result is the relatively high concentration of 12ppm cadmium in the limestone, which is the second highest concentration of this chemical species after the MSW incinerator ash (old).

3.3.3 Discussion

3.3.3.1 Comparison of Material Solid Composition to Literature Sourced Data

The chemical analysis of the solid fraction has shown the variability in the composition of the different materials. To determine whether the laboratory samples show a typical composition for each material, the data has been compared to available data from the literature. Literature data generally consists of some of the major oxides and the loss on ignition. Table 3-6a and b show the data from the literature alongside that obtained from the solids analysis. For those materials and binders excluded from Table 3-6a and b no literature sourced data was available.

Observations of Table 3-6a and b generally show a good correlation between the literature and the laboratory samples obtained for use in this research. The china clay sand, foundry sand (phenolic, brass), pulverized-fuel ash, cement and lime show excellent correlation with the data from the literature for all the major oxides for which the comparison was made. The laboratory samples of these materials are therefore considered to be representative of the material as a whole. The blastfurnace slag, basic oxygen steel slag, furnace bottom ash, minestone, and cement kiln dust show good correlation for most species although each material shows one or two species that deviate from published concentrations. Generally, these deviations are small and may often be an inherent property of the material. Therefore, these materials are also considered to suitably representative.

Table 3-6a: Comparison of the chemical composition of the solid fraction of the aggregates with data from literature

Material	Reference	SiO_2 (%)	TiO_2 (%)	Al_2O_3 (%)	Fe_2O_3t (%)	MgO (%)	CaO (%)	Na_2O (%)	K_2O (%)	P_2O_5 (%)	SO_3 (%)	LOI (%)
BFS	**Lit. 1, 7**	28-36	0.8	9-22	0.3-1.7	4-15	33-45	0.7	0.8	0.6	0.53	-
	Test	*35.65*	*0.50*	*12.87*	*0.59*	*8.38*	*39.87*	*0.20*	*0.40*	*<0.05*	*1.80*	*<0.11*
BOS	**Lit. 1, 7, 8**	9-24	0.3-1	0.5-18	5-30	0.5-8	31-51	0.05-0.3	0.02-0.4	0.03-1.8	0.04-0.9	-
	Test	*14.12*	*0.73*	*1.11*	*28.52*	*6.45*	*40.93*	*<0.05*	*0.05*	*1.51*	*0.22*	*0.43*
CCS	**Lit. 1**	75-90	0.05-0.15	5-15	0.5-1.2	0.05-0.5	0.05-0.5	0.02-0.75	1-7.5	-	-	1-2
	Test	*81.23*	*0.12*	*10.05*	*1.16*	*0.23*	*0.12*	*0.31*	*4.76*	*0.06*	-	*1.18*
FSB	**Lit. 10**	95.9	0.05	1.25	0.65	0.17	0.01	0.20	0.63	-	-	1.18
	Test	*95.68*	*0.05*	*1.47*	*0.80*	*0.06*	*0.03*	*0.12*	*0.99*	*0.01*	*<0.05*	*1.04*
FBA	**Lit.**	50	1	29	11	1.5	2	1	3.5	-	-	-
	Test	*42.09*	*0.84*	*20.81*	*17.01*	*2.35*	*5.10*	*0.87*	*2.37*	*0.43*	-	*7.77*
MS	**Lit. 1, 9**	37-55	0.5-1	16-23	0.5-11	0.9-3.2	0.4-4.9	0.1-0.8	0.8-3.6	0.1-0.25	0.5-2.5	10-40
	Test	*45.40*	*0.75*	*21.25*	*4.97*	*1.56*	*0.90*	*0.58*	*3.68*	*0.17*	*0.1*	*20.47*
MSW(N)	**Lit. 1, 6**	35-55	0.5-0.6	22-23	9-26	0.5-0.7	5-6.3	2-4	<0.5	-	<1	2-15
	Test	*50.02*	*0.91*	*9.97*	*11.94*	*1.48*	*11.21*	*3.85*	*1.18*	*0.70*	*1.35*	*3.87*
MSW(O)	**Lit. 1**	35-55	0.5-0.6	22-23	9-26	0.5-0.7	5-6	2-4	<0.5	-	<1	2-15
	Test	*43.69*	*1.06*	*8.59*	*9.12*	*1.50*	*14.86*	*5.11*	*1.64*	*1.16*	-	*9.87*
PFA	**Lit. 1**	39-56	0.5-1.2	20-34	5-16	1-4.4	1-5.4	0.05-1.7	1-4.5	0.2-0.8	0.3-1.5	1-25
	Test	*44.22*	*0.95*	*22.56*	*12.28*	*2.05*	*4.44*	*1.29*	*2.69*	*0.47*	-	*7.83*

Table 3-6b: Comparison of the chemical composition of the solid fraction of the binders and activators with data from literature

Material	Reference	SiO_2 (%)	TiO_2 (%)	Al_2O_3 (%)	Fe_2O_3t (%)	MgO (%)	CaO (%)	Na_2O (%)	K_2O (%)	P_2O_5 (%)	SO_3 (%)	LOI (%)
ce	**Lit. 2**	20.4	-	5.8	2.2	1.1	65	0.2	0.6	-	3.2	1.1
	Test	*21.12*	*0.22*	*5.35*	*2.14*	*0.98*	*65.75*	*0.20*	*0.56*	*0.08*	*2.8*	*0.91*
ckd	**Lit. 3, 4, 5**	11-16	-	2.1-5.5	1.5-2.9	0.9-2.5	43-53	0.7-0.9	6	--	2.0-7.3	21.5-29.1
	Test	*13.74*	*0.21*	*4.27*	*2.09*	*0.56*	*44.35*	*0.31*	*4.57*	*0.18*	*3.4*	*25.43*
li	**Lit. 2**	0.74	-	0.11	0.07	0.47	95.6	-	-	-	-	-
	Test	*0.73*	*0.00*	*0.08*	*0.12*	*0.43*	*93.98*	*0.01*	*0.01*	*0.01*	*0.0*	*5.51*

References to Table 3-6a and Table 3-6b

1 BS 6543 (1985)
2 Nunes (1997)
3 Davis and Hooks (1974)
4 Sherwood, Tubey and Roe (1977)
5 Millar and Zaman (2000)
6 Roe (1976)
7 Lee (1974)
8 Renfroe (1979)
9 Rainbow (1989)
10 May (1997)

The two MSW incinerator ashes vary more significantly from the composition quoted in the literature. This may have occurred for a number of reasons. Firstly, incinerator ashes are inherently variable depending, in part, upon the waste source being burnt, the type of furnace and the collection and storage of the bottom and fly ashes. Secondly, the quoted literature dates from 1976 and 1985 and since then technology has moved on to provide more efficient incineration, in different plant with tighter restrictions on gaseous emissions. All of these factors may effect the composition of the resulting ash.

In general it can, however, be concluded that the laboratory samples of the different materials offer a good representation of the individual materials under investigation. This fact gives confidence in the sampling and analytical procedures used in this research.

3.3.3.2 Normalisation of the Chemical Composition Data

The absolute concentrations of the different chemical species in the solid materials provides useful information but does not give any indication of the relative toxicity of each of the chemical species and the potential risks associated with, say, 100ppm sodium compared to 100ppm cadmium. Any requirements for elemental composition, whether it be the solid components of a contaminated soil or the dissolved components of drinking water, are set at different limit concentrations depending, principally, on the toxicity, or ability to cause harm, of the individual chemical species. To enable the variable elemental toxicity to be included in the discussion of the solid composition of the materials, the data has been normalised with respect to published acceptance criteria for the assessment of contaminated soil. In this instance the normalisation has been made, arbitrarily, to the Netherland’s Target Values for concentrations in soil [See Appendix A]. It is not suggested that such criteria should be used as a means of assessing the acceptability of any material for use (since the chosen criteria are not likely to be directly applicable to the construction situation being investigated) but, rather, to enable comparative interpretation of the data. Table 3-7a and b show the components of each material (aggregates and binders respectively) that were greater or less than the value quoted in the criteria. The comparison has been made for all of the chemical species that were both determined in the solid materials and listed in the criteria. Those species were chromium, cobalt, nickel, copper, zinc, molybdenum, cadmium, barium, lead, arsenic, mercury and PAH. Species not listed in Table 3-7a and b may exist in that material but may not have a criteria value to normalise to, or are below the limits of analytical detection. In either case, normalisation is not possible.

Table 3-7a: Normalised aggregate solid composition data

Aggregate	Normalised concentration in material			
	< 1	1-10	10-100	>100
AP	Co Ni Cu Zn Mo Pb As Hg	Cr Ba	PAH	
BFS	Cr Co Ni Cu Zn Mo Pb As	Ba		
BOS	Ni Cu Zn Ba	Cr	Cd	
CCS	Cr Co Ni Cu Zn Mo Ba Pb As Hg			
CR	Cr Ni Mo Ba	Cu Cd	Co	Zn PAH
FSB	Co Ni Cu Zn Mo Ba Pb As	Cr PAH		
FBA	Pb Hg	Cr Co Ni Cu Zn Mo Ba As PAH		
GR	Cr Ni Zn Mo Pb As Hg	Co Cu Ba		
LS	Cr Co Ni Cu Mo Pb As	Zn Ba	Cd	
MS	Cu Zn Mo Pb As Hg	Cr Co Ni Cd Ba	PAH	
MSW (N)	Mo	Cr Ni Cd Ba	Zn PAH	Cu
MSW (O)	Co As	Cr Ni Mo Ba PAH	Cu Zn Cd Pb Hg	
PFA	Zn	Cr Co Ni Cu Mo Cd Ba Pb As Hg PAH		
SSA	As Hg	Co Ni Zn Mo Cd Ba Pb PAH	Cr Cu	

Table 3-7b: Normalised binder and activator solid composition data

Binder/ Activator	Normalised concentration in material			
	< 1	1-10	10-100	>100
ce	Cr Co Ni Cu Zn Mo Pb As Hg	Cd Ba		
ckd	Cr Co Ni Cu Zn Mo Ba As Hg	Cd Pb		
fgd	Cr Co Ni Cu Zn Mo Ba Pb As	Cd	Hg	
li	Cr Co Ni Cu Zn Mo Ba Pb As Hg			

It can clearly be seen from Table 3-7a and b that only the china clay sand and quicklime contain all the listed elements at normalised concentrations less than unity (i.e. absolute concentrations below the corresponding criteria). The crumbed rubber and MSW incinerator ash (old incinerator) contain components at concentrations at over 100 times the normalising level. Interestingly, even the limestone and granite primary aggregates contain several elements at normalised concentrations greater than unity, and for the limestone, the normalised concentration of cadmium was greater than 10.

From these latter observations, it becomes apparent that such a technique is unsuitable for assessing the acceptance or not of a material for use in road construction. If such criteria were used as acceptance levels, then it would result in the exclusion from use of these primary aggregate materials, which would be inappropriate on three grounds. The first is pragmatic, in that if primary aggregate materials were excluded from use then it would largely prevent road construction of any type. The second reason is due to the natural geological source of such materials. It can hardly be considered deleterious to put back into the environment the same material as has just been abstracted. Thirdly, the solid composition of the material does not provide any indication of whether the components of a material will actually be available to the wider environment. Although such observations emphasise the limitations of using such a comparison for the purpose of material assessment, it does however, highlight the particular components of each material that could be present in the leachates to be examined later.

In addition to the identification of the individual species described above, another tool that was considered to be of use in the evaluation of the solids composition data was a weighted indicator of the relative overall toxicity of one material compared to another. Such an indicator should enable the large data set to be reduced to a single value for each material, thus making it a simplistic task to compare the different materials. Scott et al. (1995) addressed a similar issue for establishing the cumulative toxicity of landfill gases. Drawing on information from the American Conference of Governmental Industrial Hygienists and the UK Health and Safety Executive (HSE) (for the purpose of the COSHH Regulations), Scott et al. proposed that the toxicological effects of mixed compounds could be considered additive when the actual effect of the mixture was unknown. Equation 3-1 was applied to the mixed gases to give a toxicity factor for a particular target.

$$\frac{C_1}{T_1}+\frac{C_2}{T_2}+\frac{C_3}{T_3}+\dots\dots\dots\frac{C_n}{T_n}=F \qquad \textbf{Equation 3-1}$$

Where,

C_n = concentration of compound n,

T_n = toxicity threshold limit of compound n,

F = toxicity factor on target.

A similar approach has been applied to the material solid composition data presented in this chapter. The normalised data for each species for which a criteria value was available (Table 3-7a and b) were summed to give a normalised approximation to a toxicity factor or index. Again, it is not suggested that such a toxicity index should be used as a means of assessing the acceptability, or not, of any material for use, but, used here, it provides a useful tool for simplifying the data set to enable comparison.

The normalisation was repeated for both the maximum and minimum possible concentrations of each species. So, for example, a concentration of <2ppm, was interpreted as zero ppm when normalising to the minimum and 2ppm when normalising to the maximum. The difference between the two interpretations was minimal and only affected the ranking (by one position) of a couple of materials at the lower end of the range. Table 3-8 provides a summary of the toxicity index (based on maximum species concentrations) for each material, ranked in descending order.

Table 3-8: Relative 'Toxicity Index' of the composition of the solid fraction for the aggregates and binders

Rank	Material	Toxicity Index
1	CR	422
2	MSW(O)	167
3	MSW(N)	160
4	SSA	75
5	AP	53
6	PFA	36
7	MS	28
8	FBA	24
9	**LS**	**22**
10	BOS	22
11	FSB	20
12	CKD*	16
13	**GR**	**10**
14	Ce*	9.4
15	BFS	6.7
16	FGD*	6.4
17	CCS	5.0
18	Lime*	3.7

- = binder or activator, **bold** = primary aggregates

Note: Such rankings should not be used as a means of accepting or rejecting a material for usage.

The conclusions to be drawn from Table 3-8 are similar to those made from Table 3-7a and b. Again, the china clay sand and quicklime show the least potential toxicity whilst the crumbed rubber and MSW incinerator ashes show the greatest potential toxicity, based on the solid composition data. The primary aggregates, especially the limestone, also fall surprisingly high in the rankings. One interesting interpretation of this data could be that any material with a toxicity index less than, or in the same region as, the primary aggregates could be regarded as acceptable based on the argument presented earlier for the unconditional use of primary materials. However, because of the possible inappropriateness of the applied acceptability criteria and probable inability of the solids composition to make any reasonable prediction of component leachability, any such interpretation should be treated with extreme caution.

3.3.4 Summary and Conclusions

- 14 aggregate materials and 4 binders were analysed to determine the concentrations of chemical components in the solid materials.

- With the exception of crumbed rubber, all materials have either silicon or calcium as their major component. Other major constituents included iron, sulphur and aluminium.

- The actual concentrations of the individual chemical species vary by over 3 orders of magnitude or more between the different materials.

- The solid composition of each material that was tested generally showed a good correlation with typical composition values obtained from the literature. Thus it can be concluded that the laboratory samples are a good representation of each material and that the material source and sampling procedures did not introduce any significant cross-contamination.

- The solid composition data was normalised by reference to published acceptance criteria. This assessment illustrated the importance of considering the toxicity of different chemical species when making a judgement about concentrations of different elements within a material. It also highlighted how established acceptability criteria may not be appropriate for the assessment of construction materials, since the conventional limestone and granite aggregates contained chemical species at concentrations above those specified in the criterion.

- A "toxicity index", or sum of normalised concentrations, was used as a tool for weighting the concentrations of the individual species to give an indication of the relative environmental quality of each material. Although such methods provide a useful tool, they should be used with caution as they do not illustrate the complete picture of the material.

3.4 Rapid Leaching Characterisation

To establish some knowledge of the leachability of the various components of each material, a simple and rapid leaching test was required. This initial leaching characterisation was not expected to directly simulate conditions found within a road construction because, in general, the material properties and leaching conditions applied in such tests are far removed from those that would be found in-situ. In this way, such rapid testing is likely to be inappropriate as an assessment tool. However, the characterisation provides a quick approximation of leachability that can indicate whether the components of a material have the ability to present any potential risk. In addition, it enables the relationship between the presence of a component in the solid fraction and the ability of that component to leach to be investigated.

Numerous leaching tests exist for simplistic investigation of leaching potential as described in Chapter 2 [Environment Canada, 1990]. The rapid assessment method chosen was the draft compliance test being developed by CEN Technical Committee 292, prEN 12457 "Compliance test for leaching of granular wastes and sludges" [CEN, 1996] (Procedure C). Procedure C is a two-batch, agitated leaching test performed at a liquid to solid ratio (L/S) of 2 for the first 6 hours, followed by 18 hours at a cumulative L/S of 10. The reasons for selecting this test procedure included:

a) At the time of study, this test was the most up-to-date test being developed in Europe by CEN Technical Committee 292 "Characterisation of waste” as part of a series of co-ordinated standards dealing with leaching procedures;

b) The adopted test method was recommended by Baldwin et al. (1997) following their use of this and other test methodologies on selected alternative aggregates;
c) The procedure enables some limited understanding of the leaching behaviour to be obtained by distinguishing between constituents that leach rapidly, consistently and variably.

Since the commencement of this research the CEN TC 292 compliance test for leaching of granular wastes and sludges has been formalised as a British and European Standard for the characterisation of waste [BS EN 12457 – Part 3, 2002]. The test procedure is largely unchanged from the 1996 draft although some modifications to the methods of data analysis and the size of the material sample have been made.

Other reasons for selecting the CEN compliance test as the rapid leaching characterisation test were:

- The test allows the pH of the leachates to be governed by the solid material being tested rather than independently controlling the pH of the leachate. This is considered to be appropriate since the leachants in the in-situ environment are likely to be rainwater or groundwater that generally have a near neutral pH;
- On a more practical level the test only requires two samples of leachate to be analysed for each test, which is fewer than in many other rapid leaching test procedures. Each additional analysis increases the cost of carrying out the leaching test, and minimising the cost of analysis was important for this research and for the construction industry as a whole.

The CEN TC 292 draft “Compliance test” was not developed specifically to assess leachability for the scenario of road construction materials. It was developed purely as a leaching test for verifying the leachability of different granular wastes and sludges. The conditions employed within the test differ substantially from those occurring in the road construction environment, as follows:

a) The test materials are size reduced to 100% below 10mm and 90% below 4mm. Typical graded aggregates have particle sizes ranging from dust up to 40mm or sometimes 75mm. The size reduction significantly increases the surface area to volume ratio of the material, thus increasing the surface area in contact with the leachate. The smaller particles also have a shorter diffusion path length within each particle, which may accelerate leaching by diffusion. Particle size reduction also exposes "new" or "fresh" surfaces which have not previously been weathered. All of these factors may result in accelerated and/or increased leaching;

b) The material and leachant are mixed continuously by agitating in a tumbling device. Such harsh mixing would never occur in a road construction scenario and accelerated and/or increased leaching was anticipated;

c) The test is carried out at liquid to solid ratios (L/S) of 2 and 10, which is much higher than those found (at any one time) in the road construction environment, where the void ratio of a compacted aggregate would, typically, be approximately 0.3, and imply a L/S ratio by volume of approximately 0.5. The large L/S ratios potentially enable a larger quantity of each component to be leached (particularly if the leaching process is limited by the solubility of the species). However, dilution of the leached species may also occur, resulting in lower species concentrations than that which would have arisen from a lower L/S ratio;

d) The test is principally for use in assessing leaching of inorganic fractions. Errors may occur in determining the leaching of certain organic fractions because the plastic leaching vessels and other equipment used to minimise inorganic cross-contamination may contribute organic species.

3.4.1 Methodology

The rapid leaching characterisation tests were carried out in accordance with Procedure C of the draft "Compliance test for leaching of granular wastes and sludges" [CEN, 1996]. The details of the methodology developed from the procedure are given in Box 3-1 below.

Box 3-1: Methodology for the rapid leaching characterisation test [CEN, 1996]

1) *Obtain a representative sub-sample* of the material with a mass of approximately 2kg, by using coning and quartering [BS 812: Part 102, 1989; BS EN 932-1, 1997].
2) *Air dry* the material if it contains particles larger than 4mm in diameter.
3) *Sieve* the material through 10mm and 4mm aperture sieves. 100% of the material must pass the 10mm sieve and at least 90% must pass the 4mm sieve.
4) *Crush* any oversized material. Any material greater than 10mm is crushed to below 10mm and then any material greater than 4mm is crushed so that less than 10% of the material is greater than 4mm. Any size reduction should be kept to a minimum.

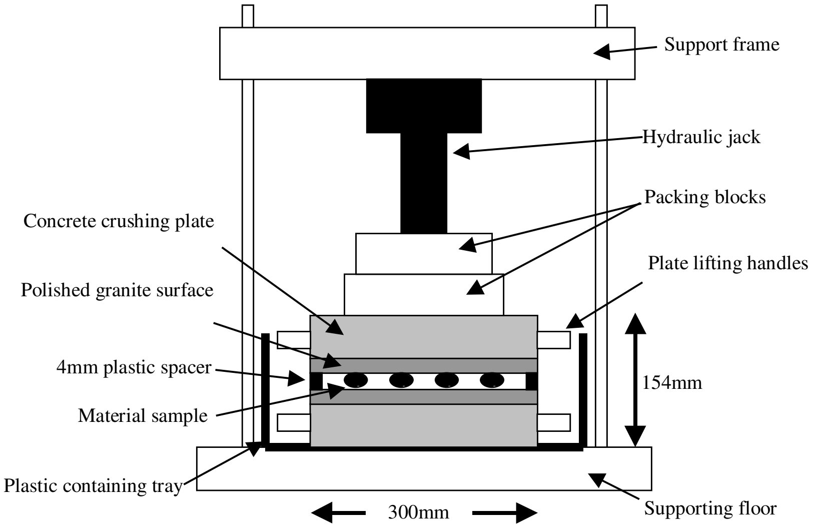
5) *Determine the residual moisture content* of the test material by oven drying a sample to constant weight [BS 812: Part 109, 1990; BS EN 1097-5, 1999]. The held water is taken into account in establishing the test liquid to solid ratio.
6) *Phase 1 leaching.* Add 100g ± 5g (dry weight) of material to a 1 to 2 litre leaching vessel. Quantitatively add distilled water leachate (5<pH<7, conductivity <5µS/cm) to the leaching vessel to establish a liquid to solid ratio of 2 l/kg ± 2% (L/S=2), taking into account the moisture held within the test sample. Agitate the sample for 6 hours ± 0.5 hours.
7) *Filtration.* Allow the suspended solids to settle for 15 minutes ± 5 minutes. Filter the leachate through a 1.2µm glassfibre filter and a 0.45µm nylon filter paper. The time allowed for filtration is 1000/A (minutes), where A is the area of the filter paper (cm^2) (approximately 57 minutes for a 47mm diameter filter paper). Where filtration cannot be achieved within this time the leachant can be centrifuged prior to filtration. Record the mass of leachate that is removed.
8) *Sample preservation.* Measure and record the pH and conductivity of the leachate. Retain 3 samples of the leachate for the analysis (1 x unacidified (~30ml), 1 x 1% nitric acid (~20ml), 1 x 1% hydrochloric acid (~20ml)).
9) *Phase 2 leaching.* Quantitatively add distilled water leachant to the leaching vessel from the phase 1 leaching to establish a cumulative liquid to solid ratio of 10 l/kg ± 2% (L/S=10). Leachant with a mass of approximately 8 times the mass of the material should therefore be added. To minimise any loss of material during the filtration process, rinse the filtration equipment with the new leachant and add the used filter papers to the leaching vessel. Agitate the sample for 18 hours ± 0.5 hours.
10) *Filtration and sample preservation 2.* Repeat stages 7 and 8 for the leachant generated during phase 2 leaching.

3.4.2 Equipment Development

Test procedures and equipment were selected to minimise any cross-contamination of inorganic substances. Plastic (polyethylene or polypropylene) laboratory ware, with high strength and low chemical adsorption, was used. This included the leaching vessel, filter flask and any beakers or other containers that were in contact with the material sample, the leachant or leachate.

3.4.2.1 Equipment for Particle Size Reduction

To carry out any particle size reduction the procedure states that "crushing equipment" should be used and that "a jaw crusher is the preferred device for size reduction". Although a jaw crusher device was available for use it was not considered to be suitable because of the build up of dust and potential contaminants in the equipment from previous use. Therefore, an alternative crushing device was designed and fabricated which was easy to clean and would minimise cross-contamination of the sample. A schematic diagram of the set up is shown in Figure 3-4.



(NOT TO SCALE - APPROXIMATE DIMENSIONS)

Figure 3-4: Schematic diagram of the particle size reduction equipment

The two crushing plates were made from polished granite slabs (300mm x 300mm x 25mm). This material was chosen because of the low contaminating composition, reasonable strength properties and easy to clean surface. The granite slabs were mounted on a block of high compressive strength concrete (approximately 50mm thick) to increase the overall strength of the slabs. Sockets for taking lifting handles were set into the sides of the concrete to enable lifting. The material to be crushed was placed between the two granite surfaces of the slabs. The crushing plates were placed in a plastic tray to collect any material sample that overflowed the plates. To prevent excessive crushing of the sample, 4mm spacers were placed between the slabs. The crushing load was applied using a hydraulic jack attached to a steel rig over the crushing plates until the 4mm spacers were contacted.

3.4.2.2 Equipment for Sample Agitation

For carrying out the mixing or agitation of the samples during the test, the procedure recommends using, “an end-over-end tumbler (5 rpm - 10 rpm) or rollertable rotating at about 10 rpm". The end-over-end method was selected as it was considered to be a more aggressive mixing action. Commercially available agitation devices were generally not capable of holding several leaching vessels of the sizes that would be required for this test (500ml, 1000ml and 2000ml). Therefore, a device was designed and fabricated to meet the necessary requirements and is illustrated schematically in Figure 3-5 and photographically in Figure 3-6.

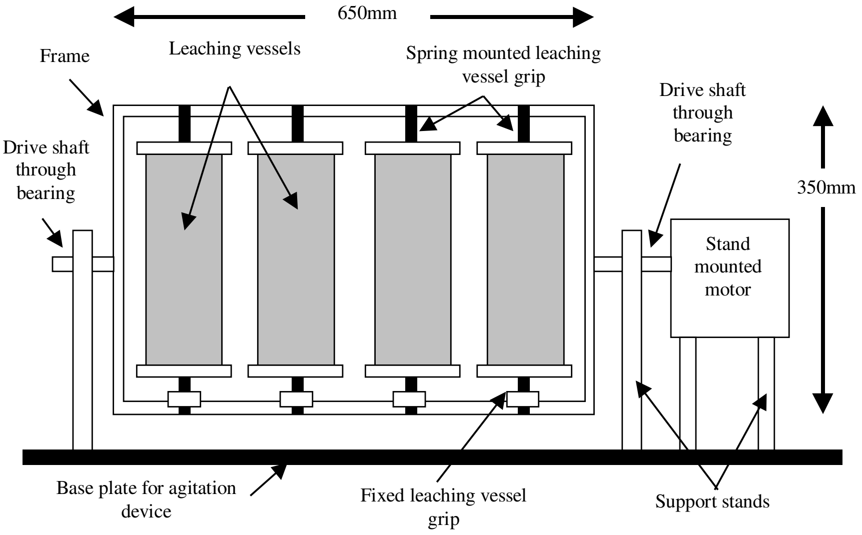


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Figure 3-5: Schematic diagram of the end-over-end agitation device

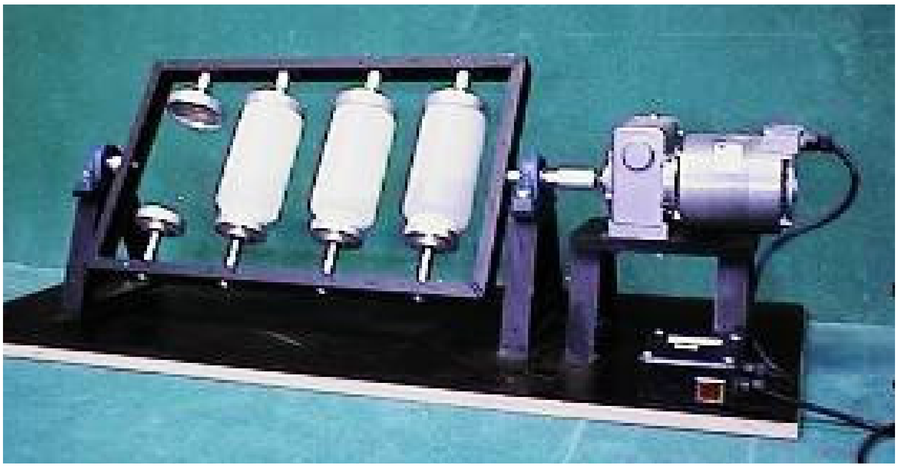


Figure 3-6: Photograph of fabricated end-over-end agitation device

In the agitation device the leaching vessels are clamped inside a rectangular metal frame that is rotated about its central horizontal axis by an electric motor operating at 8rpm. The leaching vessels are clamped in place using a pair of metal discs that have been machined to fit the ends of the leaching vessels. The metal discs and supporting rods are interchangeable to enable leaching vessels of various sizes to be held. One of the discs is on a fixed support whilst the other is spring mounted to enable the leaching vessels to be inserted.

3.4.3 Leachate Analysis

The pH and conductivity of the leachate samples were measured in the laboratory, using ion selective electrodes, immediately following sample filtration. The full chemical analysis of the leachate samples was carried out by the British Geological Survey, in Keyworth, Nottingham, England. The leachates were typically analysed for the 37 different components listed in Table 3-9. Where the analytical methods allowed, the majority of the determinands were measured in each leachate. The analytical laboratory conducted ionic balances on the results as a quality check to ensure that the data was reliable. The data generally has a confidence limit of about ±10 to 20%. For very low concentrations (i.e. parts per billion) and concentrations close to the limit of analytical detection, the degree of uncertainty may be substantially greater [Cave, 1998].

Table 3-9: Analytical determinands for liquid leachate samples

Test	Determinands
Leachates	pH, conductivity, Ca, Mg, Na, K, Total alkalinity (HCO_3), Cl, sulphate (SO_4), nitrate (NO_3), Br, nitrite (NO_2), F, Total organic carbon (TOC), Total inorganic carbon (TIC), Total P, Total S, ammonium (NH_4), Si, Ba, Sr, Mn, Total Fe, Al, Co, Ni, Cu, Zn, Cr, Mo, Cd, Pb, V, Li, B, As, Se

The leachate samples were stored in 30ml ‘Sterylin’ tubes and refrigerated, prior to analysis. Three samples of each leachate were retained (See Box 3-1, step 8) and used in the analysis in the following ways:

a) *Unacidified sample* - used to determine the concentration of the different anions (including chloride, sulphate, nitrate and nitrite). The analytical techniques included ion selective electrodes and ion chromatography;

b) *Sample acidified to 1% nitric acid* - used to determine the concentration of the majority of the cations. The analytical techniques of ICP-AES and ICP-MS were used (See Section 3.3);

c) *Sample acidified to 1% hydrochloric acid* - used to determine the concentration of specific elements (including mercury, arsenic and selenium) that exhibit limitations (such as high detection limits) when determined using ICP techniques.

A number of analytical methods, including atomic absorption spectrometry (AAS) and atomic fluorescence spectrometry are used [British Geological Survey, undated].

3.4.4 Sample Preparation and Testing

The rapid leaching characterisation was carried out on all of the aggregate materials, binders and activators under investigation in this research, with the exception of the bitumen, for which the test could not be carried out, and the granulated blast furnace slag, which is assumed to be similar to the blastfurnace slag. The materials which were subjected to the test are listed in Table 3-10a and b (for aggregates and binders respectively) alongside details of the sample preparation that was applied to each material.

Table 3-10a: Sample preparation for the rapid leaching characterisation tests on the aggregates

Material	Abbreviation	Air Drying	Size Reduction
Asphalt planings	A P	✓	✓
Air-cooled blast furnace slag	B F S	✓	✓
Basic oxygen steel slag	B O S	✓	✓
China clay sand	C C S	×	✓
Crumbed Rubber (0.5-1.5mm)	C R (0.5-1.5)	×	×
Crumbed Rubber (1-4mm)	C R (1-4)	×	×
Foundry sand (phenolic,brass)	F S B	×	✓
Furnace bottom ash	F B A	✓	✓
Granite	G R	✓	✓
Limestone	L S	✓	✓
Minestone	M S	✓	✓
MSW incinerator ash (New incinerator)	M S W (N)	✓	✓
MSW incinerator ash (Old incinerator)	M S W (O)	✓	✓
Pulverised fuel ash (Conditioned)	P F A	×	×
Sewage sludge incinerator ash	S S A	×	×

Table 3-10b: Sample preparation for the rapid leaching characterisation tests on the binders and activators

Material	Abbreviation	Air Drying	Size Reduction
Cement	ce	×	×
Cement kiln dust	ckd	×	×
Flue-gas desulphurisation gypsum	fgd	×	×
Quicklime	li	×	×

To determine the extent of any cross-contamination that results from the leaching test procedure, two blank rapid leaching tests were performed. These tests involved following the defined test procedure but with no solid material in the leaching vessel. The results of these blank tests were to be used to quantitatively adjust the data obtained for the tests on the different materials to account for the species that resulted from the test procedure. Samples of the distilled water leachant were also analysed to determine whether the said species arise from the leachant or from the test procedure.

To give an indication of the test repeatability the complete procedure of sample drying, size reduction and testing was repeated on the granite. Ideally, repeat tests would have been carried out on all materials. However this was not possible due to the expense of the analytical testing.

The issue of repeatability was discussed in a workshop at the WASCON 2000 conference [Harmonisation Network, 2000] and is now included as an appendix to BS EN 12457 (2002). It was found that the parameters that imparted the most significant variation to the test result were the temperature, method of agitation and method of size reduction. The liquid to solid ratio, mass of solid sample and diameter of the filter paper were sometimes significant. In this research the method of sample size reduction and agitation were constant and the temperature of the tests reasonably well controlled.

In carrying out the sample preparation, testing and analysis, a number of difficulties were encountered with certain materials and leachates.

- The temperature of the asphalt planings had to be reduced to below 0°C to enable the material to be crushed (rather than compressed);
- The MSW incinerator ash (new) contained 2.7g of non-crushable material from the laboratory sample of 1328g;
- When adding the leachant to the sample of quicklime a considerable amount of heat was generated by the hydration reaction. To prevent the polypropylene leaching vessel from melting, the leachant had to be added over a period of approximately 1.5 hour. To compensate the sample was agitated for the minimum specified time of 5.5 hours, in phase 1 of the test (L/S=2);
- A number of the leachates could not be filtered through the 0.45µm filter paper within the specified time (57 minutes for a 47mm diameter paper).

- For the test on the minestone, the leachate proved difficult to filter due to the high proportion of clay in the sample. The samples were centrifuged and the filtration process took approximately 2 to 3 hours at each stage of the test.
  - For the test on the quicklime it was not possible to filter a sufficient quantity of the phase 1 leachate (L/S=2) for the analysis. The phase 2 leachant (L/S=2-10) was simply added to the leaching vessel and the test was continued.
- The leachates from the cement and cement kiln dust samples contained relatively high concentrations of certain alkali and alkali earth metals. High concentrations of such species can result in interference and errors in the analytical methods, which require such leachates to be diluted prior to analysis. However, dilution of the sample results in elevated detection limits and some species that may have been at detectable concentrations in the original leachate may have become undetectable.

3.4.5 Results

3.4.5.1 Correction for the Blank

A summary of the data obtained from the blank compliance tests and distilled water leachant samples is provided in Table 3-11, which shows the average concentrations of chemical species in the leachate in milligrams per litre (mg/l). The average concentrations could only be calculated for those chemical species that were present above the limits of analytical detection. The species that are listed in Table 3-9 but are not in Table 3-11 may exist but at concentrations below the limits of analytical detection.

Table 3-11: Species detected in the blank rapid leaching tests (average of 2) and distilled water leachant (average of 4)

Test	**L/S**	**pH**	**Cond. µS/cm**	**Concentration of detectable determinands, mg/l**				
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**
Blank rapid leaching test	**2**	4.7	43	Sr	Ba Zn	Ca Na Cl Si Se		TOC
	2-10	4.6	39	Sr	Ba	Ca Na Cl Si Zn Se		TOC
Distilled water leachant		5.2	7		Ba Zn	Ca Na Cl TIC Si Se	TOC	

From Table 3-11 it can be seen that the concentrations of the detectable species were generally of the same order of magnitude in the blank rapid leaching test and the distilled water leachant. The maximum variation in the concentrations of any species was by a factor of around 2 or 3, with the higher species concentration generally shown in the blank. The calcium and TOC were more then 2 times higher in the blank than the leachant, implying that the test method generated some cross-contamination of these species, potentially resulting from the leaching vessel and filter papers. However, the TIC was higher in the leachant, which may imply that the species was adsorbed during the leaching test. The zinc that was present in all of the samples probably originates from the lid of the Sterylin tubes used to store the samples [Cave, 1998].

The mean blank test data was used to quantitatively reduce the concentrations found in the leachates from the rapid leaching tests on the different materials, in order to account for those species that are, thus, deemed to result from the test method or leachant. Adjustments were made for conductivity and for all of the determinands listed in Table 3-11.

3.4.5.2 Test Repeatability

Test repeatability was investigated by performing two separate tests on the granite material (GR1 and GR2). Figure 3-7 shows the excellent correlation between the pH and conductivity of the leachates generated from the two tests. Figure 3-8a and b, show the average and range of concentrations of all species detected in the leachates from both phases of the test (L/S=2 (a) and L/S=2-10 (b)).

It can be seen from Figure 3-8a and b that no determinand varies by more than an order of magnitude and for most, the variation is considerably less. For most species the variation in the results from the two tests is less than 10% from the mean. The bromine, chlorine, barium manganese and lithium showed larger variations, in one or both phases of the test. However, in all cases the measured concentrations were close to the limits of analytical detection, where the accuracy of the data cannot be guaranteed. The average variation of all species that were present above the limits of analytical detection was 9.1% for the C2 phase of the test and 6.1% for C2-10 phase.

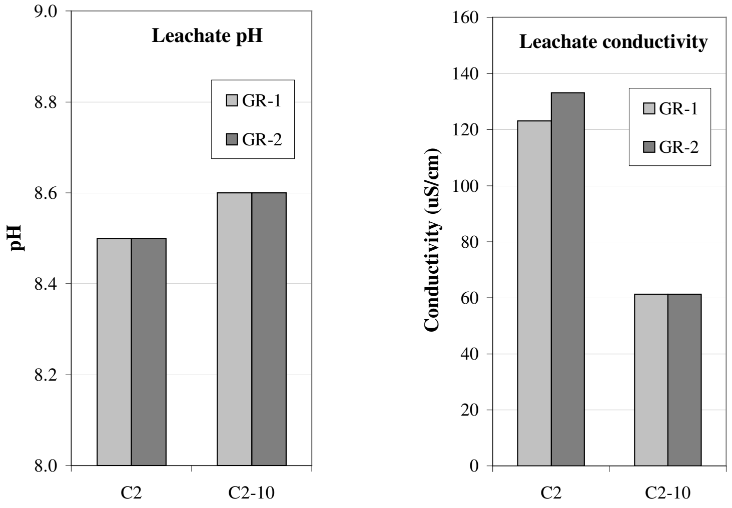


Figure 3-7: pH and conductivity data for the repeated rapid leaching characterisation tests on granite (C2 = L/S=2; C2-10 = L/S=2-10)

A number of species, which were analysed for, were below the limits of detection for the analytical methods that were used. This is particularly the case for the transition elements and such species are not shown in Figure 3-8a and b. The limits of analytical detection vary depending upon the species being detected and interference resulting from other species in solution. Generally, the limits of detection are in the order of 0.001 to 0.1 mg/l (or parts per million (ppm)).

A good correlation is shown between the repeat tests for the majority of species, however it should be pointed out that only two tests were performed and the variation observed between more numerous tests may potentially be greater [Harmonisation Network, 2000]. For species present at concentrations close to the limits of analytical detection, the data should be used cautiously and regarded only as semi-quantitative. The good degree of correlation between the repeat tests does, however, provide reassurance of the quality of the sample preparation and testing that was performed.

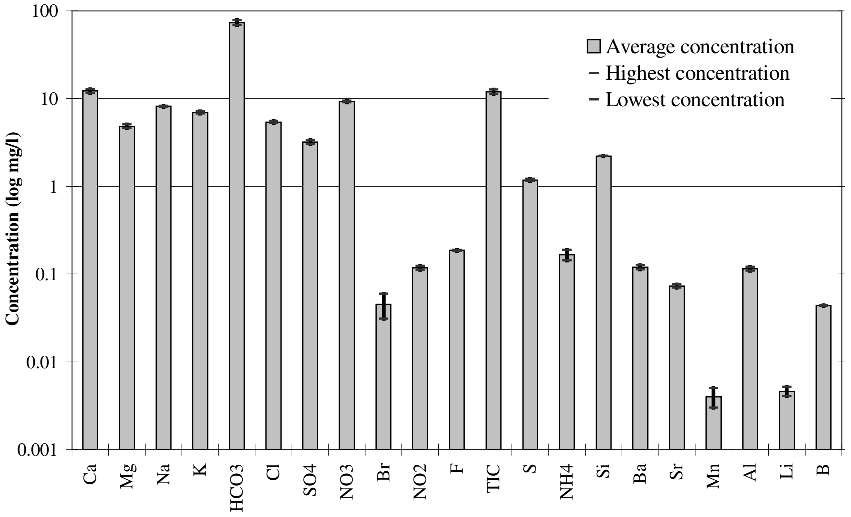


Figure 3-8a: Average and range of concentrations of determinands from the repeat rapid leaching characterisation tests on granite at L/S=2 (C2)

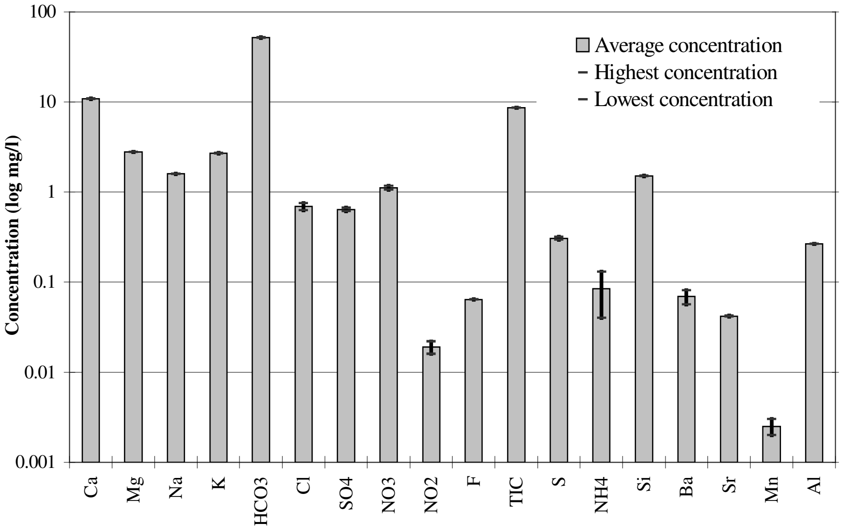


Figure 3-8b: Average and range of concentrations of determinands from the repeat rapid leaching characterisation tests on granite at L/S=2-10 (C2-10)

3.4.5.3 Results of the Rapid Leaching Characterisation Test

The chemical composition of the leachates from all of the rapid leaching characterisation tests were quantitatively adjusted to account for those species that resulted from the test method or leachant. A summary of the test data is given in Table 3-12a and b (for aggregates and binders respectively) where, for each test, the detected species are tabulated within specific concentration ranges. The data in Table 3-12a and b is quoted as a concentration, in milligrams per litre (mg/l), which is obtained directly from the analysis of the leachates (following the adjustment mentioned in earlier paragraphs). Any species that are listed in Table 3-9 but not in Table 3-12a and b may be present in the leachate but are at concentrations below the limits of analytical detection.

A graphical representation of the pH of all of the test leachates is given in Figure 3-9. Although presented on a linear scale, it should be remembered that pH is a logarithmic function, indicating that an order of magnitude change in the hydrogen ion (H^+) concentration will result in a change in pH of only 1. From Figure 3-9 it can be seen that, in general, the leachates fall in the neutral (~6-8) to alkaline (>8) range of pH. With the exception of the minestone (C2-10 leachate), all of the leachates are on the alkaline side of the average pH recorded in the blank tests (which were 4.7 in the C2 and 4.6 in the C2-10 leachates). For the majority of the materials, the pH of the leachates from the L/S=2 and L/S=2-10 phases of the test, vary by less than 1 unit of pH. This provides a good indication that the material under test is dictating the pH of the leaching environment.

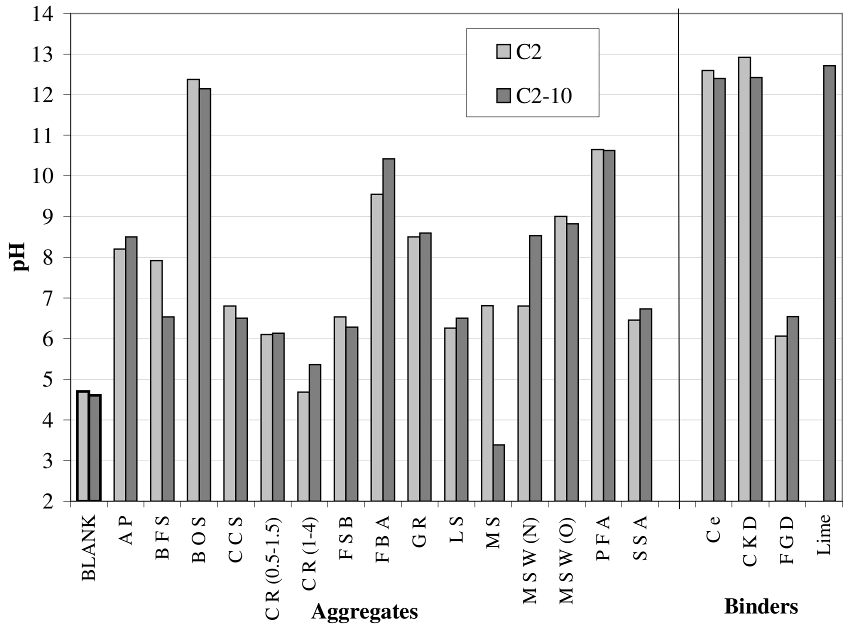


Figure 3-9: pH of the leachates from the rapid leaching characterisation tests (C2 = L/S=2; C2-10 = L/S=2-10)

Table 3-12a: Concentration ranges of determinands in the rapid leaching characterisation tests on aggregates

Material	L/S	pH	Cond. µS/cm	Concentration in leachate (mg/l)							
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**	**100-1000**	**1000-10000**	**>10000**
AP	**2**	8.2	290	TOC Fe Ni Mo Cd V Li	NO_2 NH_4 Mn Al Cu Zn	NO_3 Br F Ba Sr B	Mg Na K Si	Ca HCO_3 Cl SO_4 TIC S			
	2-10	8.5	85	NO_2 TOC Mn Fe Ni Cu Zn Mo Cd V Li	Ba Sr Cr B	F Al	Mg Na K Cl SO_4 TIC S Si	Ca HCO_3			
BFS	**2**	7.9	2567	Fe Ni Cu Zn Cr Cd V Se	NH_4 Mn	Ba Sr Al Li B	Mg F Si	Na K Cl TOC	Ca S	SO_4	
	2-10	6.5	386	Mn Fe Ni Cu Zn Cr Cd V Se	Ba Li	F TIC NH_4 Sr Al B	Mg Na K Cl Si	Ca TOC S	SO_4		
BOS	**2**	12.4	7937	Mn Fe Ni Cu Cr Cd V Li As Se	Br NH_4 Zn B	NO_2 F TIC Si Ba Al	Na K Cl SO_4 NO_3 Sr	TOC S	Ca	HCO_3	
	2-10	12.2	7501	Mn Fe Ni Cu Zn Cr Cd Pb V Li As Se	Ba	Na K NO_3 NO_2 F Si Sr Al	Cl SO_4 TIC S	TOC	Ca	HCO_3	
CCS	**2**	6.8	97	Mn Fe Ni Cu Cr Cd Pb V Li As Se	NO_2 NH_4 Ba Sr Zn B	Mg F P Al	Na K Cl SO_4 TIC S Si	Ca TOC			
	2-10	6.5	64	Mn Fe Ni Cu Zn Cr Cd Pb V Li As Se	NO_2 Ba Sr B	Mg Na Cl SO_4 F S Al	K TIC Si	Ca TOC			
CR (0.5-1.5)	**2**	6.1	189	Ni Cu Cr Cd Pb V Li As Se	Ba Sr Fe Co	F P Si Mn B	Mg Na K Cl Br S NH_4 Zn	Ca HCO_3 SO_4 TOC TIC			
	2-10	6.1	55	Fe Ni Cu Cr Cd Pb V Li As Se	F Ba Sr Mn	Mg Na K Cl NO_2 Si B	Ca SO_4 Br TIC NH_4 Zn	HCO_3 TOC S			
CR (1-4)	**2**	4.7	38	Si Ni Cu Cr Cd Pb V Li As Se	NO_2 Ba Sr Mn Fe	Mg Na K Cl F	Ca SO_4 Br TIC S NH_4 Zn	HCO_3 TOC			
	2-10	5.4	9	Na Si Ba Sr Fe Ni Cu Cr Cd Pb V Li As Se	Cl F Mn	Mg SO_4 S NH_4	Ca TIC Zn	TOC			
FSB	**2**	6.5	1887	Ni Cr Cd Pb Li As Se	Sr Mn Cu V B	Mg NO_2 F P NH_4 Ba Fe Al Zn	Ca Cl S Si	Na SO_4 TIC	K TOC		
	2-10	6.3	332	Sr Mn Ni Cu Cr Cd Pb V Li As Se	NO_2 NH_4 Ba Zn	Ca Mg Cl F P Fe Al	Na SO_4 S Si	TOC TIC	K HCO_3		
FBA	**2**	9.6	814	Mn Ni Zn Cd Se	As	F NH_4 Ba Sr Li	NO_3 NO_2 TIC Si Al B	Na K HCO_3 Cl TOC S	Ca SO_4		
	2-10	10.4	390	Mn Fe Ni Cu Zn Cr Cd Pb Se	NH_4 V Li As	Mg NO_3 F Ba Sr B	Na K Cl TIC Si Al	Ca HCO_3 SO_4 TOC S			
FBA	**2**	9.6	814	Mn Ni Zn Cd Se	As	F NH_4 Ba Sr Li	NO_3 NO_2 TIC Si Al B	Na K HCO_3 Cl TOC S	Ca SO_4		
	2-10	10.4	390	Mn Fe Ni Cu Zn Cr Cd Pb Se	NH_4 V Li As	Mg NO_3 F Ba Sr B	Na K Cl TIC Si Al	Ca HCO_3 SO_4 TOC S			

Table 3-12a: Concentration ranges of determinands in the rapid leaching characterisation tests on aggregates (continued)

Material	**L/S**	**pH**	**Cond. µS/cm**	**Concentration in leachate (mg/l)**							
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**	**100-1000**	**1000-10000**	**>10000**
GR	**2**	8.5	123	TOC Mn Fe Co Ni Cu Zn Mo Cd V Li	Br Sr B	NO_2 F NH_4 Ba Al	Mg Na K Cl SO_4 NO_3 S Si	Ca HCO_3 TIC			
	2-10	8.6	61	TOC S Mn Fe Co Ni Cu Zn Mo Cd V Li	NO_2 F Ba Sr	Cl SO_4 Al	Mg Na K NO_3 TIC Si	Ca HCO_3			
LS	**2**	6.3	149	Mn Fe Ni Cu Cr Cd Pb V Li Se	Br NH_4 Zn B	K Si Ba Sr	Mg Na Cl SO_4 NO_3 F TIC S	Ca TOC			
	2-10	6.5	74	Mn Fe Ni Cu Zn Cr Cd V Li Se	Al	Mg Na NO_2 NH_4 Si Ba Sr	Cl SO_4 NO_3 F TOC TIC S	Ca HCO_3			
MS	**2**	6.8	3657	Fe Cu Cr Cd V Se	Ba Mn Al Zn Li	F Sr B	Mg Br TIC NH_4 Si	Ca K HCO_3 TOC	Na Cl SO_4 S		
	2-10	3.4	672	Mn Cr Cd V Li Se	Sr Fe Cu	Mg Br F NH_4 Ba Al Zn B	Ca K Si	SO_4 S	Na Cl TOC		
MSW(N)	**2**	6.8	5167	Cr Cd V Se	Mn Fe Ni Zn Li	NO_2 F NH_4 Si Ba Mo	Br TIC Sr Al Cu B	Mg P	Ca Na K Cl TOC S	SO_4	
	2-10	8.5	1414	Mn Ni Cr Cd Pb V Li Se	Si Fe Zn Mo	Br NO_2 F NH_4 Ba Sr Cu B	Mg TIC P	Na K HCO_3 TOC Al	Ca Cl SO_4 S		
MSW(O)	**2**	9.0	27157	Mn Fe Zn Cr Cd V	Ni Li Se	Mg F Si Ba Cu Mo B	NO_2 TIC P Sr	HCO_3 Br NH_4 Al	TOC	Ca Na K Cl SO_4 S	
	2-10	8.8	5191	Mn Fe Ni Zn Cr Cd Pb V Li Se	Si Ba Mo	Mg NO_2 F P Cu B	Br TIC NH_4 Sr	HCO_3 TOC Al	Ca Na K Cl S	SO_4	
PFA	**2**	10.7	2557	Ba Mn Ni Zn	As Se	NO_2 F NH_4 Sr Cr V	Br TIC Si Al Mo Li B	Ca Cl TOC	Na K HCO_3 SO_4 S		
	2-10	10.6	669	Mn Fe Ni Cu Cd	Mg Zn Cr As Se	NO_2 F NH_4 Ba Sr Mo V	Cl Br TIC Si Al Li B	Ca Na K HCO_3 TOC S	SO_4		
SSA	**2**	6.5	2677	Fe Ni Cu Cr Cd Pb	Mn Zn V As Se	P NH_4 Ba Sr Al Li	F TIC Si Mo B	Mg Na K HCO_3 Cl Br TOC	Ca S	SO_4	
	2-10	6.7	1099	Ba Fe Ni Cu Zn Cr Cd	NH_4 Mn V Li As Se	P Si Sr Al Mo B	Na K Cl Br F TOC TIC	Mg	Ca SO_4 S		

Table 3-12b: Concentration ranges of determinands in the leachates from the rapid leaching characterisation tests on binders

Material	**L/S**	**pH**	**Cond. µS/cm**	**Concentration in leachate (mg/l)**							
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**	**100-1000**	**1000-10000**	**>10000**
Ce	**2**	12.6	21557	Mn Ni Zn As	NH_4 Si Se	NO_2 Ba Cr	Br F TIC Li	Cl TOC Sr	Ca Na S	K HCO_3 SO_4	
	2-10	12.4	11681	Mn Fe Ni Zn Cd Pb V As Se	Cr	Si Al	Cl SO_4 F TIC Ba Li	Na TOC S Sr	K	Ca HCO_3	
CKD	**2**	12.9	56057	Ni Cd Pb	As	NH_4 Cu Se	NO_2 Ba Fe Al Cr Li	F TOC TIC Sr Zn	Ca Na Br	HCO_3 Cl SO_4 S	K
	2-10	12.4	15331	Ni Zn As Se		NH_4 Si Ba Cr	F TIC Sr Li	Na Br TOC	Ca Cl S	K HCO_3 SO_4	
FGD	**2**	6.1	2267	Fe Ni Cu Cr Cd V Li As Se	Ba Zn Pb	K Br NO_2 Si Sr Mn Al B	Mg Na F TIC	Cl TOC	Ca S	SO_4	
	2-10	6.5	2171	Ba Fe Ni Cu Zn Cr Cd Pb V Li As Se	Mn	Mg Na Si Sr Al B	Cl F TOC TIC		Ca S	SO_4	
Lime	**2**										
	2-10	12.7	9141	Si Mn Fe Ni Cu Zn Cd Pb V As Se	Br NO_2 Cr Li	Cl F Ba Al B	Na K SO_4 TIC	TOC S Sr	Ca	HCO_3	

The furnace bottom ash and MSW incinerator ash (new) leachates exhibit a more pronounced increase in pH, whilst the blastfurnace slag and minestone show a significant decrease. In theory, to maintain a constant pH between the two phases of testing the H^+ concentration must remain constant, which, for this test, requires a five-fold increase in the absolute number of H^+ ions in solution. Any dilution of the H^+ ions will result in an increase in pH, although diluting by a factor of 5 would only result in an increase in pH of 0.7. This is however theoretical and in practice the ionic capacity of many solutions also enables H^+ ions to be taken out of solution, with resultant changes in pH.

The alkaline leachates generated by the majority of materials may provide a natural hindrance to the release of certain species into solution.

Baldwin et al. (1997) performed a series of compliance leaching tests on a range of alternative materials. They also reported some increasing and some decreasing values of pH covering a range from 7.43 to 12.75 for equivalent materials. In general, the pH values obtained by Baldwin et al. (1997) were higher (by 1 to 2) than those reported in Table 3-12a and b, for similar materials. It may be possible to explain this variation by considering the pH of the leachant used in the tests, for which the procedure specifies a range of between 5 and 7. However, Table 3-11 reports the use of leachant with a pH of 5.2 and therefore if Baldwin et al. used a leachant with a pH closer to 7, then this may explain some of the variation.

A graphical representation of the conductivity of the test leachates is shown in Figure 3-10. The conductivity of a solution is a measure of its ability to allow the passage of an electric current and is related to the concentration of dissolved ions. The materials reveal leachate conductivity values that range over 4 orders of magnitude. The cement, cement kiln dust and the MSW incinerator ash (old) display the highest conductivity values ($\sim 10^4 \mu S/cm$) whilst the asphalt planings, china clay sand, crumbed rubber, granite and limestone show the lowest values ($\sim 10^2 \mu S/cm$ or less). From Table 3-12a and b, it can be seen that the materials with the highest conductivity are those with the most chemical species in the higher concentration ranges. Similarly, those materials showing the lowest conductivity also contain the lowest concentrations of those species that are most abundant in that leachate. The leachates from the different materials that were tested, all (with the exception of basic oxygen steel slag and flue-gas desulphurisation gypsum) show a significant (half to 1 order of magnitude or more) reduction in the conductivity measured between the L/S=2 and L/S=2-10 phases of testing. The basic oxygen steel slag and flue-gas desulphurisation gypsum showed only a minimal decline in measured conductivity, which may indicate that at a L/S=2 some species contributing to the conductivity were limited by solubility. In general the leachates with a high conductivity were also those that had a high pH and vice versa. This phenomenon may be due to the bases and salts of the alkali and alkali earth metals that contribute significantly to both an alkaline pH and increased conductivity.

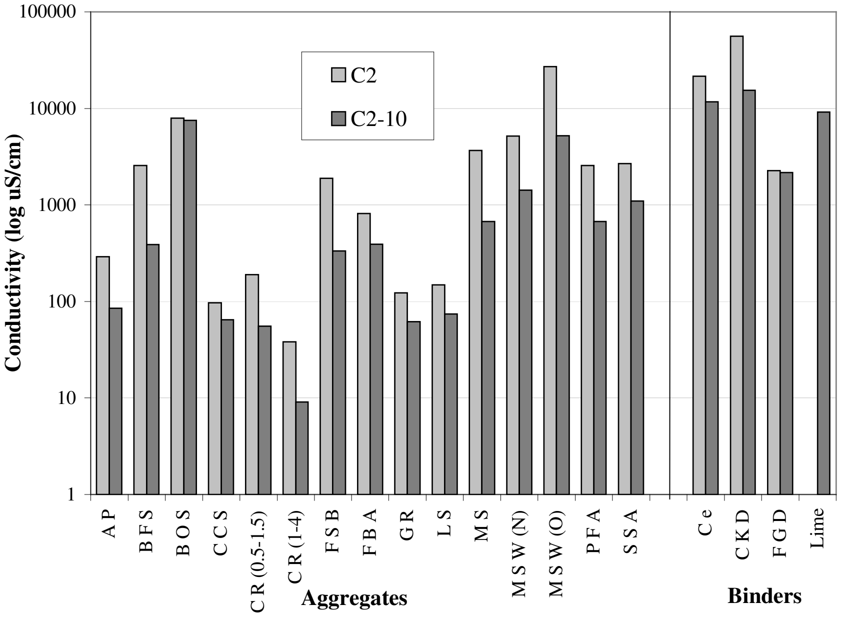


Figure 3-10: Conductivity of the leachates from the rapid leaching characterisation tests (C2 = L/S=2; C2-10 = L/S=2-10)

Table 3-12a and b present the concentration ranges of the different species found in the test leachates. It can be clearly seen that the concentrations of the various components of the leachate were detected over more than 6 orders of magnitude from over 10,000mg/l to less than 0.01mg/l. The elements that were present in the highest concentrations were sodium, potassium, calcium, chlorine and sulphur. The compounds of HCO_3 (a measure of alkalinity) and SO_4 (sulphate) are also present in relatively high concentrations in many of the leachates.

In the majority of the leachates the trace elements are present in concentrations less than 1mg/l, with most falling in the <0.01 range or below the limits of detection. Zinc is present in the 1-10mg/l range for the crumbed rubber samples and reaches 13.2mg/l in the cement kiln dust C2 leachate. This cement kiln dust leachate also contains chromium in the 1-10mg/l range. Molybdenum is found at concentrations at the lower end of the 1-10mg/l range in the C2 leachate for the pulverized-fuel ash and sewage sludge incinerator ash and copper is found in the leachate from the MSW incinerator ash (new).

Between the two phases of leaching, the concentration of the different species in the leachates, generally, remains within the same concentration range or decreases by an order of magnitude. A decrease in the order of magnitude would imply that leaching occurred rapidly in the first phase of the test. Similar species concentrations imply that either leaching continues throughout the duration of the test and is rate limited, or that it is limited by solubility. Occasionally the concentration of a species increases in the second phase of the leaching, which implies that a change in the leaching conditions has enabled the leached quantity to increase.

3.4.6 Discussion

3.4.6.1 Normalisation of the Rapid Leaching Characterisation Test Data

The results from the rapid leaching characterisation have shown the wide and varied range of species concentrations that might be expected in the leachates from different materials. The concentration of the different chemical species in the solution does not, however, give a sufficient indication of their potential to cause harm. As described in Section 3.3.3, in relation to the solid composition of the materials, the risk associated with the presence of a particular species is not directly related to absolute concentrations. The concentration at which, say, cadmium may cause harm would be significantly lower than that of, say, calcium. It is the toxicity of a species at the measured concentration that is of most important for environmental protection. To enable the variable elemental toxicity to be included in the discussion of the rapid leaching characterisation data, the leachate composition data has been compared, arbitrarily, to published acceptance criteria for water and environmental quality. It is not, however, suggested that such criteria should form the basis of any assessment of material suitability because the leachates under comparison are the result of intense testing of materials in an artificial condition. The site-specific nature of the material utilisation environment is likely to be the deciding factor for the regulator setting the specific discharge limits, which may be more or less stringent than the quality standards, used here for comparison. The normalisation against such criteria is used here purely as a tool for interpretation of the leaching test data to give a measure of the relative toxicity potential of the different leachates.

In this instance the comparison has been made, arbitrarily, to the water quality maxima summarised by Baldwin et al. (1997) from UK drinking and surface water quality standards and other commonly used environmental standards. Table 3-13 provides a summary of these criteria. It should be noted that since the commencement of this research, a number of the UK Drinking Water Quality Standards have been updated and revised as a result of an amendment to the Water Supply (Water Quality) Regulations [SI 2000/3184, 2000]. Notably, reductions in the drinking water quality standards have been introduced for chloride, nickel, copper, lead, boron and arsenic, whilst and the permitted level of sodium has increased.

The results of the comparison are listed in Table 3-14a for the aggregates and Table 3-14b for the binders. The different chemical species have been categorised according to the degree to which they exceed (or not) the water quality maxima and also by whether the species is a List I, List II or non-listed substance [76/464/EEC, 1976 - See Appendix A]. This segregation of the List I and II species was used to identify those species that may potentially cause harm, from those for which the standards apply a secondary or aesthetic quality. Any of the species that are not listed in Table 3-14a and b may exist at concentrations above or below the appropriate water quality maxima, but because the limits of analytical detection are relatively high, this cannot be assessed either way.

It can be seen from Table 3-14a and b, that in most leachates the majority of species were present in concentrations below the water quality maxima. However, all of the leachates (except those from the asphalt planings) contain at least one species at concentrations of 1 to 10 times the water quality maxima. Even the leachates from the primary limestone and granite aggregates contain species in excess of the criteria. Approximately half of the materials produce leachates with species concentrations between 10 and 100 times the water quality maxima and for a few materials this factor is greater than 100 for potassium and/or aluminium. However, neither potassium nor aluminium is a listed species and the water quality maxima are generally in place for secondary or aesthetic reasons. Other non-listed species that exceeded the water quality maxima by more than 10 times are sodium, chlorine, iron, SO_4 and NO_2.

Table 3-13: Arbitrary water quality maxima used for normalisation of the concentration of chemical species in the leachates from the rapid leaching characterisation test

Chemical Species	List I or II substance	Units	Water quality maxima
pH	-	pH units	5.5 to 9.5
Conductivity	-	uS/cm	1500
Ca	-	mg/l	250
Mg	-	mg/l	50
Na	-	mg/l	150
K	-	mg/l	12
Total Alk (HCO_3)	-	mg/l	30(minimum)
Cl	-	mg/l	400
Sulphate (SO_4)	-	mg/l	250
Nitrate (NO_3)	-	mg/l	50
Nitrite (NO_2)	-	mg/l	0.1
F	II	mg/l	1.5
Total P	II	mg/l	2.2
Ammonium (NH_4)	II	mg/l	0.5
Ba	II	mg/l	1
Mn	-	mg/l	0.05
Total Fe	-	mg/l	0.2
Al	-	mg/l	0.2
Ni	II	mg/l	0.05
Cu	II	mg/l	3
Zn	II	mg/l	5
Cr	II	mg/l	0.05
Mo	II	mg/l	0.07
Cd	I	mg/l	0.005
Pb	II	mg/l	0.05
V	II	mg/l	0.02
Li	-	mg/l	25
B	-	mg/l	2
As	II	mg/l	0.05
Se	II	mg/l	0.01

Table 3-14a: Normalisation of rapid leaching characterisation data from the aggregates to arbitrary water quality maxima

Aggregate	L/S	Normalised concentration in leachate					
		<1	1-10		10-100		>100
			II	Other	II	Other	
AP	**2**	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Al Ni Cu Zn Cr Mo Pb V Li B As Se					
	2-10	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Al Ni Cu Zn Cr Mo Pb V Li B As Se					
BFS	**2**	Mg Na Cl NO_3 P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B As Se	F	Ca K SO_4 Al			
	2-10	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd V Li B As Se		Al			
BOS	**2**	Mg Na K Cl SO_4 NO_3 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd V Li B As Se		Ca NO_2 Al			
	2-10	Mg Na K Cl SO_4 NO_3 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb V Lime B As Se		Ca NO_2 Al			
CCS	**2**	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B As Se		Al			
	2-10	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Al Ni Cu Zn Cr Mo Cd Pb V Li B As Se					
CR (0.5-1.5)	**2**	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P Ba Fe Al Ni Cu Zn Cr Mo Cd Pb V Li B As Se		Mn	NH_4		
	2-10	Ca Mg Na K Cl SO_4 NO_3 F P Ba Fe Al Ni Cu Zn Cr Mo Cd Pb V Li B As Se	NH_4	Mn NO_2			
CR (1-4)	**2**	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P Ba Mn Fe Al Ni Cu Zn Cr Mo Cd Pb V Li B As Se	NH_4				
	2-10	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P Ba Mn Fe Al Ni Cu Cr Mo Cd Pb V Li B As Se	Zn NH_4				
FSB	**2**	Ca Mg Na Cl SO_4 NO_3 F P NH_4 Ba Mn Ni Cu Zn Cr Mo Cd Pb Li B As Se	V	Fe Al NO_2		K	
	2-10	Ca Mg Na Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B As Se		K Al			
FBA	**2**	Ca Mg Na Cl NO_3 F P NH_4 Ba Mn Fe Ni Cu Zn Cd Li B As Se		K SO_4		NO_2 Al	
	2-10	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb Li B As Se	V			Al	
GR	**2**	Ca Mg Na K Cl SO_4 NO_3 F P NH_4 Ba Mn Fe Al Ni Cu Zn Cr Mo Pb V Li B As Se		NO_2			
	2-10	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Pb V Li B As Se		Al			
LS	**2**	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Al Ni Cu Zn Cr Mo Cd Pb V Li B Se	As				
	2-10	Ca Mg Na K Cl SO_4 NO_3 F P NH_4 Ba Mn Fe Al Ni Cu Zn Cr Mo Cd V Li B Se	As	NO_2			
MS	**2**	Ca Mg NO_3 F P Ba Fe Al Ni Cu Zn Cr Mo Cd Pb V Li B As Se	NH_4	Na K Cl SO_4 Mn			
	2-10	Ca Mg Na K Cl SO_4 NO_3 NO_2 F P Ba Mn Fe Al Ni Cu Zn Cr Mo Cd Pb V Li B As Se	NH_4				
MSW (N)	**2**	Mg NO_3 F Ba Mn Fe Ni Zn Cr Cd Pb V Li B As Se	P Cu Mo NH_4	Ca Na Cl SO_4 NO_2		K Al	
	2-10	Mg Na Cl NO_3 F NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B Se	P As	Ca K SO_4 NO_2		Al	

Table 3-14a: Normalisation of rapid leaching characterisation data from the aggregates to arbitrary water quality maxima (continued)

Aggregate	L/S	Normalised concentration in leachate					
		<1	1-10		10-100		>100
			II	Other	II	Other	
MSW (O)	**2**	Mg NO_3 F Ba Mn Fe Ni Cu Zn Cr Cd V Li B Se	P Mo	Ca	NH_4	Na Cl SO_4 NO_2	K Al
	2-10	Mg NO_3 F P Ba Mn Fe Ni Cu Zn Cr Cd Pb V Li B Se	Mo As NH_4	Ca Na Cl SO_4 NO_2		K Al	
PFA	**2**	Ca Mg Cl NO_3 F P NH_4 Ba Mn Fe Ni Cu Zn Li As Se	Cr	NO_2 Na SO_4 B	Mo V	K Al	
	2-10	Ca Mg Na Cl SO_4 NO_3 F P Ba Mn Fe Ni Cu Zn Cd Li B As Se	Cr Mo V NH_4	NO_2 K		Al	
SSA	**2**	Na Cl NO_3 NO_2 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Cd Pb Li B As Se	V	Ca Mg K SO_4 Al	Mo		
	2-10	Ca Mg Na K Cl NO_3 NO_2 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Cd Pb Li B As Se	Mo V	SO_4 Al			

Table 3-14b: Normalisation of rapid leaching characterisation data from the binders and activators to arbitrary water quality maxima

Binder	L/S	Normalised concentration in leachate					
		<1	1-10		10-100		>100
			II	Other	II	Other	
ce	**2**	Mg Cl NO_3 F P NH_4 Ba Mn Fe Al Ni Cu Zn Li B As Se		Ca Na SO_4 NO_2	Cr		K
	2-10	Mg Na Cl SO_4 NO_3 F P NH_4 Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B As Se	Ba	Ca Al		K	
ckd	**2**	Mg NO_3 Ni Cu Cd Pb Li As Se	F Ba Zn NH_4	Ca Na Cl	Cr	SO_4 NO_2 Al Fe	K >1000x
	2-10	Mg Na NO_3 P NH_4 Ba Mn Fe Al Ni Cu Zn Li B As Se	F Cr	Ca Cl SO_4			K
fgd	**2**	Mg Na K Cl NO_3 P NH_4 Ba Fe Al Ni Cu Zn Cr Mo Cd Pb V Li B As Se	F	Ca SO_4 Mn NO_2			
	2-10	Mg Na K Cl NO_3 NO_2 P NH_4 Ba Fe Al Ni Cu Zn Cr Mo Cd V Li B As Se	F	Ca SO_4 Mn			
li	**2**						
	2-10	Mg Na K Cl SO_4 NO_3 NO_2 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B As		Ca Al			

II = List II substance [EEC directive 76/464/EEC, 1976]; Other = Not List I or List II

The only List I substance for which the comparison was carried out, was cadmium, but none of the leachates were found to contained concentrations above the water quality maxima. This was even the case for the basic oxygen steel slag, MSW incinerator ash (old) and limestone that all contained concentrations in the solid that exceeded the normalising criteria by more than 10 times. List II substances were however found at concentrations above the water quality maxima in the leachates from all of the materials except the asphalt planings, basic oxygen steel slag, china clay sand and granite. This included the leachates from the limestone for which the concentration of arsenic was approximately twice the water quality maxima. The cement, cement kiln dust, crumbed rubber (0.5-1.5mm), MSW incinerator ash (old), pulverized-fuel ash and sewage sludge incinerator ash leachates contained List II substances (one or more from chromium, molybdenum, vanadium, lead and ammonium) at between 10 and 100 times the water quality maxima.

The fact that most of the leachates contain species at concentrations above those of the water quality maxima might reinforce the fears of the wary over the safety of using such materials in road construction applications. However, in this instance, such a comparison would have concluded that the asphalt planings is the only acceptable material and that the limestone and granite primary aggregates should be rejected on environmental grounds. This conclusion would be inappropriate as described in Section 3.3.3. Again this highlights the extensive limitations of using any such comparisons as acceptance criteria (in a regulatory sense), especially when the leaching data is obtained from intensive, non-simulative testing regimes.

Another point to note here relates to the leaching of the asphalt planings, which is a material consisting of a primary aggregate that has previously been modified, by binding with bitumen, for use in a pavement. Such a typical modification has transformed the leaching performance, as assessed in the simple characterisation test and it is likely, therefore, that modifications to behaviour will be consequent upon the mode of use of the material. Further investigation of the effect of binders on modifying leaching behaviour is included in Chapter 6 of this thesis.

Baldwin et al. (1997) carried out a similar comparison of their compliance leaching tests to the defined water quality maxima. The degree of correlation between their results and those listed in Table 3-14a and b for equivalent materials is variable. In general, most species listed above the water quality maxima in Table 3-14a and b were also detected at concentrations above the water quality maxima by Baldwin et al.. However, Baldwin et al. often detected more species, particularly the minor elements, above the water quality maxima and species were frequently present in higher concentrations. It is not possible to conclude whether this variation is the result of the test procedure or material composition, although, it may result from the inherent variability of such materials or the source and degree of weathering to which the materials have been exposed. Another, equally possible, explanation is that which was discussed earlier (as proposed by the Harmonisation Network (2000)) that concluded that significant variability may occur depending upon the particular equipment or methods used, even if they are within the scope of the specification. Such variations raise some concerns about the characterisation of, so called, "typical" material sources, and the reliability of the simulations presented by leaching tests.

The species with a normalised concentration in the leachates greater than unity, were compared to those listed in Table 3-7a and b in which the normalised solids concentrations were listed. The majority of species that were present at normalised solids concentrations greater than unity were below the water quality maxima in the test leachates. The species that exceeded unity in both considerations were molybdenum, chromium and barium. It should also be noted that the criteria for the comparison of the solid composition focuses principally on the concentration of trace elements and not the full range of species presented by the water quality maxima.

The normalisation of the concentration of species in the test leachates to the water quality maxima enabled some identification of the species that could potentially be of concern to the regulator, as listed in Table 3-14a and b. In addition to identifying the individual species of possible consequence, a crude approximation of the overall leachability of each material was also made. Continuing from the determination of the "toxicity index" for the solid composition as described in Section 3.3.3 (from Scott et al. (1995)), the method was applied to the species concentrations in the leachates normalised to the water quality maxima. Two measures of total leachability were determined. Firstly the total of all of the normalised concentrations was summed to give a total "toxicity index". To understand which species contributed most significantly to these totals, the normalised concentrations of the species were summed for the species in the different groups of the periodic table, for which the leaching patterns of the individual species were generally similar. This summation gave a good indication about which species contributed most significantly to the overall total, but did not give any weighting to those species for which the water quality maxima reflects species toxicity, compared to those that impart aesthetic or secondary properties. It was reasoned that the water quality maxima for the List I and II species (that are of most importance to the regulator) were deduced from the toxicity of each species to humans, animals, fish or the environment in general. Therefore, a summation of the normalised concentrations of these species was selected as a secondary indicator of the relative potential toxicity of the different materials.

Again, the maximum and minimum concentrations (i.e. 0.1 and 0 for a concentration of <0.1) were considered in the summation but, as with the solids comparison, had little effect on the relative "scores" of each material. Limited variation in material ranking occurred between the two phases of the test. Therefore, Table 3-15 summarises both indicators of overall toxicity, based on maximum possible concentrations in the L/S=10 phase of the test. It should be reinforced, however, that such toxicity indices should not be used as a means of assessing the acceptability, or not, of any material for use, but, are used here as a tool for simplifying a complex data set. It should also be noted here that the "toxicity index" for the total of all species is based upon the sum of 26 normalised species concentrations. The equivalent number of data points for the List I and II species summation is 13. Therefore, a "toxicity index" of 26 or 13 respectively, may not indicate that any species were actually above the water quality maxima.

It can be seen from Table 3-15 that the contribution of the List I and II species to the total toxicity is usually less than 50% and in most cases significantly less. It is interesting to note the comparative position of the limestone, which shows similar index values to a notable proportion of the alternative aggregates. It is also of interest to compare the data in Table 3-15 to that in Table 3-8, which equivalently ranked the materials based on their solid composition. Although many materials retained a similar ranking, there are a number of notable exceptions, namely, the cement kiln dust, cement and flue-gas desulphurisation gypsum that show more significant leaching, and the crumbed rubber and asphalt planings that show lesser leaching. It may be possible to explain the former of these observations by the very small size of the particles in these materials, which dictates a high surface area to volume ratio and thus a resultantly high degree of surface wash-off. A feasible explanation for the relatively minor leaching from the crumbed rubber and, particularly, the asphalt planings is that the assessment of the solid composition included an assessment of the PAH content, of which both of these materials showed significant quantities. The leaching of PAH's was not including in the analysis of the leachates from the rapid leaching characterisation tests because the test generated an insufficient volume of leachate for the necessary analytical procedure. The inclusion of such organic measures in the assessment may therefore have increased the apparent toxicity of such materials.

Table 3-15: 'Toxicity index' of the rapid leaching characterisation test data (L/S=10) for the aggregates and binders (summation of the maximum concentrations)

Rank	Material	Toxicity Index
		All species
1	CKD*	259
2	MSW(O)	170
3	MSW(N)	89
4	PFA	81
5	Ce*	42
6	FBA	35
7	SSA	25
8	FGD*	24
9	FSB	16
10	BFS	14
11	**LS**	**13**
12	MS	12
13	CR(0.5-1.5mm)	11
14	BOS	11
15	Lime*	10
16	CR(1-4mm)	6.3
17	**GR**	**5.9**
18	CCS	5.6
19	AP	5.5

Material	Toxicity Index
	List I and II species
CKD*	46
PFA	17
SSA	13
Ce*	10
MSW(O)	8.0
CR(0.5-1.5mm)	7.3
MS	6.7
LS	**6.3**
BFS	6.2
MSW(N)	5.8
FGD*	5.3
CR(1-4mm)	4.7
FBA	4.2
AP	3.8
GR	**3.8**
Lime*	3.7
BOS	3.2
FSB	2.9
CCS	2.7

* = binder or activator, **bold** = primary aggregate

Note: Such rankings should not be used as a means of accepting or rejecting a material for usage

3.4.6.2 Identification of Leaching Behaviour

The discussion on the rapid leaching characterisation tests has so far been carried out using data presented as concentrations of the different species in the leachate.

Although it is the species concentration that is usually of importance in terms of environmental loading, determining the quantity of each species that has been leached from the solid can aid the understanding of the leaching process. To determine the quantity (in milligrams per kilogram, mg/kg) of each species that was leached during the two phases of the rapid leaching test, Equation 3-2 and Equation 3-3 were applied.

$$A_2 = C_2 \times \frac{(L_2 + RM)}{M_{DW}}$$

Equation 3-2 [CEN, 1996]

Where,

A_2 = leached quantity of the chemical species at L/S=2 (mg/kg of dry matter)

C_2 = concentration of the chemical species in the leachate at L/S=2 (mg/l)

L_2 = volume of leachant used in L/S=2 (ml)

RM = residual moisture in the tested material (ml): $RM = MC \times M_{DW}$

- MC = moisture content of the tested material (% of dry weight)
- M_{DW} = dry mass of the tested material (g)

$$A_{2-10} = \frac{[(L_{2A} \times C_2) + (L_{2-10} + L_2 + RM - L_{2A}) \times C_{2-10}]}{M_{DW}}$$

Equation 3-3 [CEN, 1996]

Where, additionally,

A_{2-10} = leached quantity of the chemical species at L/S=10 (mg/kg of dry matter)

C_{2-10} = concentration of the chemical species in the leachate at L/S=10 (mg/l)

L_{2-10} = volume of leachant added in L/S=10 (ml)

L_{2A} = volume of leachate removed at C2 (ml)

In the compliance test procedure [CEN, 1996], it is proposed that the leaching behaviour of different species can be identified by comparing the quantity of a determinand released during consecutive phases of the test. The so-called "Q" values are determined from Equation 3-4.

$$Q = \frac{A_{2-10}}{A_2}$$

Equation 3-4 [CEN, 1996]

Where, A_2 and $A_{2\text{-}10}$ are as previously defined.

The interpretation of the "Q" value is still under debate by CEN TC292, however, the procedure defined by CEN (1996) proposes that:

1) If Q is low (e.g. $Q \leq 2$), the species is either largely washed out in L/S=2 (i.e. very mobile) or the matrix has an extremely high degree of retention (i.e. low mobility);
2) If Q is high (e.g. $Q > 6$), the leachability increases substantially after the leaching (and subsequent dilution at L/S=2-10) of solubility controlling constituents or major changes in leachability controlling conditions (i.e. pH);
3) If Q is intermediate (e.g. $2 < Q < 6$), the leachability is largely solubility controlled and the behaviour is reasonably well defined.

It should be noted that the calculation and assessment of the results of this test using the Q-value approach does not form part of the now published CEN compliance test (BS EN 12457, 2002) and thus the assessment presented here should be used with caution.

Such an interpretation of the Q values provides a useful tool for the simplistic understanding of leaching behaviour. However, such an approach could lead to significant inaccuracies in interpretation because it does not take account of the complexities of the leaching process. This interpretation does not allow any identification of species precipitation and/or the reduction of species concentration in solution by other mechanisms. Theoretically, such processes could be identified for those species with a Q value less than 1. Such an interpretation may indicate that leaching is solubility limited during the L/S=2-10 phase of the test and give further information as to what changes have occurred in the leaching environment.

Perhaps more importantly, the Q value classification does not fully account for the variation in species solubility with pH which may, potentially, be very significant. In theory, leaching controlled purely by solubility constraints would result in a Q value of 5. The concentration of species in the leachates from the two phases of the test would be equal, and thus the Q value would simply reflect the ratio of the volumes of leachate (i.e. L/S from 2 to 10). However, this may often not be the case as it was shown from Figure 3-9 that the leachate pH sometimes varies between the two phases of the test resulting in a change of species solubility. Although the range of Q values which define solubility controlled leaching are between 2 to 6, this may only absorb some of the variations resulting from such changing solubility. Figure 3-11 shows typical curves of pH verses solubility for the leaching of zinc and molybdenum from pulverized-fuel ash [Kossen and van der Sloot, 1997]. Similar patterns have been shown to exist in many materials and also for other species [van der Sloot et al. 1997]. For illustration purposes, if the leachate pH changed from 13 to 10 between the two phases of the test, the resulting Q values, for solubility controlled leaching, would be 0.5 for zinc and 100 for molybdenum. Similarly, a pH change from 8 to 6 would result in Q values of approximately 250 and 1.4. Such Q values would not indicate to the reader, that the leaching was solubility controlled. More generally, Conner (1990) presented simplified solubility curves for a range of metal hydroxides. In the pH range of 7 to 12 (which is typical of the leachates obtained from the tested materials), the solubility of many of the species varied by about an order of magnitude for each unit of pH. Such changes in solubility would lead to Q values ranging from 0.5 to 50, which again are outside the range suggested by CEN (1996) to indicate solubility control. In a similar manner, errors could result from the interpretation of species that have reached their solubility limits in only one of the two leaching phases. Other factors including co-ion effects and temperature may also effect the rate of leaching and hence the calculated Q values. Therefore, the leaching behaviour theoretically defined by the Q values, are used here only as a broad guidance indicator.

The Q values calculated for the materials under investigation are summarised in Table 3-16a for the aggregates and Table 3-16b for the binders (excluding quicklime for which the Q values could not be calculated because no C2 sample was obtained). An additional category of Q<1 has been included to identify those species which have come out of solution in the L/S=10 phase of the test. The Q values for many of the trace elements could not be determined because the concentrations in the leachate fell below the limits of analytical detection. For this reason, the Q value of many of the species is not listed and some species listed as having a Q value less than 2 may actually have Q values less than 1.

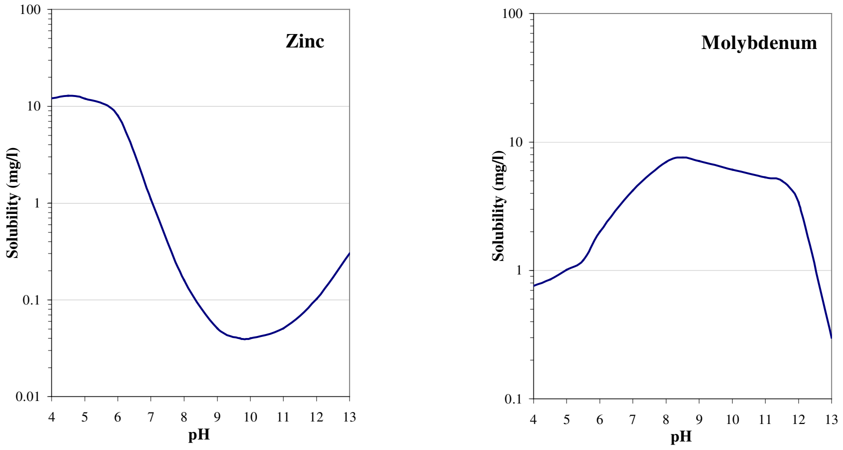


Figure 3-11: Typical curves of solubility verses pH for the leaching of zinc and molybdenum from pulverized-fuel ash [from Kossen and van der Sloot, 1997]

Table 3-16a: Leaching behaviour of the aggregates in the rapid leaching characterisation test (Summary of "Q" values)

Aggregate	**Q values (A2-10/A2)**			
	Q < 1	**Q ≤ 2**	**2 < Q < 6**	**Q ≥ 6**
AP	NO_2 Zn	Mg Na K Cl SO_4 NO_3 Br S Mn Cu	Ca HCO_3 F TIC Si Ba Sr B	Al
BFS	Mn	Ca Mg Na K Cl SO_4 S Sr Li	F TOC Si Ba B	NH_4 Al Se
BOS	Zn	Na K Cl NO_3 NO_2 Sr	Ca HCO_3 SO_4 F TOC S Si Ba Al	TIC As
CCS	Zn	Na Cl SO_4 F S As	Ca Mg K NO_2 TIC Si Ba Sr Al B	TOC Mn
CR (0.5-1.5)	Na Fe	Mg K Cl SO_4 Br TOC NH_4 Si B As	Ca HCO_3 F TIC Ba Sr Mn Se	S Zn
CR (1-4)	Na Cl Br Ba Se	SO_4 S NH_4 Sr Mn As	Ca Mg F TOC TIC Zn	
FSB	SO_4 TOC Se	Na K F P S Ba Sr Cu Zn V As	Ca Mg Cl NO_2 TIC NH_4 Si Mn Fe Al	HCO_3
FBA	NO_2	Na K Cl NO_3 Li	Ca SO_4 F TOC TIC S NH_4 Si Ba Sr Zn B As Se	HCO_3 Al
GR		Na Cl SO_4 NO_3 NO_2 F S NH_4 B	Ca Mg K HCO_3 TIC Si Ba Sr Mn	Al
LS	Zn Se	Na Cl SO_4 NO_3 B	Ca Mg F TOC S Si Sr As	NO_2 TIC NH_4 Ba Al
MS	Ca Mg Sr	Na K Cl SO_4 Br S NH_4 Mn Li Se	F Si	TOC Ba Fe Al Cu Zn B
MSW(N)	Se	Ca Mg Na K Cl SO_4 Br TOC P S NH_4 Mn Fe Ni Cu Zn Mo	F TIC Si Ba Sr B	HCO_3 NO_2 Al
MSW(O)		Na K Cl Br TOC P NH_4 Si Cu Mo Se	Ca Mg HCO_3 SO_4 NO_2 TIC S Ba Sr Al B	F
PFA	Cl	Na K SO_4 Br S Cr Mo V Li Se	Ca HCO_3 F TOC TIC Si Sr B As	NO_2 NH_4 Ba Al
SSA	Cl Ba Zn	Ca Mg Na K SO_4 Br TOC NH_4 Mo Li B Se	F TIC S Si Sr Mn V As	P Al

Table 3-16b: Leaching behaviour of the binders and activators in the rapid leaching characterisation test (Summary of "Q" values)

Binder/ Activator	**Q values (A2-10/A2)**			
	$Q < 1$	**$Q \leq 2$**	**$2 < Q < 6$**	**$Q \geq 6$**
Ce	Cl SO_4 S Cr Se	Na K Li	HCO_3 F TOC TIC Sr	Ca Si Ba
CKD	SO_4 TIC S Fe Al Cu Zn As Se	Na K Cl Br NO_2 TOC NH_4 Ba Sr Cr Li	Ca HCO_3 F	
FGD	Mg Na Cl Zn	Br Ba B	Ca SO_4 F TOC TIC Si Sr Mn Al As Se	S

In general, the majority of species have a Q value either less than 2 or between 2 and 6. Only a few species have Q values greater than 6. For the test on the minestone, there are relatively more species with a Q value greater than 6, and the alkali earth metals have Q values less than 1. This can probably be explained by the reduction in pH between the C2 and C2-10 leachate (from pH 6.8 to 3.4) and the resultant harsher leaching environment and increase in solubility limits of some lower mobility species.

To provide further data on the mobility of each chemical species for each material, the percentage of the total composition of the solid that was leached, was calculated. Table 3-17 summarises the percentage of the total solid composition of each species that has been released during the L/S=2-10 phase of the leaching test. The data is only available for those species that were measured in detectable proportions in both the solid material and in the leachates.

It can be seen from Table 3-17a and b that the most mobile species are generally the alkali and alkali earth metals, particularly sodium, potassium and calcium. Barium and strontium have varying mobility and magnesium has low mobility except in the flue-gas desulphurisation gypsum and sewage sludge incinerator ash. The mobility of the transition elements is generally low, except for molybdenum of which more than 5% is released from most materials. In addition, a number of materials show a release of greater than 5% of the zinc and chromium and greater than 10% of the sulphur and selenium. For the materials where these mobile species were found to have a normalised solids concentration greater than unity (see Table 3-7a and b), the test leachates generally had normalised concentrations greater than unity of that species (see Table 3-14). Again, this illustrates the species mobility.

Table 3-17a: Percentage of total solid composition released during the rapid leaching characterisation test at L/S =2-10 (aggregates)

Aggregate	Test pH	% of total solid composition released					
		<0.1	0.1-1	1-5	5-10	10-50	50-100
AP	8.5	Si Mn Fe Al Zn	Ca Mg Na K Ba Sr Cr	S			
BFS	6.5	Mg P Si Mn Fe Al Zn	Ca Ba Sr	Na Se	K	S	
BOS	12.2	Mg Si Mn Fe Al Cr V	Zn Se	K Ba	Ca	S	
CCS	6.5	K Si Mn Fe Al	Mg Na Ba Sr Zn As				Ca
CR (0.5-1.5)	6.1	TOC Si Fe Al Co Cu	Na Zn			Ba	Ca
CR (1-4)	5.4	Na TOC Si Fe Al Co Cu	Zn	Ba	Ca		
FSB	6.3	Si Mn Fe Al Cr	Mg Ba Cu As	P Zn Se	Ca	Na K TOC	
FBA	10.4	Mg P Si Mn Fe Al Ni Cu Zn Cr V	K TOC Ba As	Na	Ca Se		
GR	8.6	Na Si Mn Fe Al Zn V	Ca Mg K Ba Sr				
LS	6.5	Ca Si Mn Fe Zn Se	Mg Sr Al	Ba	Na	As	
MS	3.4	Si Mn Fe Al Cr V	Mg K Ba Sr Cu	TOC Se	Ca Zn	Na	S
MSW (N)	8.5	Si Ba Mn Fe Ni Zn Cr Se	Mg Al Cu	Na P	Ca K TOC	S	
MSW (O)	8.8	Mg Si Mn Fe Zn Cr Cd	P Ba Al Cu	TOC As Se	Mo	Ca Na K	
PFA	10.6	Mg P Si Mn Fe Al Ni Cu	TOC Ba Zn Cr V As	K Se	Ca Na	Mo	
SSA	6.7	P Si Ba Mn Fe Al Cu Zn Cr	K V As	Mg Na TOC	Se	Ca Mo	

Table 3-17b: Percentage of total solid composition released during the rapid leaching characterisation test at L/S =2-10 (binders)

Binder/ Activator	Test pH	% of total solid composition released					
		<0.1	0.1-1	1-5	5-10	10-50	50-100
Ce	12.4	Mg Si Mn Fe Al Zn As		S	Ca Cr Se	Ba Sr	Na K
CKD	12.4	Si Mn Fe Al	As	Ba Se	Ca Sr	S Zn Cr	Na K
FGD	6.5	Fe	Si Al Zn Se	Na S Ba Sr As	Ca Mg		
Lime	12.7	Mg Si Mn Fe Zn	Al		Ca	Na K Se	Ba Sr

The release data shown in Table 3-17 also allows the relationship between the total solids composition and the leached quantities to be considered. It can be concluded that the mobility of a particular species varies by up to 5 orders of magnitude between different materials and that for a particular material the mobility of the different species shows a similar variation. This is illustrate more clearly in Figure 3-12 which shows the proportion of sodium and zinc released from each material, and Figure 3-13 which shows the proportion of each detectable species leached from both the pulverized-fuel ash and limestone. The relationship between composition and leachability is not consistent and therefore knowledge of one cannot be used to predict the other.

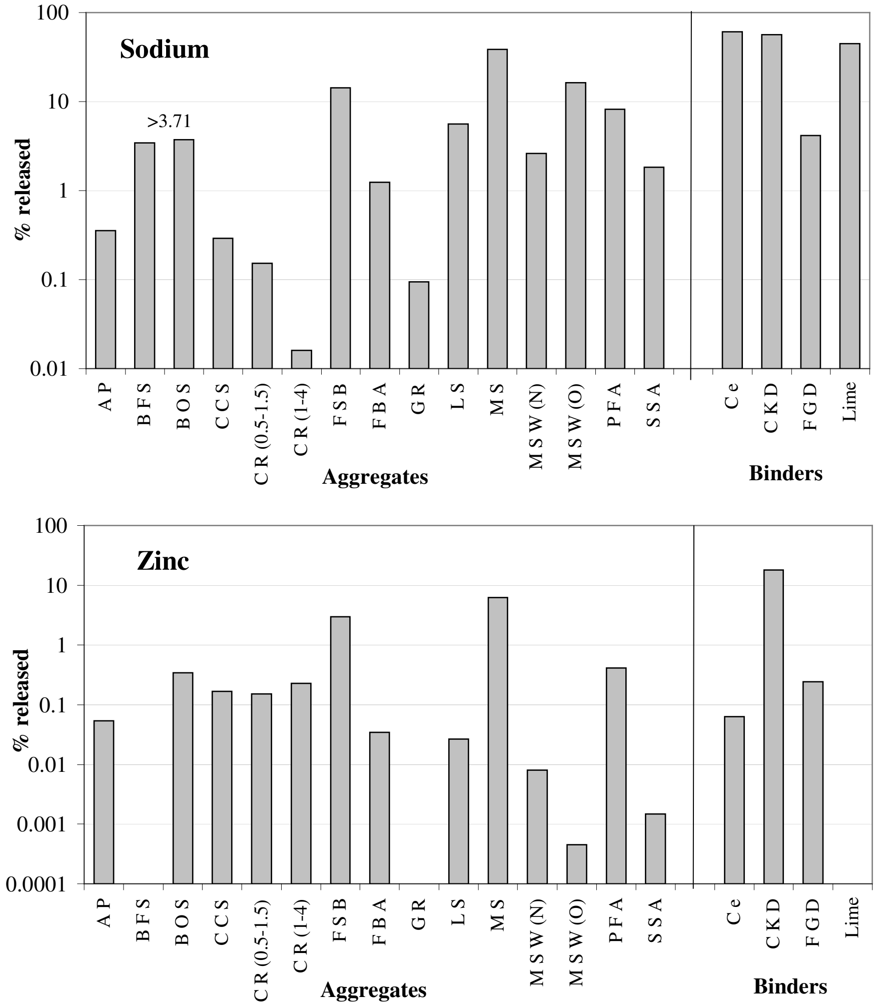


Figure 3-12: Release of sodium and zinc from materials in the rapid leaching characterisation test at L/S=2-10

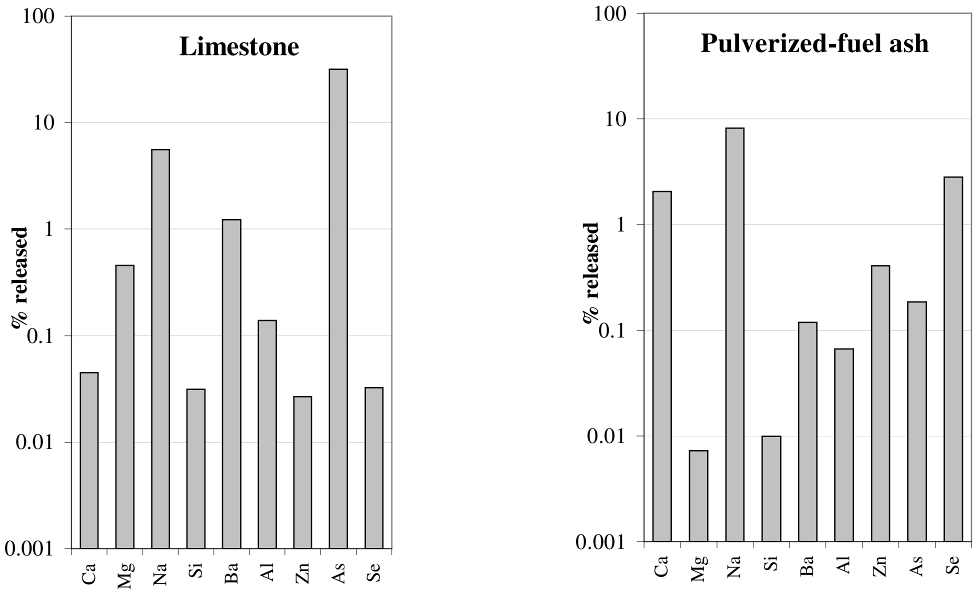


Figure 3-13: Release of different species from pulverized-fuel ash and limestone in the rapid leaching characterisation test at L/S=2-10

3.4.7 Summary and Conclusions

- The draft CEN compliance leaching test was selected and used as a rapid leaching characterisation test for obtaining an estimation of the leaching properties of the aggregates, binders and activators.

- Equipment was designed and fabricated for material particle size reduction and for agitation of the leaching vessels.

- 14 aggregate materials and 4 binders/activators were tested using the rapid leaching characterisation test procedure. The repeatability of the test is thought to be good.

- The pH of the leachate from the rapid leaching characterisation test appears to be controlled by the materials being leached, rather than the pH of the leachant.

- The results of the rapid leaching characterisation test were normalised arbitrarily to water quality maxima derived from UK Drinking Water Quality Standards, to provide an indication of the relative toxicity of the concentrations of different chemical species. All materials tested, including the limestone and granite primary aggregates showed at least one chemical species at concentrations in excess of the water quality maxima. Thus, a direct comparison would again have led to the exclusion of the limestone and granite aggregates from use, which is considered to be inappropriate and therefore implies that either the test and/or the comparison to the water quality maxima are inappropriate as methods of material assessment.

- A comparison between leached quantity and the total solid composition was used to give an indication of species mobility. In general, it was found that the alkali and alkali earth metals were the most mobile species with the exception of magnesium, and occasionally barium and strontium. The transition metals exhibited low mobility, except for molybdenum and, to a limited extent, zinc and chromium. When leaching is controlled by low mobility, it is possible that at lower L/S ratios, such as those likely to be found within a road construction, resulting concentrations may be higher.

- Species mobility was not found to be consistent for all chemical species within a material or for an individual chemical species in different materials. It was therefore confirmed that knowledge of either the solid composition or species leachability could not be used to effectively predict the other.

- The rapid leaching characterisation provided initial information on material leaching which can be used as a benchmark for the more simulative testing discussed later in this thesis.

3.5 Summary

- A selection of 14 aggregates and 6 binders were chosen for investigation. Conventional primary aggregates and traditional binders were included within the selection to enable a comparison to be made to the alternative materials.

- A representative sample of each material was obtained from the source that was considered most simulative of the material that would be used in-situ.

- The aggregate materials and binders were characterised by determining the chemical composition of the solid fraction of the material and then by performing a rapid leaching characterisation test.

- The examination of the chemical composition of the solid fraction revealed that all materials (except crumbed rubber) have either silicon or calcium as their major component, with iron, sulphur and aluminium also making significant contributions.

- In the rapid leaching characterisation test, chemical species were leached at concentrations between 10,000 and 0.001 mg/l. In general, the alkali and alkali earth metals were the most mobile species, with the exception of magnesium and sometimes the barium and strontium that exhibited lower mobility. The transition metals generally exhibited low mobility, except for molybdenum and, in some cases, zinc and chromium.

- The variation in toxicity of different chemical species was considered by comparison of the solids composition and rapid leaching characterisation test data to published quality criteria. All materials, including the primary aggregates, contained either solid or leachate species concentrations above the appropriate criteria. This highlights the limitations of using such assessment criteria for determining acceptability.

4 Assessment of Leaching by Diffusion

4.1 Introduction

The material characterisation described in Chapter 3 generated some basic knowledge and understanding of the composition and leaching potential of each of the materials under investigation. Constituents of potential concern have been identified for each material and, to a limited extent, these constituents have been compared for the different materials. Chapter 3 concluded that if a direct comparison between characterisation data and any existing arbitrary criteria was used for acceptability, then most of the materials, including primary aggregates and traditional binders, would be excluded from use in road construction or bulk fill, as a result of one or more species measured at concentrations in excess of the criteria. This is obviously inappropriate, as it would exclude the use of long-established materials, such as limestone, granite or cement, in the majority of utilisation scenarios.

It has already been seen in Chapter 3 that the composition of the solid fraction cannot generally be used to predict what quantity or concentration of a particular constituent will leach. Similar concerns arise when considering whether the rapid leaching test can be used to assess what will leach from materials when used in a road construction or bulk fill application. There are many discrepancies between the conditions found within the test and those likely to be experienced in an application environment. In a road construction or bulk fill application, the physical and hydraulic conditions to which the aggregate materials are exposed are very different to those reproduced in the rapid leaching test. The principal examples of these differences are:

a) The rapid leaching characterisation test uses size-reduced material, with 90% less than 4mm and 100% less than 10mm. Conventional aggregate material (such as crushed rock) conforms to a specific grading with particle diameters generally ranging from dust (<75μm) up to, maybe, 35 to 75 mm. The particle size and grading curve may also affect the quantity and rate of leaching. Larger particles may leach at a different (potentially slower) rate than smaller particles of the same material. Larger particles have a smaller surface area to volume ratio, which decreases the amount of leachant in direct contact with the material. In addition, larger particles have a longer, and hence slower, internal diffusion pathway. Crushing the material samples in the laboratory also exposes "unweathered" surfaces that may contribute differently to the quantity and rate of leaching when compared to the "weathered" surfaces of the full graded "as-supplied" aggregate.

b) In a road construction or bulk fill application the aggregate materials are almost always used in a compacted condition. Compaction is used to improve the mechanical performance of the structural layer but may also effect the quantity and rate of leaching. Compaction has the effect of reducing the volume of the void space within the material and also reducing the material permeability. Both of these are likely to reduce the volume of leachate coming into contact with the material, and hence reduce the quantity of leachate (compared to the same material with a higher void ratio and permeability). However, in a lower permeability material the contact time between the leachant and the material is increased and therefore the concentration of species in that leachate may increase (although solubility and other pH and co-ion effects may be more dominant than time). Some alternative aggregate materials exhibit self, or hydraulic, binding properties when mixed with water. This chemical reaction may alter the leaching properties of the material. For these reasons, some compacted aggregates may leach more like a monolith than a granular material. This may be more apparent for the fine grained and low permeability materials.

c) The rapid leaching test has an aggressive mixing action. Under realistic conditions such mixing would never occur. The type of contact between the material and leachate will depend, principally, upon the material void ratio and permeability, as described above. Other factors will also be involved including the mechanisms by which any leachant enters the construction, the local hydrological conditions and the nature of the construction particularly in terms of the foundations and drainage. The hydraulic conditions within the construction layer may effect which type of leaching process dominates. For example, in materials with a relatively high permeability, leaching by advection (surface wash-off) may be the dominant process. However, as the permeability drops and residency times increase, leaching by diffusion may become more important. It was shown by van der Sloot (1991) that for granular materials and sludges (particle diameter <40mm) leaching is dominated by advection, whilst for monolithic materials (solid matrix, particle diameter >40mm) leaching is dominated by diffusion. Again, it is considered that compacted aggregate materials or large particles within an aggregate material may leach more like a monolith than a granular material.

The rapid leaching characterisation test does not replicate the physical or hydrological conditions found within a real road construction or bulk fill scenario. To investigate how the limitations of the rapid leaching characterisation test effect leaching in the road construction application environment, more realistic simulations are required. A tiered testing strategy was developed to investigate how the particular physical and hydrological considerations of the road construction and bulk fill scenario may affect the leaching process as follows:

a) Hydraulic properties:
   - Leaching by diffusion into predominantly still or periodically moving pore water;
   - Leaching by advection into predominantly flowing pore water.
   - The relative importance of the diffusive and advective leaching properties in relation to the overall quantity of leached species.

b) Physical properties:
   - Realistic material gradings;
   - Realistic material gradings compacted to realistic densities.

The following sections of this chapter present the results of the tiered approach described above for leaching by diffusion. Leaching by advection is described in Chapter 5.

4.2 Assessment of Leaching by Diffusion

Diffusion refers to molecular motion. It can be explained by considering that any liquid solution (that is kept at constant temperature) will eventually reach a uniform composition throughout the system, independent of the original distribution. Diffusion also takes place in solids, although the rate is much slower [Sharp, 1990].

Leaching of chemical species from aggregate materials by diffusion occurs as molecules move along concentration gradients that build up in the leachant, between the leachant and the solid material and in the solid material. To assess diffusive leaching the leachant and pore water must be predominantly still or periodically moving and should continually surround the material.

Diffusion is generally seen as the dominant mechanism for leaching from monolithic materials. A number of countries have developed diffusive leaching tests, especially tank-leaching methods, that have generally evolved for use on monolithic materials. The typical arrangements of a tank-leaching test are shown in Figure 4-1. Table 4-1 summarises the various procedures and conditions used in the Dutch [NEN 7345, 1994], German [Bialucha, 2000] and draft CEN TC 154 "aggregates" [prEN 1744-3, 2000] versions of a tank-leaching test. It should be noted that the draft CEN TC 154 tank-leaching test was formalised as a European and British Standard test [BS EN 1744 – 3, 2002] after this section of the testing programme had been commenced and was therefore not used as a reference for developing the utilised procedure, but is included here for completeness.

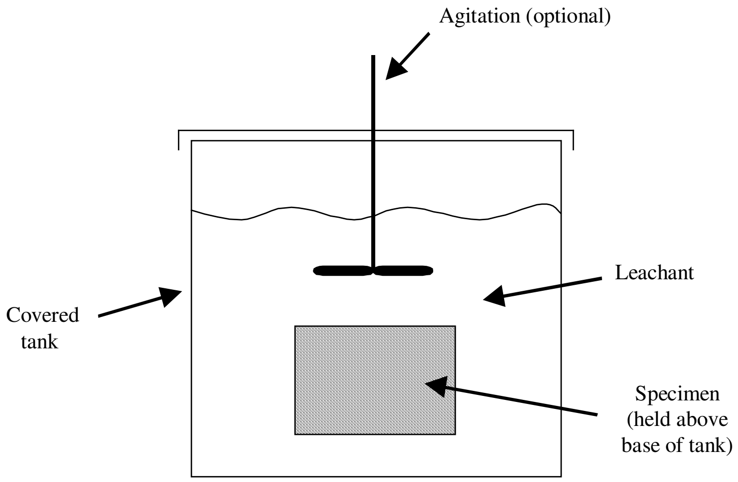


Figure 4-1: Typical arrangement of apparatus in tank-leaching test

A tank-leaching test was chosen as the method for investigating diffusive leaching from the materials being examined. The reasons for choosing a tank-leaching test procedure include:

a) At the time of study, the tank-leaching test had been utilised for assessing leaching from monolithic and granular materials;

b) A tank-leaching test was being developed by CEN Technical Committee 154 to assess leaching from aggregate materials and was likely to become a European Standard method;

c) The test methodology can be adapted to enable materials in various physical forms (e.g. at realistic gradings, compacted, bound), to be examined.

Table 4-1: Test conditions for different tank-leaching test standards

	Tank-Leaching Test Method		
Test Conditions	**Dutch [NEN 7345, 1995]**	**German [Bialucha 2000]**	**CEN TC 154 [Draft prEN 1744-3, 2000]**
Apparatus (for testing inorganic species)	-Closable polyethylene tank -Sample stands on plastic support or suspended from plastic thread	- Polypropylene or glass cylindrical or rectangular tank -Cover for tank -Sample on glass, teflon or polypropylene screen insert, mesh size 2mm	- Polypropylene or glass cylindrical tank -Sealed cover for tank -Sample on glass, teflon or polypropylene screen insert, mesh size 2mm
Leachant	-Demineralised water -Acidified to pH 4 with nitric acid -Unagitated	-Demineralised water -Agitated using magnetic stirrer	-Distilled or demineralised water -Agitated using glass or teflon motor driven dip stirrer
Leaching duration	-Leachant renewal -8 cycles -6hrs to 64 days	-24 hours	-24 hours
Material sample	-Monolithic and stabilised materials -Sample dimension >40mm (cuboide)	-Bound or unbound aggregates -2kg sample	-Unbound, uncompacted aggregates -Particle size up to 32mm -2kg sample
Liquid/solid ratio	-Liquid/material volume=5	-Liquid/material mass=10	-Liquid/material mass = 10
Comments	-Theoretical calculations of diffusion coefficients		

In more practical terms, a tank-leaching test was considered to be a suitable test because:

a) The procedure could be adjusted to meet the requirements of the research. For example, the liquid to solid ratio, leachant type and sampling frequency could be chosen;
b) The equipment is simple to develop and the test is relatively easy to perform. This is important if the test is to be suitable for application in industry;
c) Use of a similar test procedure on materials in various physical forms enables easy comparison between tests, and hence identification of the resultant effects on leaching.

The tank-leaching test may be more simulative of any potential utilisation scenarios than that shown by the rapid leaching characterisation test. However this does not mean that the tank-leaching test will directly simulate what would happen in the real situation. There are many differences between the leaching conditions within a tank-leaching test and those experienced under field conditions. Some important differences include:

a) The environmental conditions of the in-situ scenario are not illustrated. Factors such as the design of the construction, methods of leachant ingress and egress, temperature variations and biological action are not represented;
b) The liquid to solid ratio used in the test is much greater than the volume of the void space in the material. This quantity of leachant therefore represents many bed volumes of leachate in the in-situ scenario.

4.3 Tank-Leaching Test Conditions

The procedure adopted in this study combined various aspects of the existing tank-leaching test procedures (see Table 4-1). A summary of the adopted test conditions is given in Table 4-2 below followed by the reasoning behind these decisions.

Table 4-2: Tank-leaching test conditions used in the research

	Tank-leaching test on material	
Test Conditions	**Realistic Grading**	**Real Grading and Compacted**
Apparatus	-Polypropylene leaching vessel with tap at base -Polypropylene and geotextile sample support frame	
Leachant	-Distilled water (5<pH<7, conductivity <0.5mS/m) -Unagitated	
Leaching duration	-Sampling at intervals up to 64 days	
Material sample	2kg	-Compacted cylindrical specimen; diameter 150mm, height 70mm
Liquid/solid ratio	-Liquid/material mass=10	-Liquid/material volume=10

The tank-leaching tests were principally investigating the leaching of inorganic constituents. To minimise any contamination from the apparatus, the leaching vessel, material support stand and other apparatus that were to be in contact with the leachant or material sample were made from plastics. The leaching vessel was made of polypropylene and the material support stand from polypropylene with a geotextile grid.

A cylindrical shaped tank was chosen for a number of reasons. Firstly, Bialucha (2000) reported problems with the accumulation of fine-grained materials (that had fallen through the retaining mesh support) in the corners of rectangular shaped leaching vessels, and therefore recommended the use of a cylindrical vessel. Secondly, this test was developed for application to compacted and bound material samples as well as for uncompacted materials. Since the compacted specimens were to be cylindrical in shape (since such samples are easier to fabricate (Nunes, 1997)) a cylindrical shaped leaching vessel would allow the material sample to be centred in the leachate, equidistant from the walls of the vessel. The dimensions of the chosen tank were approximately 380mm in diameter and 390mm in height, giving a maximum volume of about 40 litres. Such dimensions are comparable to those used in other procedures (see Table 4-1) and were suitable for testing all of the materials at the various L/S ratios required in the different types of tank-leaching test.

The leachant chosen for the tank-leaching tests was the same as that used in the rapid leaching characterisation tests (distilled water ($5<pH<7$, conductivity <0.5mS/m)), thus allowing simple comparison. The leachate was not pH adjusted or agitated. Both were thought to be unrealistic of the in-situ environment where the material will generally govern the pH of the pore water as it moves slowly through the pavement under low hydraulic gradients. Agitation has since been shown to increase the rate of leaching [Wahlstromöm et al., 1998].

Preliminary testing was carried out to determine the time taken for the tests to establish equilibrium (or quasi-equilibrium) conditions. Using measurements of the pH and conductivity of the system, it was found that the time to equilibrium varied between materials. It was concluded that the tests should generally be carried out over a period of 64 days, although this could be changed depending on the material being tested.

Ideally, samples of the leachate would have been analysed after 2 hours, 6 hours, 24 hours, 2 days, 4 days, 8 days, 16 days, 32 days and 64 days. However, the expense of such frequent sampling was beyond the budget of the project. Therefore, a complete chemical analysis of the leachate was carried out on the samples taken after 6 and 24 hours, as they correspond to the sampling times in the rapid leaching characterisation test. In addition, leachate analysis was typically carried out after 4 days, 16 days and 64 days.

To test materials at realistic gradings, a material sample with a mass of 2kg was chosen, as used in the procedure described by Bialucha (2000). This sample mass enabled a representative material sample to be obtained for most materials whilst keeping the sample and corresponding volume of leachant of a manageable size. The fixed mass of material makes it easier to compare the results to the rapid leaching characterisation test, which also used a fixed mass sample.

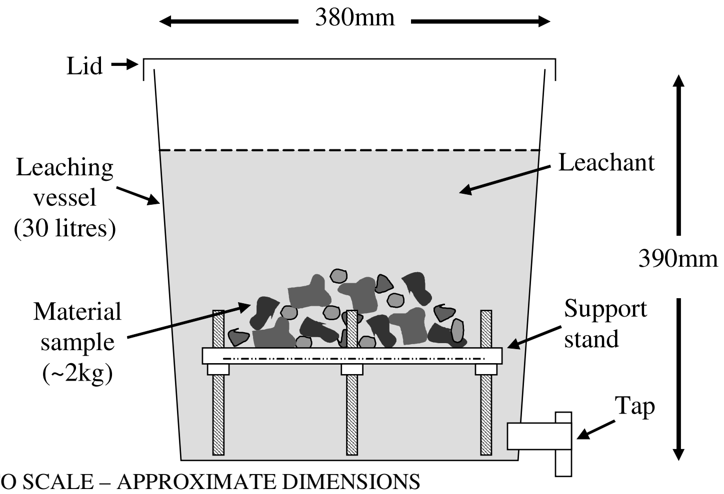
The tests on compacted materials utilised a sample with a fixed volume, similar to that of the NEN 7345 (1995) procedure. In the field, a material is generally required to perform a filling task (e.g. pavement layer thickness) for which a volume of material is required. Thus, using a fixed sample volume takes account of the variations in the density of the different materials. A cylindrical sample was chosen over a cubic sample because such specimens are easier to fabricate from unbound materials [Nunes, 1997].

For the tests on the materials at realistic gradings, a liquid to solid ratio of 10 to 1 by mass was chosen. This allowed a direct comparison to be made to the rapid leaching characterisation test. For the tests on the compacted materials a liquid to solid ratio of 10 to 1 by volume was chosen. Again, this volumetric ratio was chosen to account for the density variations between materials.

4.3.1 Equipment Development

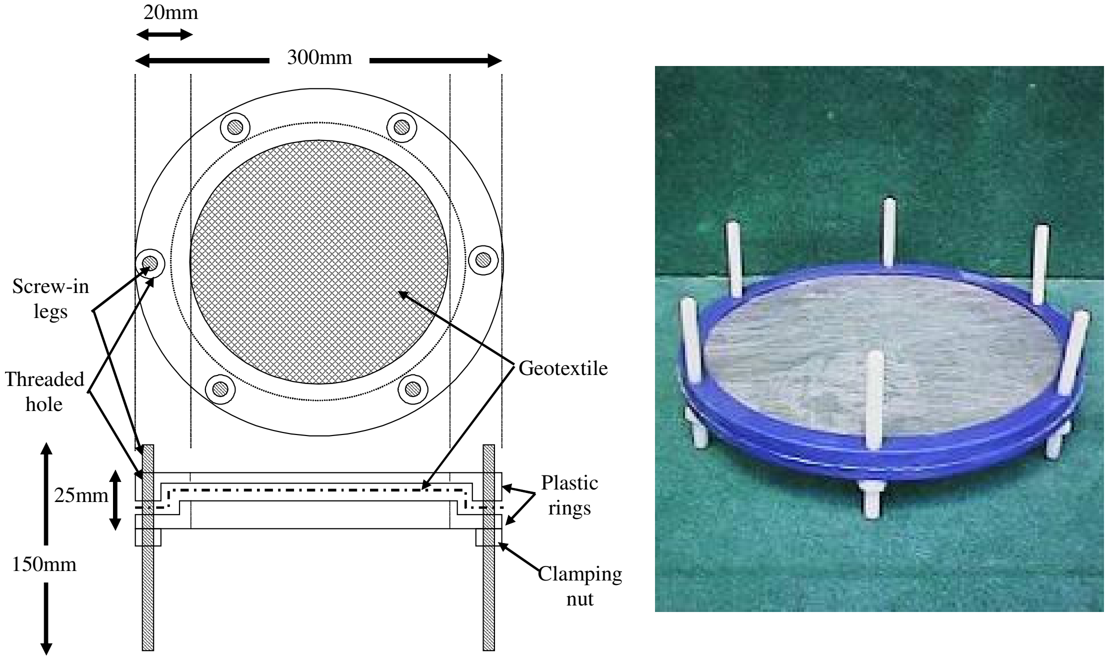
As in the rapid leaching characterisation test, all laboratory ware was made from plastic with high strength and low chemical adsorption. All test equipment that would come into contact with the leachant or material sample was developed from similar plastic materials. A leaching vessel and material support frame were designed and fabricated for use in the tank-leaching tests and are illustrated in Figure 4-2 and Figure 4-3.

The body of the leaching vessel was made from a "food quality", high-density polyethylene container with a lid. The container was cylindrical (with a slight taper, to enable stacking when not in use) with a maximum internal diameter of 381mm and a height of 388mm, giving a volume of approximately 40 litres. To enable leachate samples to be taken without significant disturbance of the leachate, a drum tap, made from polythene, was fixed into the side of the leaching vessel near the base. A schematic diagram of the set up for the tank-leaching test is shown in Figure 4-2.



NOT TO SCALE – APPROXIMATE DIMENSIONS

Figure 4-2: Schematic diagram of the equipment for the tank-leaching test (shown with uncompacted specimen)



NOT TO SCALE - APPROXIMATE DIMENSIONS

Figure 4-3: Detail and photograph of support stand for the tank-leaching tests

The material sample needed to be surrounded by leachant and held equidistant between the base of the leaching vessel and the surface of the leachant. The height of the support stand therefore needed to be adjustable to accommodate the various volumes of leachant that were required for the range of tests. The design of the material support stand should enable the leachate to freely circulate around the material. The support stand that was manufactured is illustrated in Figure 4-3. The frame is made from two interlocking plastic rings supported on six screw-in legs. The two plastic rings clip over each other and are used to secure a fine geotextile mesh (Terram 500) inside the rings. The material sample sits on the geotextile which can be replaced between tests to minimise cross-contamination between samples. The screw-in legs pass through the geotextile and screw into threaded holes in the plastic rings. The legs are screwed to the required height and secured in place with a clamping nut.

4.4 Diffusive Leaching from Materials with Realistic Grading

To investigate the effect of material grading on diffusive leaching the tank-leaching test was performed on a selection of the materials with differing particle sizes and material origins.

4.4.1 Methodology

The tank-leaching test methodology that was developed to investigate diffusive leaching from materials with realistic gradings is divided into two sections, sample preparation and tank-leaching test. The details of the methodology are given in Box 4-1 and Box 4-2 respectively.

Box 4-1: Sample preparation methodology for the tank-leaching test on materials with realistic gradings

1) *Obtain a representative sub-sample* of the test material, with a mass of approximately 5kg, by using coning and quartering [BS 812: Part 102, 1989; BS EN 932-1, 1997]. This sub-sample will form the laboratory sample.
2) *Determine the residual moisture content* of the material in the laboratory sample by oven drying a sub-sample to constant weight [BS 812: Part 109, 1990; BS EN 1097-5, 1999].
3) *Obtain the test sample,* of approximately 2kg ± 5% of DRY material, by sub-sampling from the laboratory sample, using coning and quartering.
4) *Calculate the mass of water in the test sample.* The total mass of leachant in the tank-leaching test should include the moisture held within the test sample.

Box 4-2: Methodology for the tank-leaching test on materials with realistic gradings

1) *Determine the volume of leachate required* to establish a liquid to solid mass ratio of 10. The total mass of leachant that is required is equal to 10 times the DRY mass of the test sample. The mass of leachant that has to be added to the leaching vessel is calculated by subtracting the moisture held within the test sample (See Box 4-1, Part 4) from the total volume of leachant that is required.
2) *Prepare the tank-leaching test apparatus.* Clamp a new sheet of geotextile into the support stand for the test sample. Depending upon the volume of leachant that is being used, adjust the length of the supporting legs, to ensure that the material sample will be centred vertically within the leachant. Thoroughly clean the leaching vessel and support stand with leachant.
3) *Start the tank-leaching test.* Weigh the required volume of leachant (±2%) into the leaching vessel. Place the material sample onto the geotextile in the support stand and gently lower the stand and sample into the leaching vessel. Record the time and date at the start of the test.
4) *Leachate Sampling.* To sample the leachate drain a volume of approximately 100ml through the tap in the side of the leaching vessel. Record the mass of the leachate sample to enable the remaining liquid-to-solid ratio to be calculated. Do not replace the leachate.
5) *Filtration and Sample Preservation.* Allow any suspended solids in the leachate to settle for up to 15 minutes. Filter the leachate through a 0.45μm filter paper using a vacuum or syringe filtration apparatus, or similar. Measure the pH and conductivity of the leachate. Retain 3 samples of the leachate for analysis (1 x unacidified, 1 x 1% nitric acid, 1 x 1% hydrochloric acid) (See Section 3.4.3 for details). Refrigerate the samples until analysis.
6) *Leachate Sampling Frequency.* Leachate sampling and analysis is normally carried out after 6 hours, 24 hours, 4 days, 16 days and 64 days. If necessary, adjust the test duration and the leachate sampling frequency according to the nature of the material being tested. The variation in the pH and conductivity of consecutive samples of the leachate, can be used to indicate whether the composition of the leachate is still changing or whether equilibrium (or near equilibrium) conditions have been achieved. If consecutive samples reveal no change in the pH or conductivity then the test can be concluded. Tests would normally be expected to last for at least 30 days, with the majority continuing to the full 64 days. The test duration can also be extended beyond 64 days if significant changes are still occurring.

The leachates generated from the tank-leaching tests were analysed for the complete range of 37 determinants that are listed in Table 3-9. Where the analytical method allowed, the majority of determinants were measured for each leachate.

4.4.2 Tank-Leaching Tests on Materials at Realistic Gradings

The tank-leaching test on materials with realistic gradings was carried out on the 9 materials listed in Table 4-3. The materials were selected to cover the full range of gradings presented by the materials under investigation in this research. The materials were also selected to include the various material types including primary aggregates, processed rock and residues from high temperature processes.

Table 4-3: Summary of the materials tested in the tank-leaching tests at realistic gradings

Material	Test Abbreviation	Material Grading	Material Source
Air-cooled blastfurnace slag	BFS-L	DoT Type 1	Metalliferous slag
China clay sand	CCS-L	Medium	Processed rock
Foundry sand (phenolic,brass)	FSB-L	Fine	Treated sand
Granite	GR-L	DoT Type 1	Primary aggregate
Limestone	LS-L	DoT Type 1	Primary aggregate
Minestone	MS-L	Medium to coarse	Processed rock
MSW incinerator ash (New incinerator)	MSW(N)-L	Medium	Incinerator bottom ash
Pulverised fuel ash	PFA-L	Fine	Power station fly ash
Sewage sludge incinerator ash	SSA-L	Fine	Incinerator bottom ash

In addition to the tests listed in Table 4-3, a blank tank-leaching test was performed to determine, quantitatively, whether the test procedure results in any contamination of the leachant. The test was performed as described in Box 4-2 but without adding a material sample to the leaching vessel. A leachate volume of 20 litres (±2%) was used, which corresponds to the volume required for a material sample of exactly 2kg. As with the rapid leaching characterisation test, the blank was used to quantitatively adjust the concentration of species in the leachates from the material tests. The initial composition of the leachant in the blank test was also used as the "zero hours" concentration for the material tests.

The expense of the chemical analysis of the leachates limited the number of analyses that could be performed. Because of this, it was not possible to carry out repeated or multiple tests on every material. To give an indication of the repeatability of the tank-leaching test, a repeat test was carried out on the blastfurnace slag. The complete material sampling and testing procedure was performed twice.

The tank-leaching tests on materials at realistic gradings were generally performed with few problems, although a number of minor difficulties were encountered. The most significant problem was that one of the leachate samples taken from the foundry sand (phenolic, brass) test after 4 days, was broken during transportation or storage prior to analysis. Therefore some determinants could not be measured for this leachate.

4.4.3 Results of Blank Tank-Leaching Test

The composition of the leachates obtained from the blank tank-leaching test are summarised in Table 4-4 along with the detectable components in the distilled water leachant as previously reported in Chapter 3. Any species that were listed in Table 3-9 but are not in Table 4-4 may exist but are at concentrations below the limits of analytical detection.

From Table 4-4 it can be seen that the species detected in the distilled water leachant sample are also present in the leachates from the blank tank-leaching test at similar concentrations, as expected. Additionally, the leachate from the blank tank-leaching test, at 0 hours, contains detectable concentrations of strontium, total phosphorus and total sulphur and the concentration of calcium is an order of magnitude higher than that in the distilled water leachant sample. This implies that these species have originated from initial wash-off from the apparatus. However, the detected phosphorus and sulphur are at concentrations around the limits of analytical detection experienced in the distilled water leachant sample and therefore, it is not possible to conclude whether the tank-leaching test procedure has contributed to these species. A similar situation arises for strontium, where the concentrations in the blank are of the same order of magnitude as some, but not all, of the distilled water leachant samples. Throughout the duration of the test (64 days) most of the detectable species show some fluctuation, with many showing falls as well as rises in concentration. Some species, including calcium and strontium, show an increase in concentration over time in a similar pattern to the increase in pH. The chlorine and TIC also exhibit increases in concentration in the later stages of the test. The nitrite, sulphate and ammonium appear to increase over time and may be the result of interaction with the air in the leaching vessel. The concentrations of these species are, again, close to the limits of analytical detection thus increasing potential errors and making it difficult to confirm or reject such a trend. As discussed in Section 3.4.5, the presence of zinc in the solutions has most probably originated from the lid of the leachate sample storage containers. Generally, it can be concluded that initially the tank-leaching test apparatus only contributes detectable quantities of calcium into the leachate. As a result of time, or from slow leaching of the apparatus, the concentrations of some species increase, by up to one order of magnitude, by the end of the test.

Table 4-4: Species detected in the blank tank-leaching test

Test	**Sample Time**	**pH**	**Cond. µS/cm**	**Concentration of determinants (mg/l)**			
				<0.01	**0.01- <0.1**	**0.1- <1**	**1- <10**
Blank tank-leaching test	**0 hrs**	5.0	10	Sr	P S Ba Zn	Na Cl TIC Si	Ca TOC
	6 hrs	6.7	12	Sr	Ba Zn	Na Cl TIC Si	Ca TOC
	24 hrs	6.6	14	Ba Sr	K NO_2 S Zn	Na Cl TIC Si	Ca TOC
	4 day	7.4	15	Ba Sr	NO_2 S Zn	Na Cl SO_4 TIC Si	Ca TOC
	16 day	7.4	18		NH_4 Ba Sr Zn	Na Cl SO_4 Si	Ca TOC TIC
	64 day	7.5	37		S Ba Sr Zn	Na SO_4 Si	Ca K Cl TOC TIC
Distilled water leachant		5.2	7	Se	Ba Zn	Ca Na Cl TIC Si	TOC

The blank test data was used to quantitatively reduce the concentrations of the species detected in the leachates from the tank-leaching tests on the different materials, in order to account for those species that are, thus, deemed to result from the test method or leachant. Adjustments were made for conductivity, calcium, sodium, chlorine, TOC, TIC, silicon, barium, strontium and zinc, which are the same species for which adjustments were made to the rapid leaching characterisation test data (with the exception of the TIC). No correction was applied for the remaining species listed in Table 4-4 since those species were not detected in all blank leachate samples and the accuracy of any adjustments might be low.

4.4.4 Tank-Leaching Test Repeatability

The repeatability of the tank-leaching test was investigated by performing the procedure on two separate samples of the blastfurnace slag (BFS1-L and BFS2-L). Figure 4-4 shows the pH and conductivity measured during the two tests. The variations in these determinants are summarised in Table 4-5. The pH shows very good correlation with an average deviation from the mean of 1.3%. The maximum deviation of 3.7% occurs from the leachates sampled after 6 hours. The conductivity follows the same pattern in both tests but has a higher average deviation from the mean of 2.6%. The maximum variation of 5.7% resulted from the leachates sampled after 16 days.

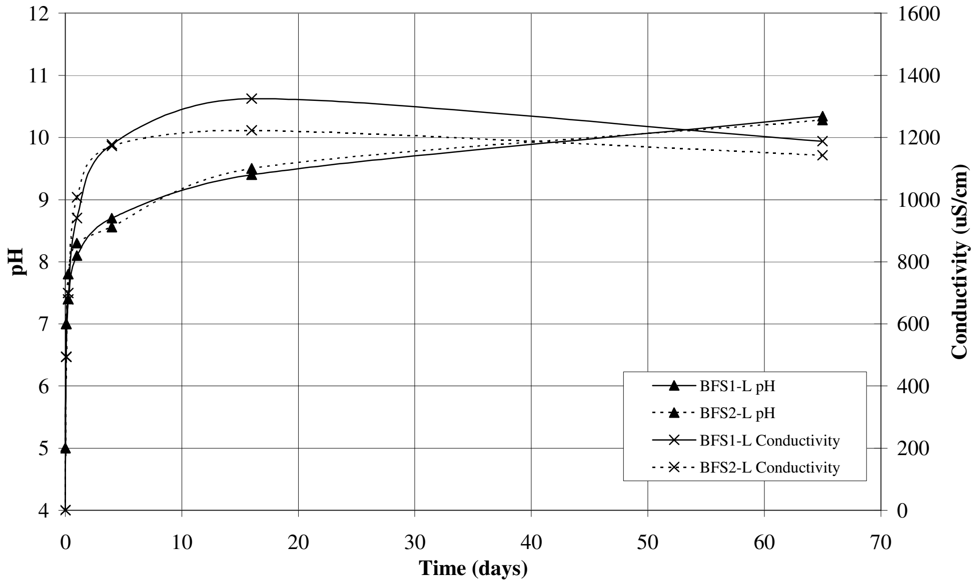


Figure 4-4: pH and conductivity data for the repeated tank-leaching tests on blastfurnace slag (BFS1-L and BFS2-L)

Table 4-5: Summary of the standard deviation (SD) of species concentrations in the leachates from the repeated tank-leaching tests

Average SD from mean (%)	**Sampling time (days)**						**Average SD from mean for all leachates (%)**
	0.08	**0.25**	**1**	**4**	**16**	**64**	
pH	0	3.7	1.7	1.1	0.7	0.4	**1.3**
Conductivity	0.1	2.2	4.9	0.3	5.7	2.7	**2.6**
All other detected species	15.8	17.6	21.4	13.2	13.3	7.9	**14.9**
All other detected species except TOC and TIC	7.1	8.3	15.8	5.1	10.7	7.1	**9.0**
All other detected species except TOC, TIC and Zn	6.1	5.1	6.8	5.4	11.5	7.5	**7.1**

Figure 4-5a and b show the average and range of concentrations of all species present above the limits of analytical detection in the leachates from the repeated tank-leaching tests after 6 hours (Figure 4-5a) and 64 days (Figure 4-5b). The concentrations presented have been adjusted to account for those species that were present in the blank (as described in the previous paragraph). It can be seen from these figures that, at any one time, the concentration of all species in the repeated tests varied by less than one order of magnitude and for most species, that variation was considerably less.

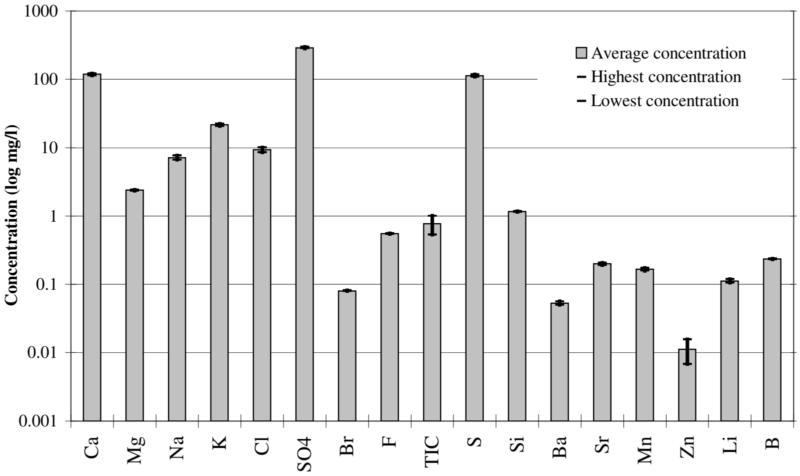


Figure 4-5a: Average and range of determinant concentrations (above the limits of analytical detection) from the repeat tank-leaching tests on blastfurnace slag after 6 hours

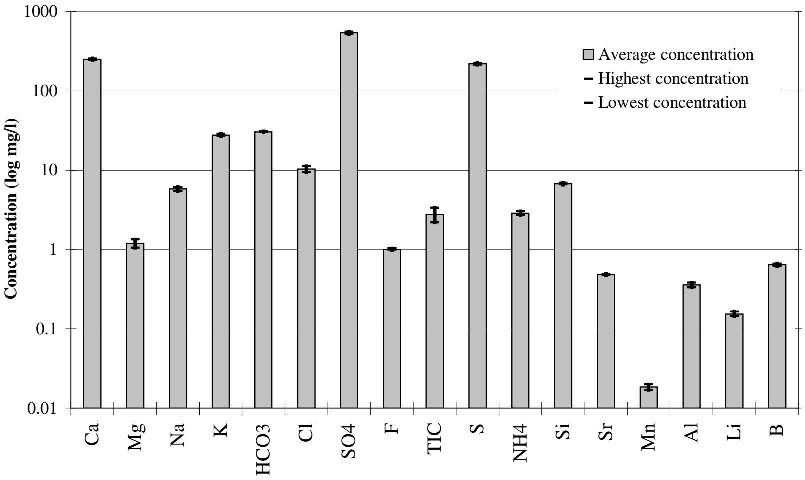


Figure 4-5b: Average and range of determinant concentrations (above the limits of analytical detection) from the repeat tank-leaching tests on blastfurnace slag after 64 days

Table 4-5 summarises the average standard deviation of all the species detected in the leachates (not including the pH and conductivity). Any species that were below the limits of analytical detection are not included in this data. Table 4-5 also summarises the standard deviations that result if the TOC and TIC, or the TOC, TIC and zinc are excluded from the calculations. It can be seen that the removal of these species results in a fall in the average standard deviation from 14.9 to 7.1%. This confirms that the standard deviation of most species will be by less than 10% from the mean.

As described in Section 3.3.2, a number of species, which were analysed for, were present at concentrations below the limits of detection for the analytical methods that were used. This is particularly the case for the transition elements. These species are not shown in Figure 4-5a and b, and are not represented in Table 4-5. The limits of analytical detection vary depending upon the species being detected and interference resulting from other species in solution. Generally, the limits of detection are in the order of 0.001 to 0.1 parts per million (ppm). Such concentrations are less than most commonly used water quality limits for the corresponding species. Therefore, where the concentration of a species was below the limits of analytical detection in both repeat tank-leaching tests, it was concluded that there was a good correlation between those species. This occurred in most cases.

In general, it has been shown that a good correlation exists between the two tank-leaching tests on blastfurnace slag and thus that the test shows an acceptable degree of repeatability for most species. TOC, TIC and other organic measures should be used with caution and cannot be guaranteed when using this test method. The high degree of reproducibility of the other species again provides reassurance of the quality of the sample preparation, testing and analysis that was performed.

4.4.5 Results

The concentrations of the species in the leachates from the tank-leaching tests on the different aggregates, were quantitatively adjusted to account for those species that had resulted from the test method or leachant. A summary of the results are given in Table 4-6 where, for each test, the species detected in the leachate after 6 hours (0.25 days), 1 day, 4 days, 16 days and at the end of the test, are tabulated within specified concentration ranges. Those species listed in Table 3-9 that are not listed in Table 4-6, may be present in that leachate but are at concentrations below the limits of analytical detection.

From Table 4-6 it can be clearly seen that the concentrations of different species in the leachates vary over at least 5 orders of magnitude. The elements that are generally leached in the highest concentrations are sodium (Na), potassium (K), calcium (Ca), chlorine (Cl) and sulphur (S). The compounds of HCO_3 and sulphate (SO_4) were also present in the highest concentration ranges in the leachates from some of the materials. Total organic carbon was also recorded in the higher concentration ranges, but the reliability of the data is not guaranteed, as noted from the repeated tests. All of these elements and compounds are the same as those listed in Section 3.4.5 as having the highest concentrations in the leachates from the rapid leaching characterisation tests.

Table 4-6 also shows that the majority of the transition elements were below the limits of analytical detection in most of the leachates. Transition elements that were detected in one or more leachates were manganese (Mn), zinc (Zn), copper (Cu), iron (Fe), nickel (Ni), molybdenum (Mo), chromium (Cr) and vanadium (V). No transition element exceeded a concentration of 10mg/l. Only molybdenum (Mo) and copper (Cu) were detected above 1mg/l and this was in the leachates from the pulverized-fuel ash and sewage sludge incinerator ash, and MSW incinerator ash, respectively.

Other List I or II species that were detected in at least one of the leachates were arsenic (As), barium (Ba), phosphorus (P), lead (Pb), ammonium (NH_4) and fluoride (F).

A typical graphical representation of the change in species concentrations in the leachate over time is given in Figure 4-6. The data presented is from the test on the sewage sludge incinerator ash, which was chosen for illustration purposes because the leachates contained a higher number of detectable species than most of the other tests.

Table 4-6: Summary of tank-leaching test data from materials at realistic gradings (concentrations in leachate)

Test	Time Days	pH	Cond. µS/cm	Concentration in leachate (mg/l)						
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**	**100-1000**	**>1000**
BFS L	**0.25**	7.4	678		Br Ba	F TIC Sr Mn Li B	Mg Na Si	K Cl	Ca SO_4 S	
	1	8.1	940		Ba	Sr Mn Li B	Mg Na F TOC TIC Si	K Cl	Ca SO_4 S	
	4	8.7	1177		NO_2 Mn	TOC Sr Al Li B	Mg Na F TIC Si	K Cl	Ca SO_4 S	
	16	9.4	1325		Al	Sr Mn Li B	Mg Na F TIC Si	K Cl	Ca SO_4 S	
	65	10.3	1188		Mn	F Sr Al Li B	Mg Na Cl TIC NH_4 Si	K HCO_3	Ca SO_4 S	
CCS L	**0.25**	6.0	0	Zn As Se		F				
	1	6.3	0	As Se	Zn	SO_4 F				
	4	6.7	4.6	Mn Cu As Se	Na Cl F NH_4 Si	Ca Mg K SO_4 S	TIC			
	16	6.9	27	Mn As Se	Na Cl	Ca Mg K SO_4 F S Si	TIC			
	22	7.0	23	As Se	Na Cl	Mg K SO_4 F P Si	Ca TIC			
FSB L	**0.25**	7.0	1241	Mn Li	P Sr Fe Cu	Mg Br Si	Ca S NH_4	Na SO_4 TIC	K HCO_3 TOC	
	1	7.1	1242	Li	Sr Mn Fe Al Cu	Ca P Si	Mg S NH_4	Na SO_4 TIC	K HCO_3 TOC	
	4	7.2	1173	Sr Li	Mn Fe Al Cu	Ca Mg P Si	S NH_4	Na	K	
	16	6.4	995	Sr Li	P Mn Cu	Mg Br NO_2 Fe Al	S Si NH_4	Na SO_4 TIC	K HCO_3 TOC	
	64	7.0	560		P Mn Cu	Mg S Fe Al	NH_4 Si	Na TIC	K HCO_3 TOC	
GR L	**0.25**	7.9	37		NO_2 F Ba Mn	Mg Na K Cl SO_4 TOC S Si	Ca NO_3 TIC			
	1	8.0	49		NO_2 Ba Sr Mn	K Cl SO_4 TOC S Si	Ca Mg Na NO_3 TIC			
	4	8.1	50		F Sr Mn	Cl NO_2 TOC S NH_4 Si Al	Ca Mg Na K SO_4 NO_3 TIC	HCO_3		
	16	8.5	64	Mn	F Sr	Cl SO_4 NO_2 S Al	Ca Mg Na K NO_3 TOC TIC Si	HCO_3		
	65	8.2	88	Mn	Sr Al	S	Mg Na K Cl SO_4 NO_3 TIC Si	Ca HCO_3		
LS L	**0.25**	8.6	56		NO_2 Si Sr	Mg Na Cl F S Ba	Ca SO_4 TOC TIC	HCO_3		
	1	8.5	61		NO_2 Si Sr	Mg Na Cl F S Ba	Ca SO_4 TOC TIC	HCO_3		
	4	8.3	58	Na	NO_2 Sr	Mg Cl NO_3 F S Si Ba	Ca SO_4 TIC	HCO_3		
	16	8.5	77		Na NO_2 NH_4 Sr	Mg K Cl NO_3 F S Si Ba	SO_4 TIC	Ca HCO_3		
	65	8.4	89		Zn	Mg NO_3 Si Ba Sr	K Cl SO_4 F TIC S	Ca HCO_3		

Table 4-6: Summary of tank-leaching test data from materials at realistic gradings (concentrations in leachate) - continued

Test	Time Days	pH	Cond. µS/cm	Concentration in leachate (mg/l)						
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**	**100-1000**	**>1000**
MS L	**0.25**	6.3	859	Se	Mn Ni Zn	F NH_4 Si Ba Sr Fe	Mg K Br	Ca S	Na Cl SO_4	
	1	6.9	1060	Se	NO_2 Mn Ni Zn Li B	F Si Ba Sr Fe	Mg K Br TIC NH_4	Ca S	Na Cl SO_4	
	4	7.4	1107	Se	Ba Mn Fe Ni Zn Li B	F Si Sr	Mg K Br TIC NH_4	Ca HCO_3 S	Na Cl SO_4	
	16	7.8	1144	Se	Ba Li B	F Sr Mn As	Mg K Br NH_4 Si	Ca TIC S	Na HCO_3 Cl SO_4	
	48	8.0	1060		Cu Li Se	NO_2 F Sr Mn B	Mg Br NH_4 Si	Ca K TIC S	Na HCO_3 Cl SO_4	
MSW (N) L	**0.25**	11.9	2778	Mn Se	Zn Mo Li	NO_2 F P Ba Sr Pb	Br TIC NH_4 Si Al Cu	SO_4 S	Ca Na K HCO_3 Cl TOC	
	1	11.9	3046	Mn Se	Ni Mo Li	NO_2 F Ba Sr Zn Pb	Br TIC NH_4 Si Al Cu	SO_4 S	Ca Na K HCO_3 Cl TOC	
	4	11.7	3205	Mn As Se	Zn Mo Li	NO_2 F P Ba Sr Pb	Br TIC NH_4 Si Al Cu	SO_4 S	Ca Na K HCO_3 Cl TOC	
	16	11.9	3122	Mn As Se	Zn Mo Li	NO_2 F P Ba Pb	NO_3 Br TIC NH_4 Si Sr Al Cu	S	Ca Na K HCO_3 Cl SO_4 TOC	
	48	11.1	2183	Mn Se	Zn Li	NO_2 F P Ba Mo Pb	Br TIC NH_4 Si Sr Al Cu		Ca Na K HCO_3 Cl SO_4 TOC S	
PFA L	**0.25**	10.1	292		NO_2 NH_4 Sr Cr As	F P Ba Mo V Li B	Cl Br TOC TIC Si Al Mo	Ca Na K HCO_3 SO_4 S		
	1	10.8	496	Zn	Mg NO_2 NH_4 Cr As	F P Ba Sr V B	Cl Br TOC TIC Si Al Mo Li	Ca Na K HCO_3 S	SO_4	
	4	11.1	685		NO_2 Zn As	Mg F P NH_4 Ba Sr Cr V B	Br TOC Si Al Mo Li	Ca Na K Cl TIC S	HCO_3 SO_4	
	16	10.9	655		NO_2 Zn As	Mg NO_3 F TOC P NH_4 Ba Sr Cr V B	Cl Br TIC Si Al Mo Li	Ca Na K S	HCO_3 SO_4	
	65	11.1	653		NO_2 As	Mg NO_3 F P Ba Sr Cr V B	Cl Br TIC NH_4 Si Al Mo Li	Ca Na K HCO_3 S	SO_4	
SSA L	**0.25**	7.1	702	V As Se	NH_4 Ba Mn Al Zn Li	F P Si Sr Mo B	Na K Cl Br TIC	Mg	Ca SO_4 S	
	1	7.6	973	As Se	NH_4 Ba Mn V Li	F P Sr Al Zn Mo B	Na K Cl Br TIC Si	Mg HCO_3	Ca SO_4 S	
	4	7.4	1418	Se	Ba Mn Zn V As	NO_2 F P NH_4 Sr Al Mo Li B	Br TIC Si	Mg Na K HCO_3 Cl	Ca SO_4 S	
	16	7.3	1804	Se	Ba Mn Zn V As	F P NH_4 Sr Al Li	Br TIC Si Mo B	Mg Na K HCO_3 Cl	Ca S	SO_4
	48	7.4	1771		Ba Mn Zn V As	P Sr Al Li	Br F TIC Si Mo B	Mg Na K HCO_3 Cl	Ca S	SO_4

It can be seen from Figure 4-6 and Table 4-6 that the majority of leached species follow a similar pattern of release, generally involving an increase in concentration over time. During the first few hours of the test, most species exhibit a period of very rapid leaching. This is followed by a period of up to a few days, where leaching continues but is at a slower rate than at the beginning of the test. For the remaining duration of the test, most species show only minor changes in concentration.

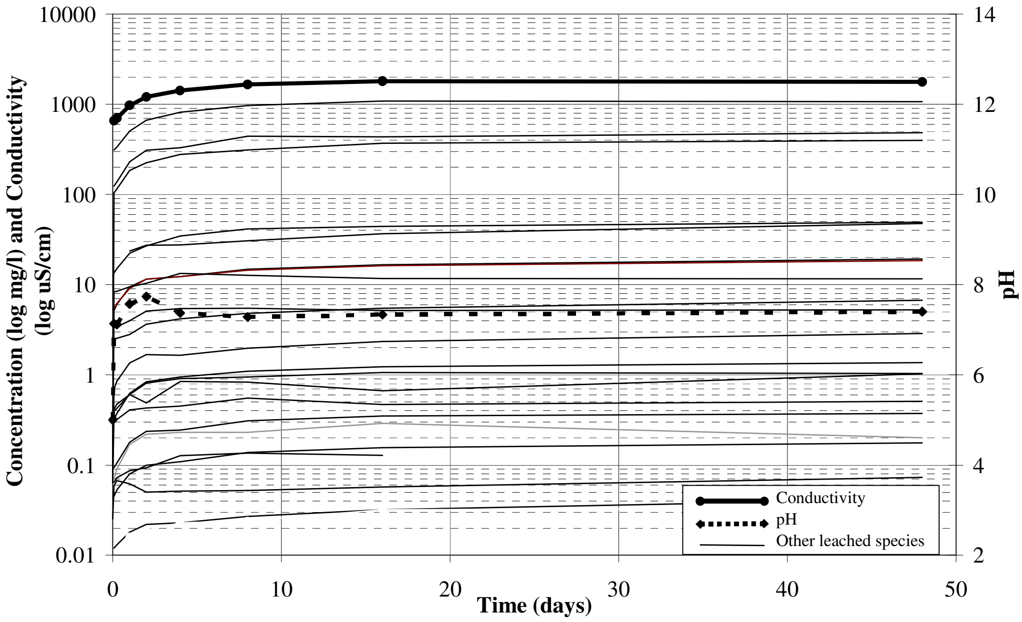


Figure 4-6: Typical change in species concentrations with time in the tank-leaching test on materials at realistic gradings (example shows the sewage sludge incinerator ash)

The change in leachate species concentration over the first 24 hours of the tank-leaching test is shown in more detail in Figure 4-7 (again, for the sewage sludge incinerator ash test). From Figure 4-7 the very rapid initial increase in concentration can be clearly seen for most species. This initial period of leaching increases species concentrations by anything between 0.1 and 3 or more orders of magnitude. From this data, it appears that the most rapid rise in concentration occurs during the first 2 hours of the test. However, because the first sample of leachate was not taken until 2 hours into the test, this data is probably skewed and the initial leaching is likely to have occurred even more rapidly than it appears from Figure 4-7. Such rapid initial leaching occurs as a result of the wash-off and dissolution of species from the surface of the material particles. Such surface species are readily available for leaching and are aided by the high concentration gradients that exist between the test material and the leachant at the beginning of the test. Rapid initial leaching was also shown in the rapid leaching characterisation tests.

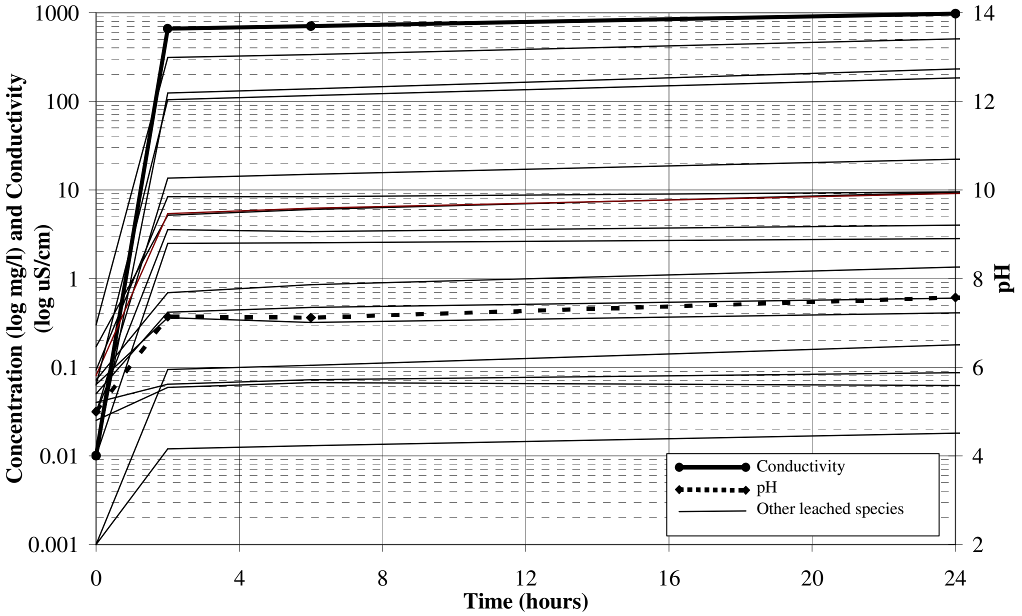


Figure 4-7: Typical change in species concentrations during the first 24 hours of a tank-leaching test on material at realistic grading (example shows the sewage sludge incinerator ash)

From Table 4-6 it can been seen that a significant proportion (about half) of the species are at concentrations within the same range as the ultimate values after a period of only 24 hours. The effect on species concentrations of this first period of leaching is more significant than the following 63 days, although it is in this latter period that diffusive leaching may become more significant.

The next phase of leaching covers the period up to between 1 and 4 days. During this time, leached concentrations generally continue to increase but at a slower rate than experienced in the initial stage of the test. It is during this phase of the test that most species reach concentrations that reflect the order of magnitude of the ultimate values. This phase of the leaching may be attributed to continued surface wash-off and also to diffusion processes. The rate of leaching may be affected by two factors, firstly, the availability of species from the solid and, secondly, the ability of the leachant to accept further species. The availability of species from the solid is influenced by boundary and leached layer phenomena, and by the rate of diffusion, which is potentially very slow. The concentration gradient that now exists between the solid and liquid phases is considerably lower than at the beginning of the test. The ability of the leachant to take on more species has decreased as many of the "sites" that were available for leached species are now occupied and some species are approaching limits of solubility.

For the remaining duration of the test, most species show only minor changes in concentration. During this phase of the test any leaching is largely by diffusion, although this process is slowed by the low concentration gradient between the solid and leachant and the impending solubility limits of a number of species. In this later stage of the test both increases and decreases in species concentrations are observed. Using a similar test, but with agitation to accelerate the leaching process, Bialucha (2000) observed similar decreases in concentrations in the later stages of a limited number of tests.

The pH of all the leachates in the tank-leaching tests on the aggregates at realistic gradings are shown in Figure 4-8. The figure clearly illustrates that the largest pH adjustment, into the alkali region, occurred for the tests on the MSW incinerator ash and the pulverized-fuel ash. For the blastfurnace slag, minestone and china clay sand the change in pH is a linear function of the log of time (R^2 of 0.98, 0.98 and 0.93 respectively). This relationship shows that leaching has continued over the duration of the test but that the rate of leaching has reduced in a logarithmic pattern. For the other materials the pattern of pH shows a more rapid increase in the initial stages of the test leading to a plateau in the later part of the test. This pattern may indicate that the leaching of species that define the pH have either reached solubility limits in solution or are restricted by availability.

A similar graph of the change in leachate conductivity with time is shown in Figure 4-9. From Figure 4-9 it can be seen that the highest conductivity leachates are generated from the test on the MSW incinerator ash. The lowest conductivity leachates are those from the china clay sand and limestone and granite primary aggregates. Most materials exhibit an increasing conductivity for the first 4 to 16 days of the test and then show a levelling or reduction in conductivity to the end of the test. Again, this illustrates that leaching is limited either by availability or solubility.

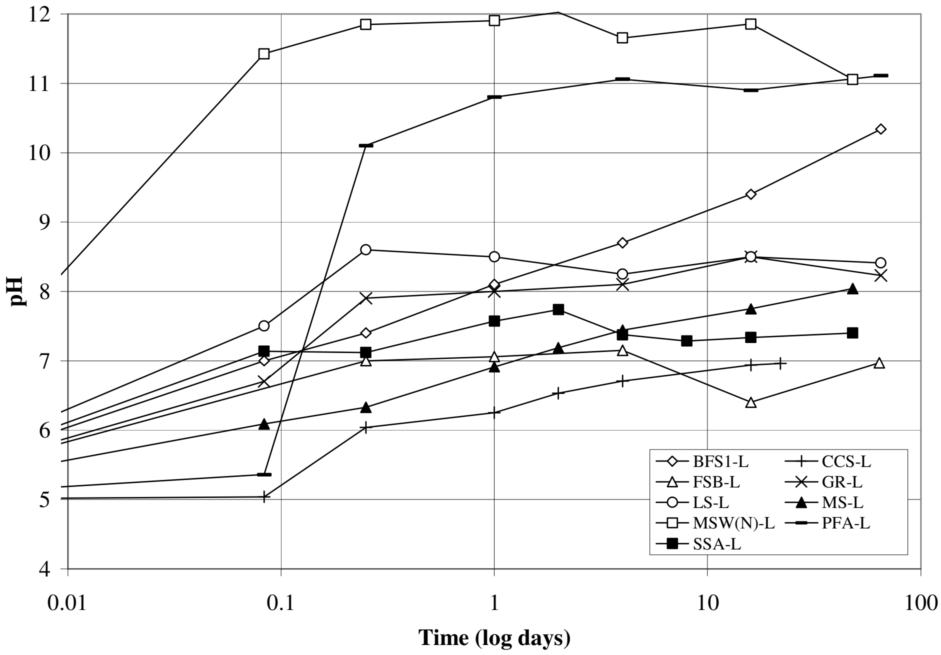


Figure 4-8: pH as a function of time for the tank-leaching tests on aggregates at realistic gradings

The foundry sand (phenolic, brass) shows an unexpected pattern of conductivity, which is reflected in the leaching of the alkali and alkali earth metals. The maximum conductivity and concentrations are shown in the first sample of leachate (which in this case represents a testing duration of 6 hours), after which a reduction in these values is observed. One conceivable explanation is that the binders used to hold the sand in the casting moulds (which in this case are alkali phenolic binders) are rapidly washed from the surface of the sand particles. After this time the remaining sand, which is relatively inert, contributes insignificant quantities to the leachate. The reactive chemistry of the phenolic compounds, particularly with chlorine, sulphur and nitrogen, may also contribute to any explanation of the observed trends.

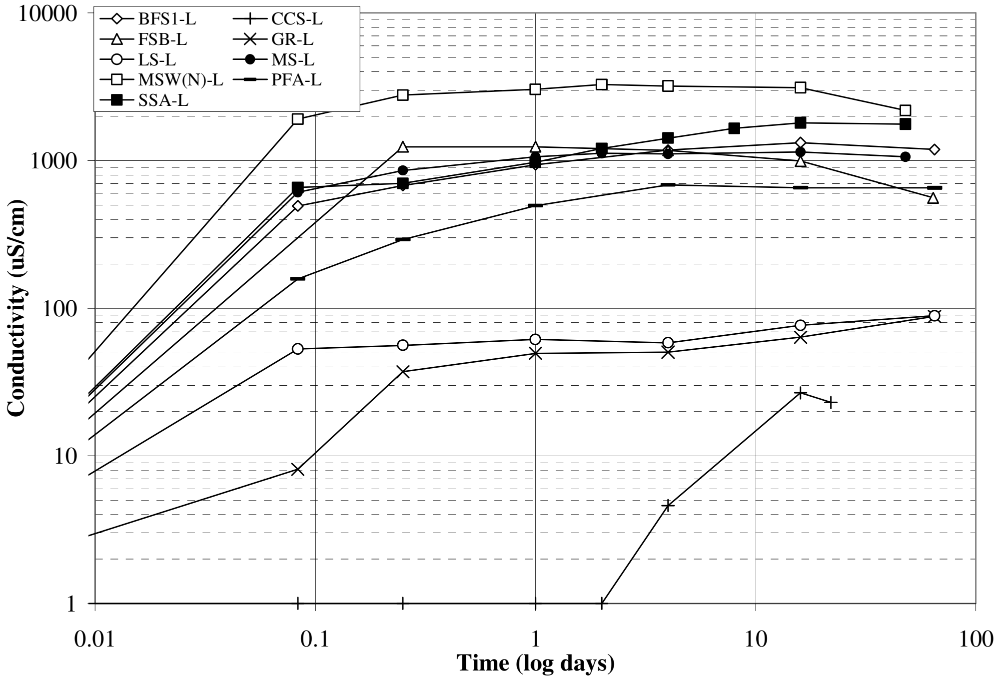


Figure 4-9: Conductivity as a function of time for the tank-leaching tests on aggregates at realistic gradings

In general, the species that showed high mobility in the rapid leaching characterisation test, namely the alkali and alkali earth metals, leached rapidly in the first few hours of the tank-leaching tests. It is thought that the lower mobility species maintain a relatively higher rate of leaching (but low absolute quantities) for a longer duration as the leachate is not so saturated with such species. This phenomenon is illustrated in Figure 4-10 for the leaching of silicon. It is not possible to confirm similar trends for other low mobility species since most such species are the transition metals, which were at concentrations below the limits of analytical detection in many of the leachates.

Similarly, it is also not possible to confirm whether leaching is pH controlled in the tank-leaching test, because many of the transition elements (for which solubility is most affected by pH) were below the limits of analytical detection.

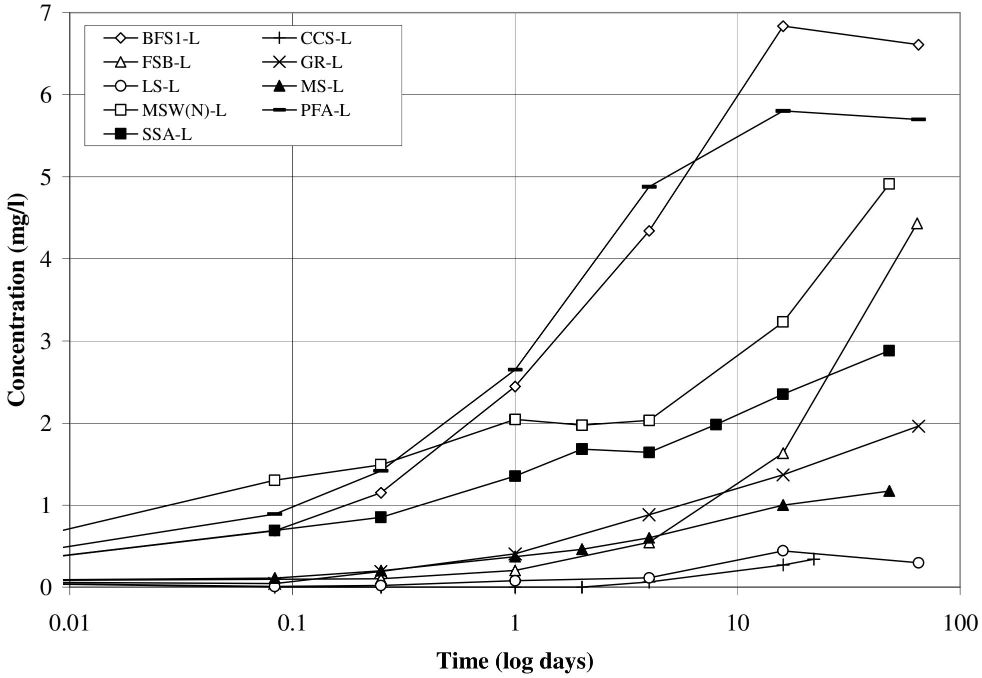


Figure 4-10: Leaching of silicon as a function of time for the tank-leaching tests on aggregates at realistic gradings

Identification of the elements within the different groups of the periodic table reveals that the species within each group generally exhibit similar patterns of leaching from a material. This trend could be used to predict leaching patterns of other species within a group based on a more complete knowledge of one species and limited information about the species that is to be predicted.

4.4.6 Discussion

4.4.6.1 Normalisation of the Data from the Tank-Leaching Tests on Materials at Realistic Gradings

The results from the tank-leaching tests on materials at realistic gradings have so far been described in terms of the absolute concentrations of species in the leachates. To enable the data to be weighted against the relative toxicity of the different species, the test data has again been normalised to the water quality maxima using the same method as described in Section 3.4.6 for the normalisation of the rapid leaching characterisation test data. The normalisation of the leaching test data against such criteria is, again, used here purely as a tool for interpretation. It is not suggested that such an assessment should form the basis of any examination of material acceptability or not.

The normalised data from the tank-leaching tests on materials at realistic gradings is presented in Table 4-7. As discussed in Section 4.4.3, the maximum concentration of a species within a test did not always occur in the last sample from that test. Therefore, the normalised concentrations listed in Table 4-7 represent the maximum concentration measured for that species at any time during the test. It can be seen from Table 4-7 that in most leachates the majority of species are present in concentrations below the water quality maxima. However, all of the materials, except the china clay sand and limestone, produce leachates with at least one species with a normalised concentration greater than unity. The foundry sand (phenolic, brass), MSW incinerator ash, pulverized-fuel ash and sewage sludge incinerator ash all contain species at more than 10 times the wqm, and for the latter three materials, those species include at least one from List II.

Table 4-7: Normalisation of data from tank-leaching tests on aggregates at realistic gradings to arbitrary water quality maxima

Aggregate	**Test**	**L/S**	**Maximum normalised concentration in leachate**					
			Detected <1	**1-10**		**10-100**		**>100**
				II	**Other**	**II**	**Other**	
BFS	**L**	**10**	Mg Na Cl NO_2 F Ba Zn Li B	NH_4	Ca K SO_4 Mn Al			
CCS	**L**	**10**	Ca Mg Na K SO_4 F P NH_4 Ba Mn Cu Zn As					
FSB	**L**	**10**	Ca Mg Na SO_4 P Mn Al Cu Zn	NH_4	NO_2 Fe		K	
GR	**L**	**10**	Ca Mg Na K Cl SO_4 NO_3 F NH_4 Ba Mn Al Zn		NO_2			
LS	**L**	**10**	Ca Mg Na K Cl SO_4 NO_3 NO_2 F NH_4 Ba Mn Zn					
MS	**L**	**10**	Ca Mg K Cl SO_4 F Ba Ni Cu Zn Cr Li B As	NH_4	Na NO_2 Mn Fe			
MSW(N)	**L**	**10**	Cl NO_3 F P Ba Mn Ni Zn Cr Li As	NH_4 Mo Pb	Ca Na SO_4 NO_2	Cu	K Al	
PFA	**L**	**10**	Ca Mg Na Cl SO_4 NO_3 NO_2 F P Ba Fe Zn Li B	NH_4 Cr As	K	Mo V	Al	
SSA	**L**	**10**	Mg Na Cl F P NH_4 Ba Mn Cu Zn Li B As	V	Ca K SO_4 NO_2 Al	Mo		

L = tank-leaching test on material at realistic grading

Again, as described in Section 3.4.6 for the rapid leaching characterisation test data, an indication of the overall quality of the test leachates was obtained by summing the individual normalised species concentrations. For the same reasons described in this earlier section, two indicators of leachate quality were used- firstly, the total sum of all the normalised species, and secondly, the sum of the List I and II species. Similarly, the maximum potential concentrations (i.e. 0.1 for a concentration of <0.1) were used in both summations. Graphical representations of these "toxicity indices" are given in Figure 4-11 and Figure 4-12, respectively.

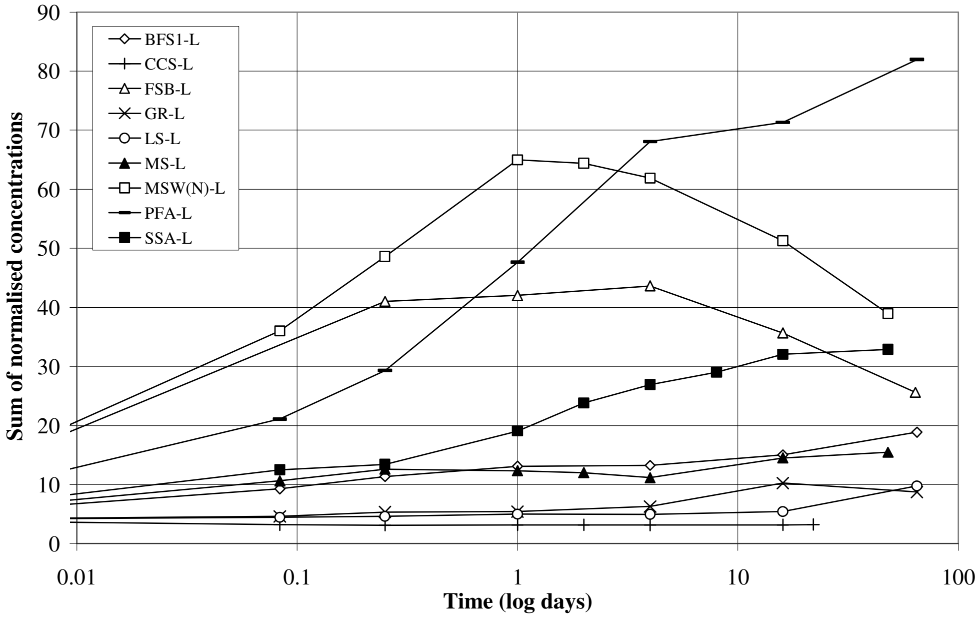


Figure 4-11: Sum of all normalised species concentrations from tank-leaching tests on materials at realistic gradings

From Figure 4-11 it can be seen that the pulverized-fuel ash, MSW incinerator ash, foundry sand (phenolic, brass) and sewage sludge incinerator ash show the highest total normalised concentration. These materials exhibit two different patterns of leaching. The first, which is demonstrated by the pulverized-fuel ash and sewage sludge incinerator ash, shows the leached concentrations increasing throughout the duration of the test. Although it cannot be concluded, this similarity in leaching pattern may be a function of the fine particle size of both materials or of the incineration processes from which the materials originate (which may encapsulate the species in a glassy matrix). The MSW incinerator ash and foundry sand (phenolic, brass) show a second pattern of leaching involving a peak in concentrations before the 4th day, or so, of the test, followed by a decline in concentration for the remainder of the test.

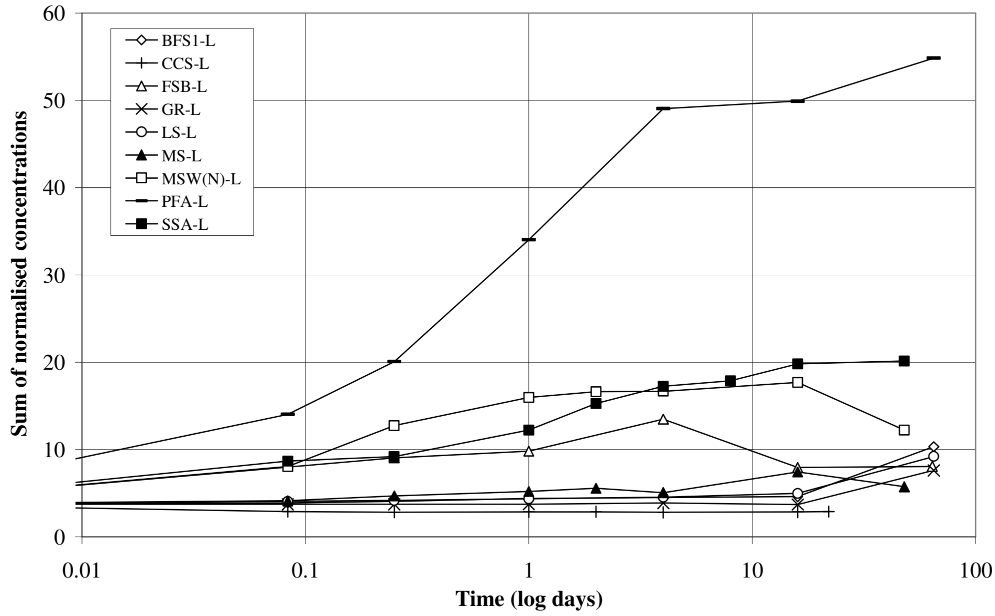


Figure 4-12: Sum of normalised species concentrations of List I and II species from tank-leaching tests on materials at realistic gradings

A comparison between Figure 4-11 and Figure 4-12, generally shows that the pattern of total leaching is reflected by the leaching of List I and II species. These latter species only contribute between 30% and 70% of the total, with the remaining proportion mainly resulting from species such as sodium, potassium, calcium, magnesium and chlorine.

The data shown in Table 4-7 was compared with the equivalent data from the C2-10 phase of the rapid leaching characterisation tests, as presented in Table 3-14. Such a comparison can only be made between these two different types of test because both establish a L/S ratio of 10. Such a comparison cannot be made between tests with differing L/S ratios since the species would be concentrated at a lower L/S ratio and diluted at a higher L/S ratio (except for those species that are limited by solubility). The comparison between the results of these two tests revealed some unexpected results. All of the materials, with the exception of the china clay sand and limestone, contained at least one species at higher concentrations than in the rapid leaching characterisation test on the same material. Equally, the tank-leaching test resulted in significantly lower concentrations of at least one species in the tests on the blastfurnace slag, granite, limestone, MSW incinerator ash and pulverized-fuel ash.

The tank-leaching test data was examined to determine whether the inflated concentrations in these leachates occurred at a time greater than the 1 day for which the rapid leaching characterisation test had been performed. In many instances this was not the case. It was therefore deduced that the increased leaching of a limited number of species was a function of the difference in the test methods.

A more detailed interpretation of the tank-leaching test data in relation to the rapid leaching characterisation test data is given in the following section.

4.4.6.2 Effect of Material Particle Size on Leaching

To infer the effect of material particle size on material leaching, the results of the tank-leaching tests on materials at realistic gradings were compared to the results of the rapid leaching characterisation tests for equivalent materials. A comparison for the pH and conductivity of the test leachates is provided in Figure 4-13 for the tank-leaching tests compared to the C2-10 phase of the rapid leaching characterisation test (L/S=10, which is the same as in the tank-leaching tests). It was expected that the materials would leach at a slower rate in the tank-leaching tests because of the larger particle sizes (smaller surface area to volume ratio) for granular materials in this test, and also because the leachant was not agitated.

The china clay sand and granite both show a higher conductivity in the rapid leaching characterisation tests but a similar pH in both tests. It is, however, observed that for both of these tests the measured values of conductivity are very low (<70uS/cm) and, from Figure 4-11, it can be seen that the total leached species are also very low. Such low values indicate that there are few species to be leached from either of these materials. If there are few species to be leached, then high concentrations will not result under any leaching conditions. This point is of interest on two levels. The first is that such materials are most unlikely to leach at concentrations of potential concern to the environment, and therefore, materials such as china clay sand can be considered as equivalent to primary materials. The second point concerns the interpretation of such low concentrations. A large percentage change to a low concentration only results in a small change in the absolute value. Since analytical errors are bigger for species at low concentrations, then these small absolute changes in concentration are potentially subject to large errors. Any trends observed in such data cannot therefore be guaranteed and should not be used to justify any theory. From Figure 4-11 and Figure 4-13, it can be seen that a similar condition is experienced for the limestone.

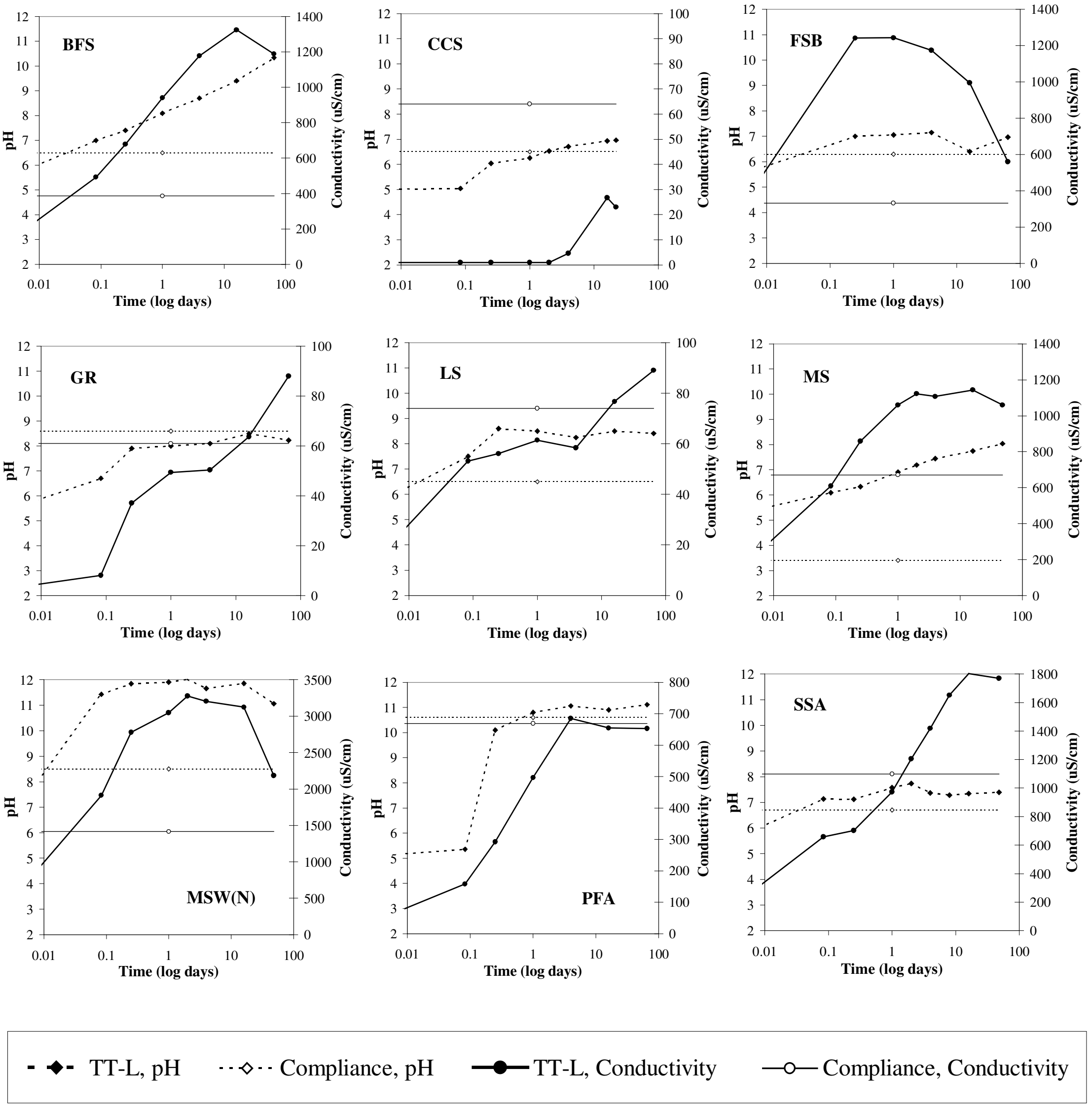


Figure 4-13: Comparison of the pH and conductivity of the test leachates from the tank-leaching test on materials at realistic gradings and the rapid leaching characterisation tests, plotted against time

For the six materials that have not yet been discussed, it can be clearly seen from Figure 4-13 that the pH and conductivity of the leachates from the rapid leaching characterisation tests are not always higher than the equivalent measures in the tank-leaching tests after 1 day. In fact, in many cases the reverse is true. For the blastfurnace slag, foundry sand (phenolic, brass), minestone and MSW incinerator ash the pH and conductivity shown in the tank-leaching tests are all higher than in the equivalent rapid leaching characterisation test. For the pulverized-fuel ash the value of pH is similar for the two tests, but the conductivity is higher in the rapid leaching characterisation test. For the sewage sludge incinerator ash the pH is higher in the tank-leaching test and the conductivity in the two tests are similar.

From these observations, it can be concluded that the pH and conductivity exhibited in a rapid leaching characterisation test cannot be used to predict the equivalent values that will result from a tank-leaching test on the same material at a realistic grading. This highlights some of the perceived limitations of using the rapid leaching characterisation test to assess materials in an "unnatural" condition.

To provide further evidence of the differences between the tank-leaching test and the rapid leaching characterisation test, the relationship between the concentrations leached during the two tests was considered. To simplify the data set, consisting of nearly 2000 data points, to a more manageable size the sum of the normalised concentrations were used as a tool to clarify the comparison. This data is presented in Figure 4-14 for the sum of all species and the List I and II species.

It is apparent, from observing Figure 4-14, that a different story is presented here from that shown by the leachate pH and conductivity illustrated in Figure 4-13. For all of the materials, except foundry sand (phenolic, brass) and, to a limited extent, minestone, the rate of total leaching observed in the tank-leaching tests is significantly lower than that shown in the rapid leaching characterisation test. Over time, however, the total concentration obtained in the rapid leaching characterisation test is reached in all tank-leaching tests except the MSW incinerator ash and china clay sand. It should also be noted that the fine-grained pulverized-fuel ash and sewage sludge incinerator ash, which did not require size reduction for the rapid leaching characterisation test, also demonstrated a reduced rate of leaching in the tank-leaching test. Different mechanisms may be in operation in these materials than those in the coarser graded materials.

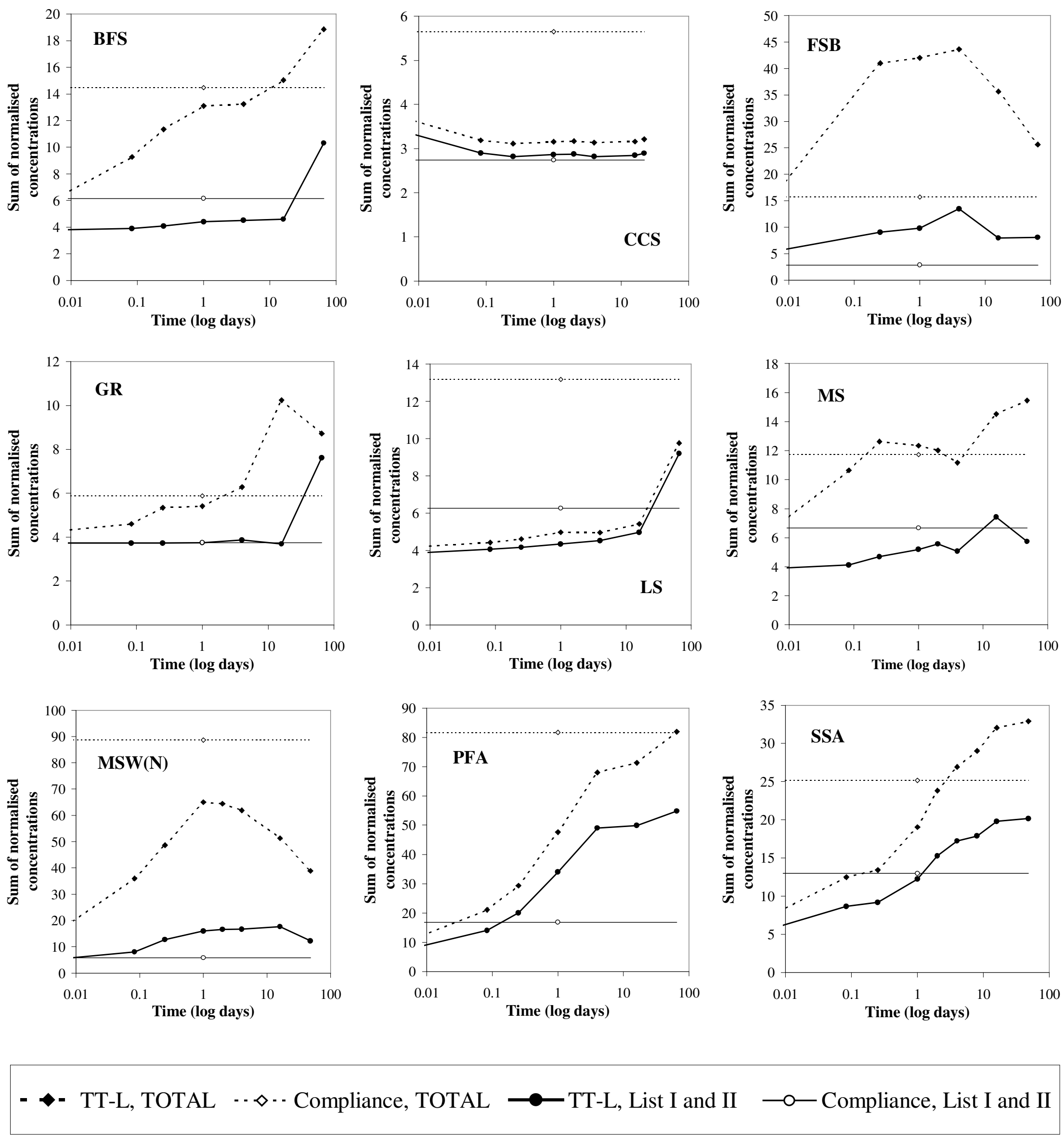


Figure 4-14: Comparison of the sum of the normalised species concentrations in the test leachates from the tank-leaching tests on materials at realistic gradings and the rapid leaching characterisation tests, plotted against time

The trends shown for the leaching of the List I and II species are not as clear as those shown for the total leached species. The blastfurnace slag, limestone and minestone all show slower rates of leaching in the tank-leaching test. It should be noted that these materials are all relatively coarse graded and would thus, have been most significantly influenced by the particle size reduction in the rapid leaching characterisation test. After 1 day of testing, the china clay sand, granite and sewage sludge incinerator ash exhibit similar normalised concentrations of List I and II species in the two types of test. Again, due to the very low species concentrations released by the china clay sand and granite, any data from these materials should be used cautiously. The foundry sand (phenolic, brass), pulverized-fuel ash and MSW incinerator ash all display more rapid leaching of the List I and II species in the tank-leaching test. For the MSW incinerator ash it may be possible to explain this trend from the inclusion, in the tank-leaching test, of the non-crushable fraction of the material, which was excluded from the rapid leaching characterisation test. This fraction largely consisted of fused metals that have the potential to contribute significantly to the leaching of List I and II species.

It can generally be concluded that the rate of leaching in the tank-leaching test on materials at realistic gradings is slower than that shown in the rapid leaching characterisation test for the same material. Leaching is however time dependant and for most materials the ultimate concentration of leached species exceeds that defined by the rapid leaching characterisation test. The time dependent nature of leaching is important when considering the road utilisation environment for these materials. The typically low permeability and drainability of compacted aggregates is likely to result in long residence times for the pore water, which may in turn result in high species concentrations, but small leachate volumes.

The slower rate of leaching, shown by most materials in the tank-leaching test, may indicate the effects on leaching of particle size and/or leaching vessel agitation. For the coarse materials, the lower surface area to volume ratio may account for such reduction. This theory is supported by the work of Bialucha (2000) who performed tank-leaching tests on different sized fraction of the same materials and concluded that the smaller fractions leached in higher concentrations than their coarser equivalent. The reduction in the rate of leaching shown for the fine-graded materials, which did not require size reduction for examination in the rapid leaching characterisation test, may be more of a reflection of the lack of agitation in the tank-leaching test. The "heaped" material sample used in this test may begin to act more like a monolith than a granular material, thus reducing the material permeability and the contact between the leachant and the material grains near the centre of the heap.

There is no conclusive evidence that leaching in the tank-leaching test is controlled by the pH of the leachate.

4.4.7 Conclusions

- The tank-leaching test that was developed for use in this research proved simple to perform and showed good repeatability for testing uncompacted materials at realistic gradings.

- Leaching is generally rapid at the beginning of the test as a result of surface wash-off from the material particles. A period of slower leaching follows in which diffusive leaching begins to contribute. For most species, concentrations in the vicinity of the ultimate values are observed after between 1 and 4 days testing. A test duration of 4 days is therefore likely to be sufficient for material assessment in this way.

- Leaching in the tank-leaching test on materials at realistic gradings generally occurs at a slower rate than that experienced in the rapid leaching characterisation test on similar materials. This fact was illustrated by comparison of the leached concentrations after 1 day of leaching. Leaching in the tank-leaching test is time dependant with ultimate concentrations generally exceeding those in the rapid leaching characterisation test. This slower rate of leaching may be attributed, principally, to the larger particle sizes of the materials in the tank-leaching test and/or the non-agitation of the leachate. PH is not thought to be a significant influence over the total leached quantities. Measures of pH and conductivity did not provide a good representation of the overall quantity of leached species.

- This section provided a first attempt at testing the materials in the physical conditions that represent those of the in-situ usage environment of the road construction. The next section of this chapter continues by investigating the effect of material compaction on the rate and quantity of leaching.

4.5 Diffusive Leaching from Compacted Materials

To investigate the effect of material compaction on diffusive leaching the tank-leaching test was again utilised. The samples of the chosen materials were prepared to realistic degrees of compaction and then cured.

4.5.1 Methodology

The tank-leaching test methodology that was developed to investigate diffusive leaching from compacted materials is described in Box 4-3 and Box 4-4 below.

The method of compaction that was chosen for preparing the test samples needed to replicate typical compaction found in the field. Many standard laboratory methods exist for compacting aggregate materials. Possible compaction devices include the proctor hammer or vibrating hammer, and compaction can be carried out in one or more layers. A cylindrical sample, rather than a cubic sample, was preferred because of the relative ease of manufacture of such samples. The size of the sample needed to be large enough to contain the particle sizes being considered, whilst also being small enough to be contained in the leaching vessel along with the relevant quantity of leachant. For consistency and simplicity compaction in a single layer was preferred. The chosen method of sample compaction used a standard vibrating hammer [BS 5835, 1980] along with a special mould developed by Nunes (1997). The compaction method had been designed to achieve a density of at least 95% of the maximum dry density (MDD), as determined using the BS5835 procedure. The compaction method uses cylindrical moulds with an internal diameter of 150mm and a height of 70mm. The moulds are made from sections of plastic piping which make them cheap and easy to manufacture in the required quantities. The metal base plate of the compaction rig offered a potential source of cross-contamination to the material sample, especially as abrasion may occur during compaction. To reduce the contact a piece of plastic sheet, cut to size, was placed in the bottom of the mould before adding the material. A schematic diagram of the compaction apparatus is shown in Figure 4-15.

The method of sample compaction, developed by Nunes (1997), was for use on stabilised (treated) materials. Initial compaction trials were carried out to determine whether the unstabilised material samples would be durable enough to handle. A number of the trial samples partially collapsed when they were removed from the moulds. To overcome this problem the samples required some additional support to hold them together once they had been removed from the mould. A geotextile sleeve was developed for this purpose. A sheet of fine geotextile (Terram 500) was formed into a cylindrical tube that could line the inside of the plastic sample mould. The geotextile was joined by using a soldering iron (with a large head to distribute the heat) to partially melt the fabric which enabled it to be joined together. A fine geotextile was chosen as it is suitably strong but also highly permeable and therefore minimising the disruption to the contact between the leachant and the sample.

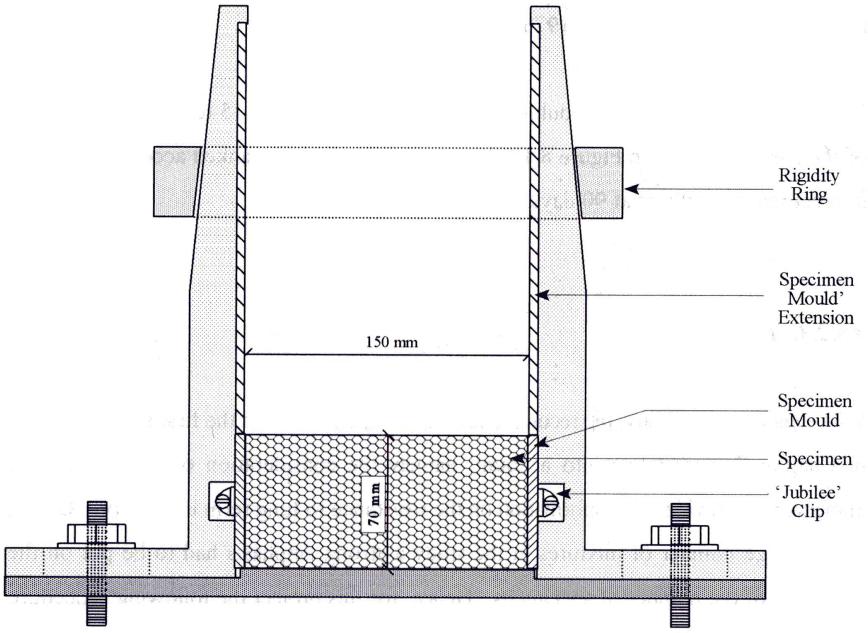


Figure 4-15: Schematic diagram of the compaction rig used to prepare the compacted tank-leaching test samples (from Nunes, 1997)

After being compacted the test specimens were stored in sealed plastic bags (to prevent loss of moisture) and cured for 90 days before being tested. Curing was considered necessary as some of the materials have limited hydraulic binding properties. Any strength development (and associated chemical reactions) that may result from the hydraulic binding needs to have occurred before testing. It was also considered important to subject the compacted materials to the same conditions as the bound materials that were to be tested later in the research (See Chapter 6). Nunes (1997) concluded that the majority of strength development had occurred by 90 days of curing and therefore this duration was considered appropriate, whilst also being reasonably practical.

Three compacted samples of each material were prepared. Since the samples were cured for 90 days it was considered important to make sure a suitable sample would be available at the end of that time. Preparing three samples allowed for any problems in the compaction and possible breakage when removing the samples from the moulds.

Box 4-3: Sample preparation methodology for the tank-leaching test on compacted materials

1) *Determine the maximum dry density (MDD) and the optimum moisture content (OMC)* of the test material [BS 5835, 1980].
2) *Obtain a representative sub-sample* of the test material by using coning and quartering [BS812: Part 102, 1989; BS EN 932-1, 1997]. Using the MDD determined in Step 1, calculate the dry mass of material required to fill 6 sample moulds (150mm ∅ x 70mm). Sub-sample approximately this mass of material to form the laboratory sample. If the material is wet, a larger sample should be taken.
3) *Determine the residual moisture content* of the material in the laboratory sample by oven drying a sub-sample to constant weight [BS 812: Part 109, 1990; BS EN 932-1, 1997].
4) *Mixing the laboratory sample to the OMC for compaction.* Weigh the whole laboratory sample and calculate the dry mass of the material and the mass of residual moisture in the sample using the moisture content determined in Step 3. Calculate the mass of distilled water to be added to the laboratory sample to achieve the OMC. Weigh the distilled water and thoroughly mix into the laboratory sample. Store this sample in a sealed plastic bag until ready to compact.
5) *Prepare the sample moulds.* Prepare 4 cylindrical sample moulds (150mm ∅ x 70mm) by cleaning and rinsing in distilled water leachant. Assemble cylindrical geotextile mould liners and wash in distilled water leachant and air-dry before use.
6) *Compact the test specimens*. Clean the compaction apparatus and assembled as described by Nunes (1997). Place a circle of plastic sheeting over the base plate of the apparatus and position the cylindrical geotextile liner inside the sample mould. Using the MDD (determined in Step 2) and the moisture content of the material (established in Step 4), calculate the mass of wet material required to fill one mould. Weigh this mass of sample and placed into the compaction apparatus. Compact the sample for 3 minutes using a standard vibrating hammer [BS 5835, 1980]. Observe the sample and determine whether it is oversize or undersize and adjust the mass of material in further samples accordingly. Remove this sample from the mould and determine the moisture content of the compacted specimen, as described in Part 3. Prepare three further compacted samples.

7) *Cure the compacted samples.* Seal each sample inside a plastic bag to prevent loss of moisture and leave to stand for 90 days. This period of curing should enable any self-binding reactions to occur, as shown by Nunes (1997).
8) *Prepare the material sample for testing*. After curing, remove the test sample from the mould using a hydraulic extraction machine. Weigh the sample and determine the dry mass of material and the mass of water in the sample using the compacted moisture content determined in 6.

The tank-leaching test methodology employed for testing the compacted materials is similar to that used for the materials at realistic grading. The differences are given in Box 4-4 with reference to Box 4-2.

Box 4-4: Methodology for the tank-leaching test on compacted materials

1) *Determine the volume of leachant required* to establish a liquid to solid volume ratio of 10. The total mass of leachant required is equal to 10 times the volume of the test sample (150mm ∅ x 70mm). The mass of leachant to be added to the leaching vessel is calculated by subtracting the mass of moisture held within the test sample (see Box 4-3, Part 8) from the total volume of leachant that is required.
2) *Follow steps 2 to 6* from Box 4-2.

The leachates generated from the tank-leaching tests on compacted materials were analysed, where possible, for the 37 determinants that are listed in Table 3-9.

4.5.2 Tank-Leaching Tests on Compacted Materials

The tank-leaching test on compacted materials was carried out on the 9 materials listed in Table 4-8. The materials were selected to cover the range of gradings and process sources. They are generally a continuation of those already tested at realistic gradings in the tank-leaching tests with the addition of the crumbed rubber that could not be tested uncompacted as it floated on the leachant.

The blank tank-leaching test, as described in Section 4.4.3, applies equally to the compacted specimen testing. Again, the data from this test (as presented in Table 4-4) was used to quantitatively adjust the concentrations of species in the leachates from the compacted material tests. The initial concentration of the leachate in the blank test was also again used as the "zero hours" concentrations for the material tests. Some small error may result from the use of this blank leaching test data for the quantitative adjustments. This is because the blank was carried out with 20 litres of leachant, thus representing the ideal L/S ratio for the tests on materials at realistic gradings. In the tests on the compacted materials, a leachant volume of approximately 13 litres was used in most cases. Therefore, any species resulting from the blank will be concentrated in this smaller volume of leachant and thus result in high species concentrations in the solution. However, because the applied corrections are generally small compared to the total, it was deduced that the use of this data would not contribute significant error.

Table 4-8: Summary of the tank-leaching tests on compacted materials

Material	Test Abbreviation	Material Grading	Material Source
Air-cooled blastfurnace slag	BFS-C	DoT Type 1	Metalliferous slag
China clay sand	CCS-C	Medium	Processed rock
Crumbed rubber (2-8mm)	CR-C	Medium	Processed vehicle tyres
Foundry sand (phenolic, brass)	FSB-C	Fine	Treated sand
Granite	GR-C	DoT Type 1	Primary aggregate
Limestone	LS-C	DoT Type 1	Primary aggregate
Minestone	MS-C	Medium to coarse	Processed rock
MSW incinerator ash (New incinerator)	MSW(N)-C	Medium	Incinerator residue
Pulverised fuel ash	PFA-C	Fine	Power station fly ash

The tank-leaching tests on compacted materials were generally performed with few problems. The most significant problem again resulted from the breakage of a leachate sample container during transportation or storage prior to analysis. In this case, a number of species could not be determined for the 64 day test from the crumbed rubber.

When the test materials were compacted into the moulds, a number of samples were a few millimetres undersize or oversize, even after the mass of material had been appropriately adjusted. When establishing the volumetric L/S ratio of 10, the measurements of such samples were taken into account.

4.5.3 Results

The physical properties and test conditions demonstrated in the tank-leaching tests for compacted materials are summarised in Table 4-9. In the tank-leaching tests on compacted materials, the aim was to maintain a consistent L/S ratio of 10 by volume. It can be seen that this was generally achieved. From Table 4-9, it can also been seen that the volumetric L/S ratio has, in most cases, resulted in a L/S ratio by mass of considerably less than the 10 used in the tank-leaching tests on materials at realistic gradings. As a result of this difference in L/S ratio (by mass) the concentration data from these two tests are not directly comparable. The lower L/S ratio in the tests on the compacted materials may artificially inflate the concentration of species in the solution.

Table 4-9: Summary of the physical properties and test conditions for the tank-leaching tests on compacted materials

Test	MDD Mg/m^3	OMC %	Mass of Test Sample Kg	Test L/S by volume	Test L/S by mass
BFS-C	2.01	6	2.510	10.0	4.93
CCS-C	1.93	10	2.181	10.7	6.07
CR-C	-	-	0.687	11.6	20.8
FSB-C	1.9	10	2.139	10.7	6.20
GR-C	2.2	7	2.847	10.8	4.68
LS-C	2.21	8	2.750	10.7	4.82
MS-C	1.72	10	2.003	11.2	6.89
MSW(N)-C	1.62	12	1.851	10.0	6.68
PFA-C	1.09	25	1.298	10.0	9.53

The concentrations of species in the leachates from the tank-leaching tests on compacted materials are summarised in Table 4-10. The species are tabulated within specified concentration ranges for leaching duration's of 6 hours (0.25 days), 1 day, 4 days, 16 days and at the end of the test. Those species listed in Table 3-9 that are not listed in Table 4-10, may be present in that leachate but are concentrations below the limits of analytical detection.

From Table 4-10 it can be clearly seen that the concentrations of the different species in solution vary over more than 5 orders of magnitude. This is the same range as presented for the tank-leaching tests on materials at realistic gradings in Table 4-6. In general, the species present in the highest concentrations are the same as those observed at the highest concentrations in the tank-leaching tests on materials at realistic gradings. A comparison between Table 4-10 and Table 4-6 reveals that, in general, each material exhibits the same detectable species in the leachates from both tests. The concentrations of these species generally fall within ±1 order of magnitude, of each other. The variation in L/S ratio between the two tests does however raise some complications over the comparison of the data. The comparison and associated complications are discussed in more detail later in this chapter.

Table 4-10: Summary of tank-leaching test data from compacted materials (concentrations in leachate)

Test	Time Days	pH	Cond. µS/cm	Concentration in leachate (mg/l)					
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**	**100-1000**	**>1000**
BFS C	**0.25**	7.5	974		NO_2 Mn Zn	Br F Sr Li B	Mg Si	Na K Cl	Ca SO_4 S	
	1	8.5	1447	Zn	Mn	Br Sr Li B	Mg F TIC NH_4 Si	Na K Cl	Ca SO_4 S	
	4	8.7	1863	Zn	Mn V	NO_2 TIC Sr Li B	Mg F	Na K Cl Si	Ca SO_4 S	
	16	8.8	2162	Zn		Br NO_2 Sr Mn Li B	Mg F TIC	Na K Cl Si	Ca S	SO_4
	64	9.2	2173	Zn		Sr Mn Al Li	Mg F B	Na K Cl Si	Ca S	SO_4
CCS C	**0.25**	6.6	0	Na Cl Mn Zn As Se	Ca	Mg SO_4				
	1	6.7	3.0	Na Cl Mn Zn Se	NH_4 Si	Ca Mg SO_4 F TIC S				
	4	7.0	32	Mn Se	Na Cl Zn	Mg K F S Si	Ca SO_4 TIC			
	16	7.1	40	Mn Zn As Se		Mg Na Cl F S Si Al	Ca K SO_4 TIC			
	22	7.4	40	Zn As Se		Mg Na Cl F S Si	Ca K SO_4 TIC			
CR C	**0.25**	6.8	8.1	Na	Mg Mn	Ca Cl SO_4 Br TIC P S Zn	K TOC			
	1	6.8	4.4	Na	Mg Mn	Ca K Cl SO_4 Br TIC P S Zn	TOC			
	4	7.1	29	Na	Mg P Si Mn	K Cl SO_4 Br S Zn	Ca TOC TIC			
	16	7.1	44	Na Sr Fe	Cl NO_3 NO_2 Ba Mn	Mg K SO_4 Br P Si	Ca TOC TIC S Zn	HCO_3		
	64	7.4	43	Na Sr	P Mn	Mg Si Ba	Ca K TOC S Zn			
FSB C	**0.25**	6.9	1794	Mn Zn	Sr Cu	Si	Ca Mg Cl NO_3 S NH_4	Na SO_4 TIC	K TOC	
	1	7.1	1749	Cl Sr Zn	Mn Fe Cu	Br Si	Ca Mg S NH_4	Na SO_4 TIC	K TOC	
	4	7.4	1581	Cl Sr Ni Zn Li	Mn Al Cu V	Br P Si Fe	Ca Mg S NH_4	Na SO_4	K TOC TIC	
	16	6.9	1238	Cl Sr Ni Li	Mn Cu Zn	Mg NO_3 Br P Fe Al	Ca S NH_4 Si	Na SO_4 TIC	K HCO_3 TOC	
	64	7.6	670	Cl Zn	Ca Mn Al Cu	Mg Fe	SO_4 S NH_4 Si	Na TIC	K HCO_3 TOC	
GR C	**0.25**	6.5	21	Sr	NO_2 Si Mn Zn	Ca Mg Na K Cl TIC S	NO_3			
	1	7.2	39	Zn	NO_2 Sr Mn	Na SO_4 S Si	Ca Mg K Cl NO_3 TIC			
	4	8.1	60	Zn	NO_2 Sr Mn Al	SO_4 TOC S Si	Ca Mg Na K Cl NO_3 TIC	HCO_3		
	16	8.3	72	Mn Zn	Br NO_2 Sr	S Si Al	Ca Mg Na K Cl SO_4 NO_3 TIC	HCO_3		
	64	8.2	95		Sr Zn	S Al	Mg Na K Cl SO_4 NO_3 Si	Ca HCO_3 TIC		

Table 4-10: Summary of tank-leaching test data from compacted materials (concentrations in leachate) (continued)

Test	Time Days	pH	Cond. µS/cm	Concentration in leachate (mg/l)						
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**	**100-1000**	**>1000**
LS C	**0.25**	8.8	54	Na Mn	F Sr Zn	Mg SO_4 NO_3 S Ba	Ca K Cl TIC	HCO_3		
	1	8.8	62	Mn	Si Sr Zn	Mg Na F P S Ba	K Cl SO_4 TIC	Ca HCO_3		
	4	8.6	74	Mn Zn	NO_2 Sr Al	Mg Na K NO_3 F TOC S Si Ba	Cl SO_4 TIC	Ca HCO_3		
	16	8.3	84		NO_2 Sr Al Zn	Mg Na K NO_3 F Si Ba	Cl SO_4 TIC S	Ca HCO_3		
	64	8.3	109	Zn	NO_2	Mg Na K Si Ba Sr	Cl SO_4 NO_3 F S	Ca HCO_3 TIC		
MS C	**0.25**	6.6	573	Se	Mn Zn	F TIC NH_4 Sr	Mg K Br	Ca Na SO_4 S	Cl	
	1	6.8	966	Zn Se	NO_2 Si Mn	F TIC NH_4 Sr	Mg K Br	Ca SO_4 S	Na Cl	
	4	7.1	1596	Zn Se	Al Li	F Si Sr Mn	Mg K Br TIC NH_4	Ca S	Na Cl SO_4	
	16	7.5	2002	Zn Se	Li B	NO_2 F Si Sr Mn	Mg Br TIC NH_4	Ca K HCO_3 S	Na Cl SO_4	
	49	7.6	1618	Zn Se	Li B	NO_2 F Si Sr Mn	Mg Br TOC NH_4	Ca K TIC S	Na HCO_3 Cl SO_4	
MSW (N) C	**0.25**	7.9	611	Se	Ba Zn	Br F NH_4 Si Sr Cu	TIC Al	Ca Na K HCO_3 SO_4 TOC S	Cl	
	1	10.1	979	Se	NO_2 Zn	Br F Sr Cu	TIC NH_4 Si Al	Ca Na K HCO_3 SO_4 TOC	Cl S	
	4	10.4	1440	Mn Zn Se	Ba Mo Li	Mg F Sr	Br TIC NH_4 Si Al Cu	Ca HCO_3 SO_4 TOC	Na K Cl S	
	16	10.7	1873	Mn Zn As Se	Ba Mo Li	Mg NO_2 F P Sr	Br TIC NH_4 Si Al Cu	HCO_3 TOC	Ca Na K Cl SO_4 S	
	63	9.5	1807	Zn As Se	Ba Mo V Li B	Mg NO_2 F P Sr	Br NH_4 Si Al Cu	HCO_3 TOC TIC	Ca Na K Cl SO_4 S	
PFA C	**0.25**	9.6	258	Se	NO_2 Ba Zn Mo V As	Br F Si Sr Li B	Cl TIC Al	Ca Na K HCO_3 SO_4 S		
	1	10.4	426		NO_2 Ba Zn Cr V As Se	Br F Sr Mo Li B	Cl TIC Si Al	Ca Na K HCO_3 SO_4 S		
	4	10.0	924		NO_2 Ba Zn As Se	F NH_4 Sr Cr Mo V	Br TIC Si Al Li B	Ca K HCO_3 Cl S	Na SO_4	
	16	10.9	1114	Zn	Ba As Se	F NH_4 Sr Fe Cr V	Br TIC Si Al Mo Li B	Ca K Cl	Na HCO_3 SO_4 S	
	63	10.5	930		NO_2 Ba Zn Cr As Se	Mg F P NH_4 Sr Mo V	Br TOC TIC Si Al Li B	Ca Na K HCO_3 Cl	SO_4 S	

Note: These species concentrations should not be directly compared to those from the tank-leaching tests on materials at realistic gradings because the difference in L/S ratio between the test methods may result in the concentration or dilution of species in solution.

In general, the patterns of leaching are similar to those shown for the tank-leaching tests on materials at realistic gradings. These patterns of leaching are depicted by the change in pH and conductivity, as illustrated in Figure 4-16 and Figure 4-17 respectively. During the test, initially a period of rapid leaching occurs. This is followed by a period of slower but continued release in which the ultimate concentration range is established for most species. In the test period, after about 4 to 16 days the change in concentration of most species is small. In a number of leachates, reductions in species concentrations are actually experienced during this period of the test.

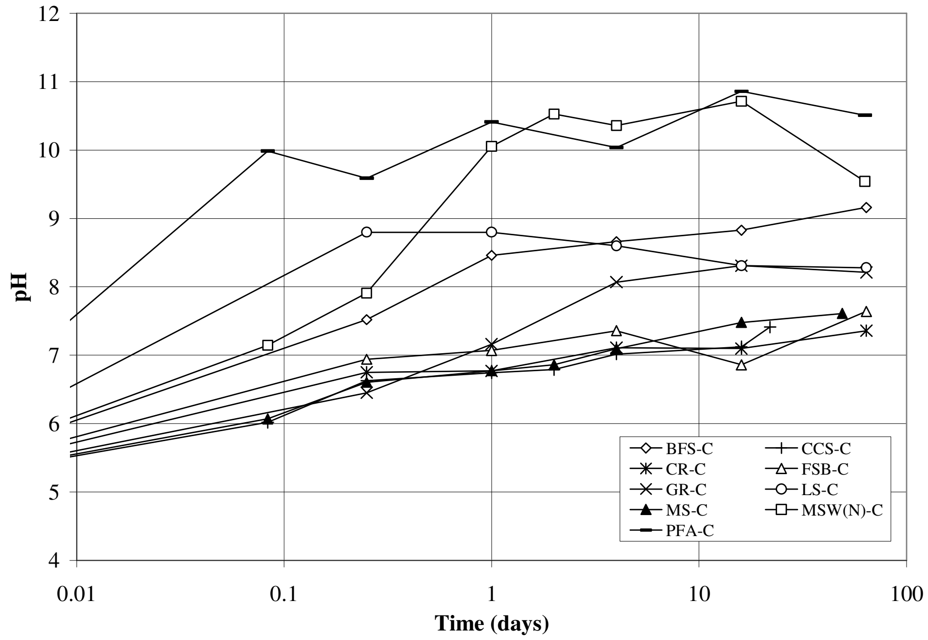


Figure 4-16: pH as a function of time for the tank-leaching tests on compacted aggregates

The results of the tank-leaching test on compacted foundry sand (phenolic, brass) again show the unusual trends that were observed in the test on the same material at realistic grading. This similar trend highlights the variability that exists between different materials and emphasises the need for a reliable assessment procedure that is applicable for all materials.

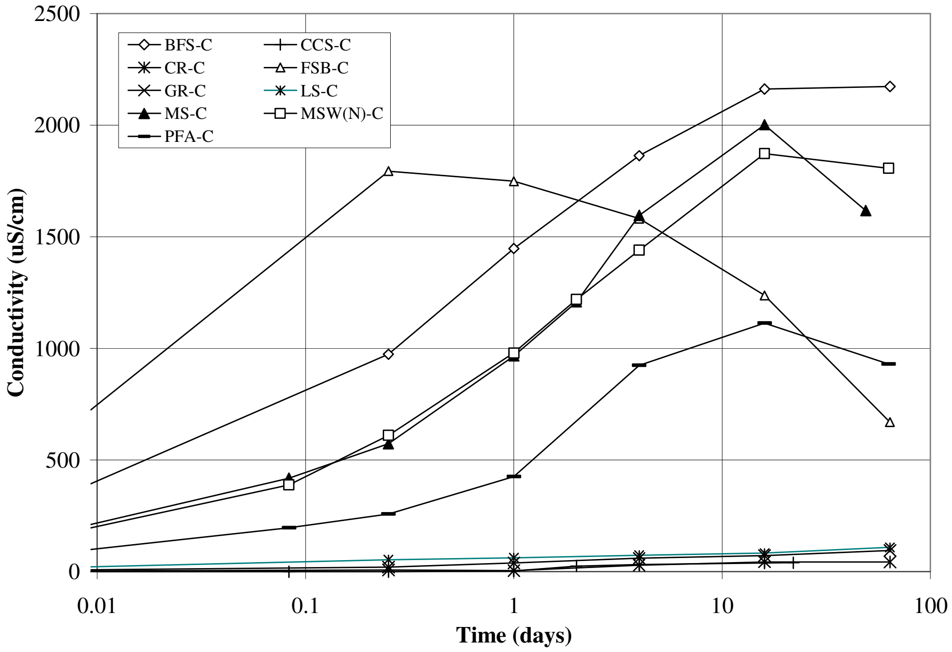


Figure 4-17: Conductivity as a function of time for the tank-leaching tests on compacted aggregates

4.5.4 Discussion

4.5.4.1 Normalisation of the Data from the Tank-Leaching Tests on Compacted Materials

To identify whether material compaction has had any relative effect on the rate of leaching from the different materials, the results of the tests on the compacted materials have, again, been normalised to the arbitrary water quality maxima as previously described in Section 3.4.6. A graphical representation of this normalised data is given in Figure 4-18, for the sum of all normalised species and Figure 4-19, for the sum of the List I and II species.

A comparison of Figure 4-18 with Figure 4-11, which shows the total normalised species for the leaching of the non-compacted materials, reveals that the total leaching of the minestone has reduced relative to the blastfurnace slag. A similar comparison of Figure 4-19 with Figure 4-12, which show the normalised concentrations of the List I and II species, also reveals a reduction in the leaching from the minestone, and also the MSW incinerator ash, relative to the blastfurnace slag. It is probable, therefore, that compaction has a greater effect on reducing the leaching of minestone and MSW incinerator ash, than it does on the blastfurnace slag. Thus, it is implied that the physical properties of the material have an effect on the degree to which leaching is modified by material compaction.

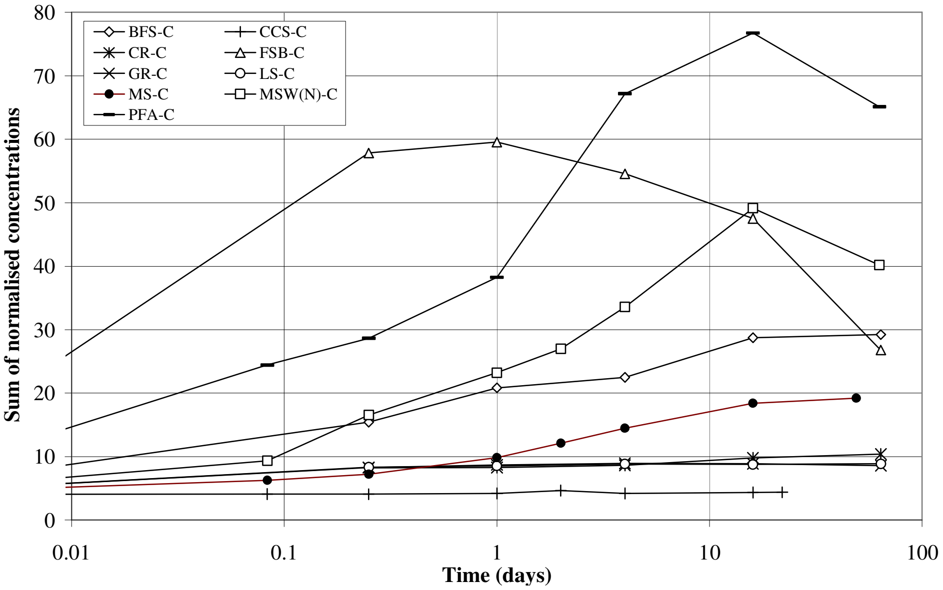


Figure 4-18: Sum of all normalised species concentrations from the tank-leaching tests on compacted materials

4.5.4.2 *Effect of Material Compaction on Leaching*

To determine the effect of material compaction on the rate or quantity of leaching, the results of the tank-leaching tests on compacted materials were compared to those from the tank-leaching tests on materials at realistic gradings. This comparison was possible for all materials except the crumbed rubber, for which a comparison was made to the results of the rapid leaching characterisation test.

When comparing the leachates generated from the two types of tank-leaching tests on the same material, the influence of the different L/S ratios used in the tests was addressed. As shown in Table 4-9, the L/S ratio, by mass, in the test on the compacted material was, generally, about half (L/S~5, by mass) of that established in the test on the material at realistic grading (uncompacted) (L/S~10, by mass). Therefore, for the same leached quantity (mg/kg) of a species, the leachates from the tests on the compacted material would exhibit a species concentration (mg/l) of about twice that shown in the equivalent test on the uncompacted material. A direct comparison of the species concentration in the two tests would, therefore, have given the inaccurate impression that leaching increased when, in fact, it remained constant. The interpretation of such data is further complicated by the possibility of leaching being controlled by species solubility.

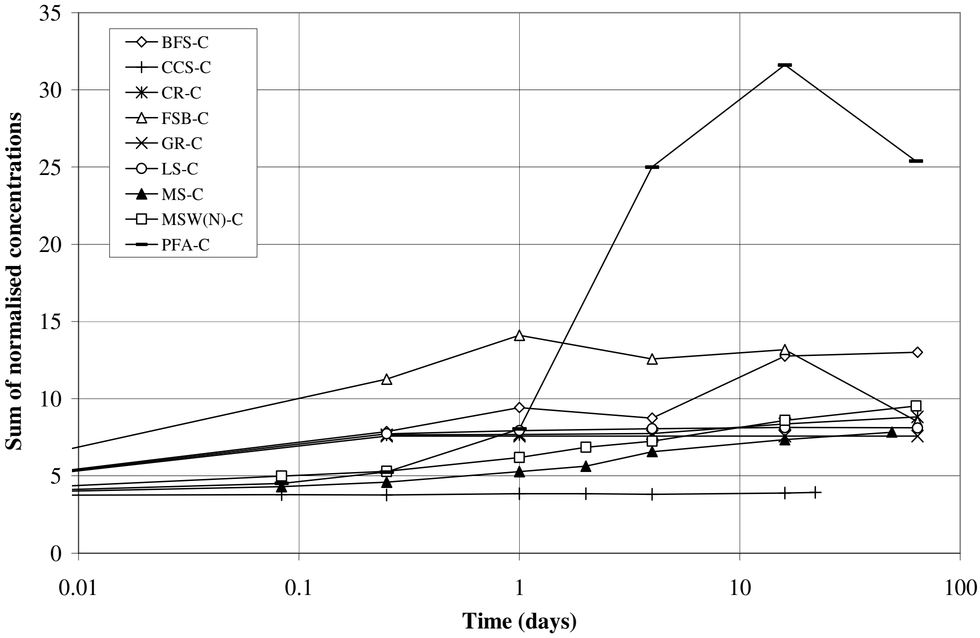


Figure 4-19: Sum of normalised species concentrations of List I and II species from tank-leaching tests on compacted materials

The possible relationships that may be deduced from a comparison between the data from the two types of tank-leaching test are summarised in Table 4-11. This table is only applicable to the situation where the L/S ratio (by mass) is greater in the test on the non-compacted material compared than in the test on the compacted material. The table shows that for a comparison based on either the species concentration (mg/l) or the leached quantity (mg/kg), the relationship in terms of the other units (i.e from mg/l to mg/kg, or mg/kg to mg/l) can be deduced. From this information it is then possible to draw conclusions about the effect of compaction on the leaching process.

To test the hypothesis that compaction will reduce the rate and quantity of leaching, it is necessary that the method of test comparison should be selected to enable this relationship to be identified. From Table 4-11 it can be seen that the only comparison that may enable such identification, is the comparison of species concentration. If such a comparison is inconclusive, then the leached quantity and the leaching behaviour identified from the rapid leaching characterisation tests can both be considered.

Table 4-11: Comparison of the data from the tank-leaching tests on materials at realistic gradings with that from the tank-leaching tests on compacted materials-Influence of variation in L/S by mass

Relationship	Deduced Relationship	Conclusion
Comparison of species concentration (mg/l), [X-L], [X-C]		
IF [X-L] > [X-C]	THEN X-L >X-C	AND compaction has reduced leaching
IF [X-L] = [X-C]	THEN X-L >X-C	AND EITHER compaction has reduced leaching, OR leaching is solubility controlled
IF [X-L] < [X-C]	THEN EITHER X-L > X-C OR X-L = X-C OR X-L < X-C	AND compaction may have increased or decreased leaching AND/OR leaching in X-C may be solubility controlled
Comparison of leached quantity of species (mg/kg), X-L, X-C		
IF X-L > X-C	THEN EITHER [X-L] > [X-C] OR [X-L] = [X-C] OR [X-L] < [X-C]	AND EITHER compaction has reduced leaching OR leaching is solubility controlled
IF X-L = X-C	THEN [X-L] < [X-C]	AND EITHER compaction has no effect on leaching OR compaction has increased leaching and X-C is solubility controlled
IF X-L < X-C	THEN [X-L] < [X-C]	AND compaction has increased leaching

[X-L] = species concentration (mg/l) in the leachate from the tank-leaching test on material at realistic grading
[X-C] = species concentration (mg/l) in the leachate from the tank-leaching test on compacted material
X-L = leached quantity of species (mg/kg) in the leachate from the tank-leaching test on material at realistic grading
X-C = leached quantity of species (mg/kg) in the leachate from the tank-leaching test on compacted material

The pH and conductivity of the leachates from the tank-leaching test on materials at realistic gradings and the tank-leaching test on compacted materials are compared for each material (with the exception of the crumbed rubber) in Figure 4-20. The tank-leaching test on the compacted crumbed rubber, included in Figure 4-20, has been compared to the crumbed rubber (1-4mm) data from the rapid leaching characterisation test.

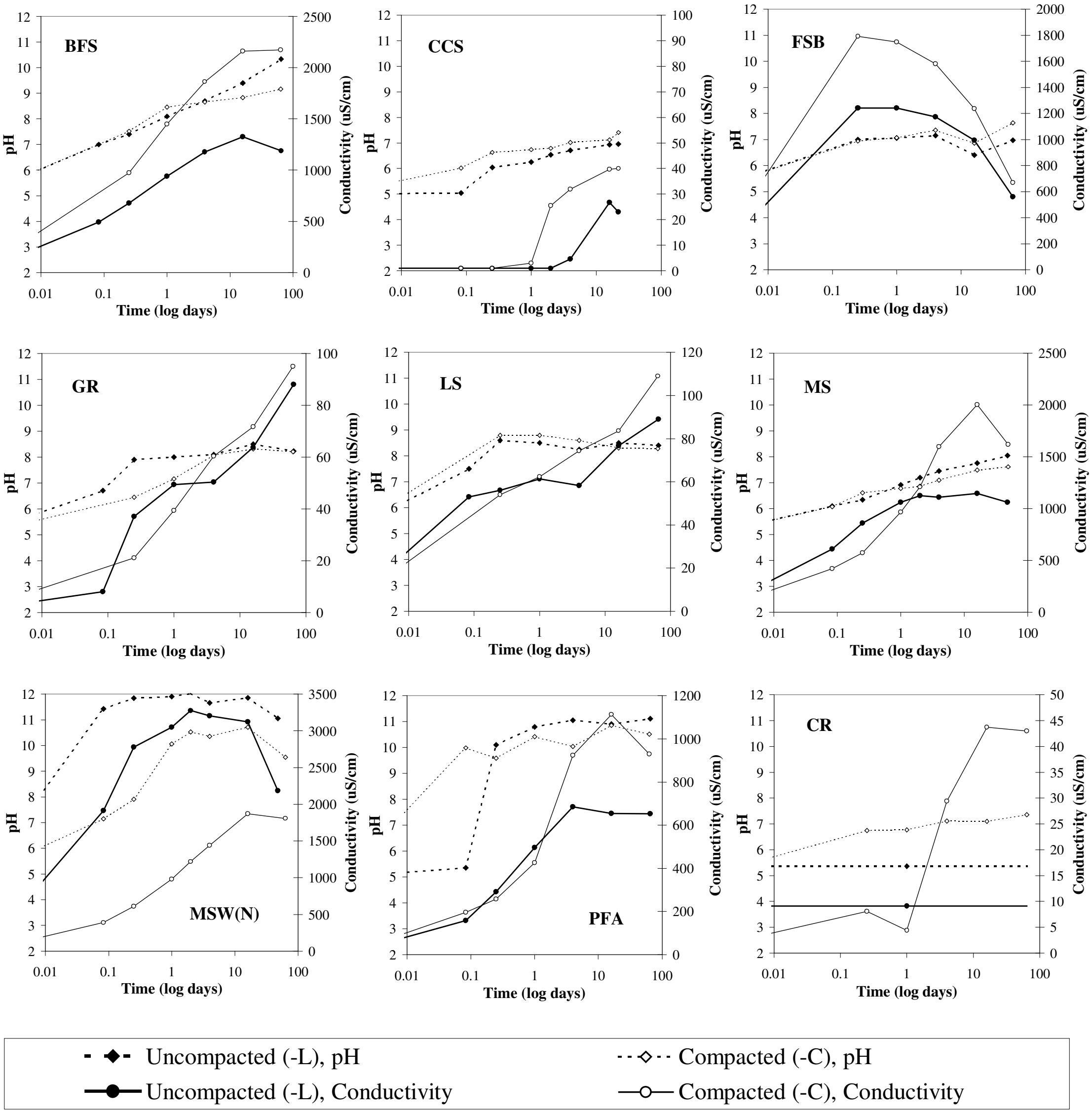


Figure 4-20: Comparison of the pH and conductivity of the test leachates from the tank-leaching tests on materials at realistic gradings (uncompacted) and the tank-leaching tests on compacted materials, plotted against time

It can be seen from Figure 4-20 that the pH of the leachates for the two types of tank-leaching test are generally of the same order of magnitude (less than 1 unit of pH deviation) for a particular material. The exception to this is the MSW incinerator ash, which shows considerably lower pH in the compacted material test. After 6 hours this difference is more than 4 units of pH and by the end of the test this difference is still greater than 1 unit. This reduction in pH modification is a potential indicator of a reduction in leaching.

The interpretation of the conductivity data is slightly more complex and the variation in L/S ratio of the tests requires consideration. For all of the tests, except the pulverized-fuel ash and crumbed rubber, the L/S ratio (by mass) in the tests on the compacted materials is approximately 5, whereas in the tests on the non-compacted materials it is approximately 10. Therefore, for similar leached quantities, the conductivity shown in the leachates from testing the compacted materials might be expected to exhibit a two-fold increase in conductivity compared to that measured in the tests on the non-compacted materials. Bearing this fact in mind, Figure 4-20 shows that most of the materials have a slower rate of leaching from the compacted material, especially in the early stages of the test. For the granite, limestone, minestone and MSW incinerator ash this trend is also illustrated in absolute terms, before applying any weighting for the difference in L/S ratio. In the later stages of the tests all materials, except the MSW incinerator ash, reveal higher conductivity in the leachates from the tests on the compacted material. However, no material exhibits conductivity greater than twice that shown in the test on the non-compacted material. Again, this evidence may indicate a reduction in leaching from compacted materials.

It may also be noted that the similarity in the patterns of pH and conductivity from the two types of testing provides encouragement for the validity of the test methods used. For example, the pH and conductivity measurements from the tests on the foundry sand (phenolic, brass) are, more or less, a scaled replica of each other.

To provide further evidence of the effect of material compaction on leaching, the concentration of species in the test leachates have again been normalised to the water quality maxima as previously described in Chapter 3 and Section 4.4.6 of this Chapter. This normalisation again provides a method of identifying general trends in species leaching. The comparison of the normalised data from the tank-leaching tests on non-compacted and compacted materials is presented in Figure 4-21 for each material. Figure 4-21 shows the sum of all the normalised species and the sum of the List I and II species.

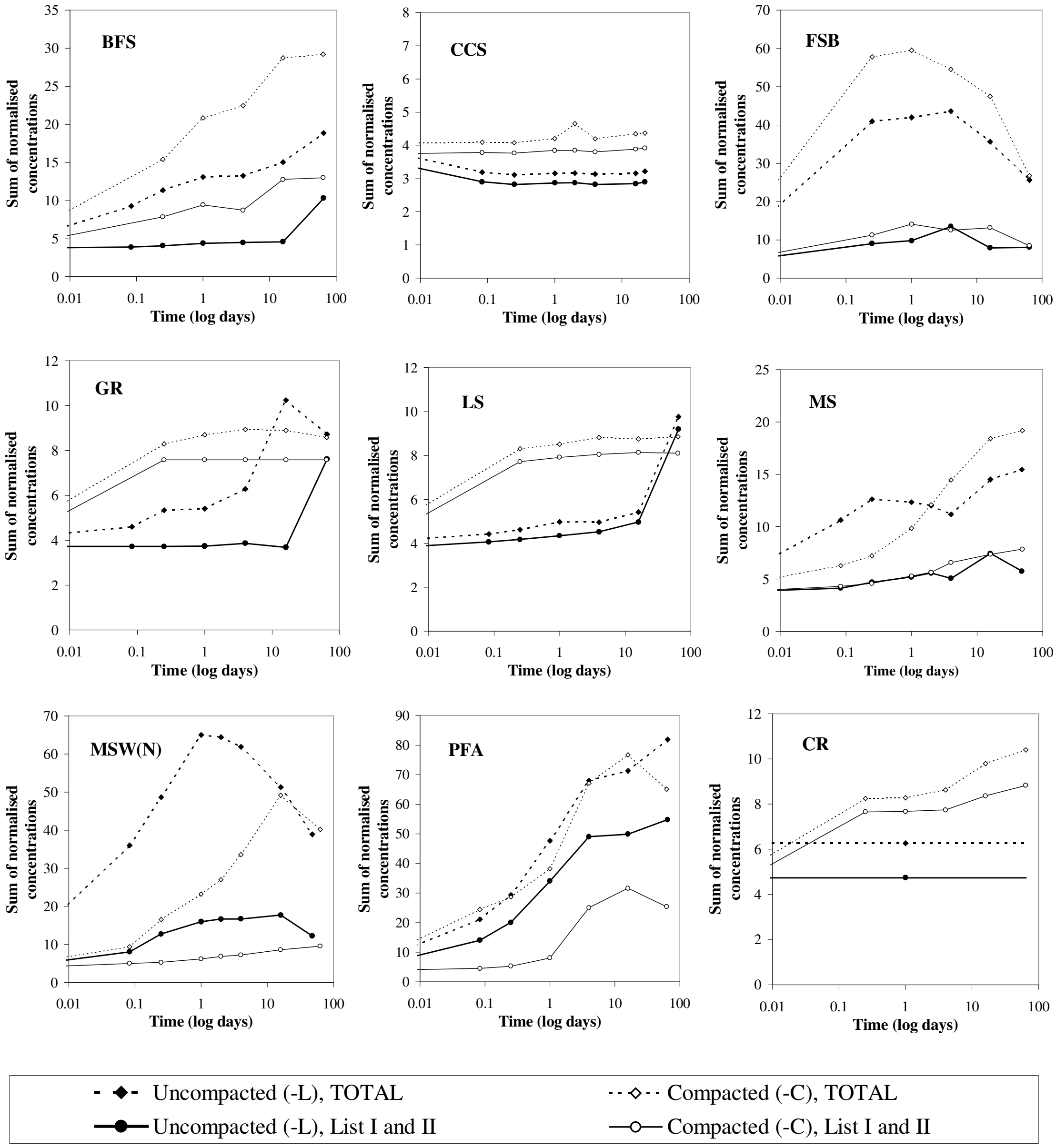


Figure 4-21: Comparison of the sum of normalised species concentrations in the test leachates from the tank-leaching test on materials at realistic gradings (uncompacted) and the tank-leaching tests on compacted materials, plotted against time

From Figure 4-21, an absolute reduction in the total normalised concentration is shown during the first 4 days of the test on the minestone and the first 16 days for the MSW incinerator ash. This is a key indicator of a reduction in the initial rate of leaching from these materials following compaction. The effect of compaction is likely to be most significant in these two materials because they are both granular materials that contain coarse particles combined with a high proportion of fine material. When compacted, such materials will exhibit a significant reduction in permeability, compared to their uncompacted state, thus restricting the circulation of the leachant through the solid material and ultimately slowing the rate of leaching.

The results from the tests on the limestone, granite and china clay sand do not contribute any evidence of a reduction, or not, in leaching as a result of compaction. The apparent shift in the data shown for these materials can actually be attributed to the lower limits of analytical detection achieved for the leachates from the tests on the non-compacted materials. The normalisation procedure used the maximum potential species concentrations (i.e. 0.1 for a concentration of <0.1) and therefore an increase in detection limit caused an increase in normalised concentration. The only conclusion that can be drawn from these test, is that compaction had not increased the rate of leaching in these materials.

The two tests on the pulverized-fuel ash were actually performed at similar L/S ratios (by mass) because of the relatively low density of this material. The test on the compacted pulverized-fuel ash had a L/S ratio of 9.53 by mass. Therefore, the interpretation of the data from this material should not be weighted for the difference in L/S ratio. From Figure 4-21 it is therefore deduced that compaction had little effect on the total leaching from this material. However, as discussed in Section 4.4.6, a significant reduction in leaching was shown between the rapid leaching characterisation test and the tank-leaching test on the non-compacted pulverized-fuel ash. This reduction was potentially attributed to the differing leaching mechanisms in the two tests and the partial consolidation of this fine-grained material in the non-compacted tank-leaching test. The low permeability of the "heap" of material in the non-compacted test would therefore be similar to that of a compacted test specimen, which could explain the apparent lack of effect of material compaction. Figure 4-21 does however reveal a reduction in the leaching of List I and II species by approximately half. It would therefore appear that another factor is affecting the leaching of these species, which are predominantly the transition elements. One possible explanation could be the effect of leachate pH. However, from Figure 4-20 it can be seen that the pH of the leachates from the two tests is relatively similar and does not change in the same patterns as the leached species. Another, possible explanation may be that the curing of the material following compaction has affected these species. Pulverized-fuel ash is a pozzolanic material that produces a stabilising cementitious reaction when activated by a source of calcium oxide (CaO). Although it cannot be confirmed here, the pulverized-fuel ash may have undergone some limited stabilisation during curing as a result of the small quantity of CaO present. Stabilisation may have reduced the permeability and leachability of some of the List I and II species. The effect of material binding on leaching is discussed in detail in Chapter 6 of this thesis.

The tests on the blastfurnace slag and foundry sand (phenolic, brass) are also inconclusive regarding the effect of material compaction on leaching. The total normalised concentrations for both materials are at levels that could be deduced from the relative L/S ratios in the tests. There is, however, certainly no evidence that compaction has increased the rate of leaching. At the end of the tests on the foundry sand (phenolic, brass) the total normalised concentrations tend to the same value. This may be an indication of a reduction in leaching but is potentially a solubility factor since the total normalised concentrations shown by this material peak after a leaching duration of only about 1 day and then decline for the remainder of the test.

4.5.5 Conclusions

- The tank-leaching test developed for assessing leaching from compacted materials proved simple to perform. A reassurance of the quality of the testing was given in the fact that the species present in the leachates generated from using this procedure were, generally, the same as those that were found from testing equivalent materials in the tests previously described.

- For most materials and species, ultimate concentrations were achieved after about 16 days, although for a number of materials leaching continued at detectable rates throughout the duration of the test. A testing duration of about 16 days is likely to be sufficient for testing materials in this way.

- It can be concluded from the data presented in Section 4.5 that compaction has a variable, but not increasing, effect on the leaching of aggregate materials. For some of the materials that were tested, namely the MSW incinerator ash and the minestone, compaction reduced the rate and/or the quantity of leaching to about a half, or even a quarter, of that shown from the same materials tested in a non-compacted condition. However, for other materials, such as the blastfurnace slag, it was not possible to identify whether any reduction in leaching occurred as a result of the compaction. The degree to which material leaching is decreased by compaction is likely to be a function of the compacted permeability, or other physical properties of the materials. The MSW incinerator ash and minestone both have low compacted permeability resulting from the high proportion, and type of, fines contained in the material. Leaching therefore becomes more a function of diffusion through the pore water than of surface wash-off of species. The leaching of coarse or continuous graded materials, or those with a high porosity, such as the blastfurnace slag, may be less effected by material compaction since they do not provide such a significant barrier to leachant permeation.

4.6 Summary of Leaching by Diffusion

- A review of existing test strategies for assessing leaching from coarse graded and monolithic materials revealed that a tank-leaching test could be used for both purposes.

- A tank-leaching test methodology was adapted from existing standards to enable conventional and alternative aggregate materials to be tested at realistic gradings in both non-compacted and compacted forms. The method was also designed to allow a direct comparison of the results to those of the rapid leaching characterisation tests performed in Chapter 3 and, also, to be capable of testing bound mixtures of materials as addressed in Chapter 6 of this thesis.

- The fabrication of the compacted test samples and the execution of the developed test methodology proved simple, and the tests were performed without significant problems. The test illustrated a good degree of repeatability and the quality of the testing was affirmed by the consistency of the species in the leachates from the different tests.

- From Section 4.4 in which materials were tested at realistic gradings, it was concluded that both material particle size and sample agitation affected the rate of leaching. The coarser graded materials that had been sized reduced for assessment using the rapid leaching characterisation test generally leached at slower rates in the tank-leaching test. For the fine-graded materials leaching was reduced because of the restricted leachate circulation to the section of the sample not in contact with the main body of the leachant. Leaching in the tank-leaching test was time dependant and ultimate leaching often exceeded that shown in the rapid leaching characterisation test.

- Material compaction was shown to reduce the rate of leaching in most materials. The effect of compaction was most pronounced for the materials that exhibit a relatively low compacted permeability compared to their uncompacted form. Compacted MSW incinerator ash and minestone leached only a quarter to a half of that shown from the non-compacted material.

5 Assessment of Leaching by Advection

5.1 Introduction

Leaching by advection occurs as a leachant flows over or through a material. It is a surface wash-off effect and is described by Conner (1990) as the dominant leaching and transportation method in permeable systems with high flow rates. Leaching by advection in compacted aggregates in road construction or bulk fill is to be examined in this Chapter.

The specific aim of this section of the testing strategy was to assess the importance of advective leaching as a mechanism for contaminant release. The relative importance of the advective and diffusive elements of the leaching process were assessed for the leaching scenario of the road construction and bulk fill applications being examined in this thesis.

Any laboratory assessment of advective leaching requires the leachant to be physically flowing through the material sample. To ensure that leaching by advection dominates over the diffusion processes in the advective leaching tests, the hydraulic scenario necessitates a 'high' flow rate and 'low' residence time.

Flow-through tests have been widely utilised for the laboratory determination of both material leachability and permeability. Many countries have standards and specifications for these tests. Some examples of flow through tests are presented by Abbott (1990), Birtwhistle (1997), Gardner and Theis (2000), CEN TC 292 (1998), NEN 7343 (1993) and NT ENVIR 002 (1995), to name a few.

In practical terms, a flow-through test was considered to be a suitable test for use in this assessment strategy because:

a) The size of the test apparatus could be adapted to enable a representative material sample to be incorporated into the test, with granular materials being assessed in a larger scale apparatus than fine-grained materials or powders.
b) The equipment and experimental procedure could be adjusted to meet the requirements of the research. For example, the liquid to solid ratio, leachant type,

flow rate and sampling frequency could be chosen, and the hydraulic head applied to the samples could be varied to ensure a defined flow rate was achieved;

c) Available equipment (typically used for the determination of material permeability) was simple to convert for application to leaching tests. Such equipment would also be widely available to industry if such a test was to be deemed appropriate to be applied in a regulatory sense;

d) The defined flow-through test could equally be applied to both unbound and bound materials, since the test parameters were easily varied, as described in Part b) above. This is important if such a test method is to be applied to the bound materials described in Chapter 6 of this thesis.

At the University of Nottingham, four types of permeameter apparatus were available for consideration. A summary description of each of these apparatus is given in Table 5-1.

Table 5-1: Test conditions for different permeameter apparatus

Test Condition	**Permeameter Test Method**			
	Large horizontal permeameter	**Large vertical permeameter**	**Double ring laboratory permeameter (DoRLaP)**	**Small vertical permeameter**
Apparatus	-Fixed wall	-Fixed wall	-Fixed wall -Double ring for determining side-wall leakage	-Fixed wall
Leaching vessel	-Galvanised Steel -Square cross section 0.3x0.3m x 1m or 0.3m	-Galvanised Steel -Dimensions: 0.3m∅ x 0.3m	-Perspex -Dimensions: 100mm∅ x 50 to 150mm	-Perspex -Dimensions: 50mm∅ x 50 to 90mm
Direction of flow	-Horizontal	-Vertical, downwards	-Vertical, upwards or downwards	-Vertical, upwards or downwards
Maximum hydraulic head (m)	~1	~1	~200	~200

The selection of the most appropriate test method was principally made on the basis of the following factors:

a) The test hydraulic regime, such as flow rate, hydraulic gradient and direction of flow;

b) The physical properties of the material to be tested, such as particle size, grading, porosity and permeability, which will influence the hydraulic conditions achievable and hence the size and type of test apparatus required to achieve verisimilitude;

c) The details of the test apparatus, for example, the materials from which they are fabricated and whether side-wall leakage can be interpreted, if thought to be necessary.

Consideration of the wide range of materials to be tested and the extent of their associated properties was made to determine the most suitable test methodology. A summary of these issues is described below.

The hydraulic flow regime established within the test will affect the duration and the quantity of contact between the leachant and the solid material. The following four flow regimes were considered:

1) Fixed hydraulic gradient, realistic of road construction conditions;
2) Fixed 'leachant volume to material volume ratio' flow rate;
3) Fixed 'leachant mass to material mass ratio' flow rate;
4) Fixed 'leachant volume to material bed volume ratio' flow rate.

The flow regime was selected to enable the advective and diffusive elements of the leaching process to be separated. For this to be achieved, the residence time of the leachate within the sample was required to be a constant so that the time dependant diffusive element of leaching would be a constant error. The only flow regime that would maintain a constant leachant residency time within the pores for different materials, and thus maintaining the diffusive element of the leaching process as a constant, is the fixed 'leachant volume to material bed volume ratio' flow rate. To achieve such as flow regime, it was necessary for the test method to include a facility for changing the hydraulic gradient applied over the test sample.

From data available in the literature [Biczysko, 1985; Abbott, 1990; Baldwin et al., 1997], permeability values of between, maybe, 10^{-1} to 10^{-8} m/s were expected for the materials under investigation. The higher permeability materials were expected to be the well-graded, coarse and single sized materials, such as Type 1 limestone, blastfurnace slag and crumbed rubber, whilst lower permeability will be shown by materials that have a high fines content, such as the incinerator ashes [Abbott, 1990;

Head, 1980; Das, 1983]. Material permeability will infer the hydraulic gradient that is required to achieve the defined flow rate and thus, for the lower permeability materials, higher hydraulic gradients were required to achieve the hydraulic conditions defined above.

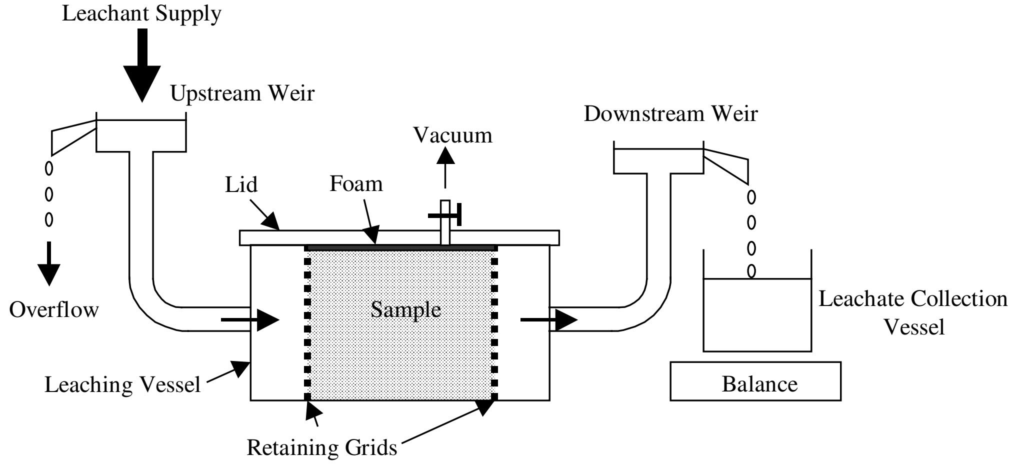
The degree of material compaction will also affect permeability and hence the hydraulic gradient required to achieve the defined flow regime. The materials were to be tested at a degree of compaction which was realistic of the usage conditions being investigated (e.g. at least 95% of proctor compaction). Testing compacted granular materials presents further complications since anisotropic values of permeability are observed. Material segregation and particle alignment that occur during compaction generate a material with a horizontal permeability up to several orders of magnitude in excess of that shown in the vertical plane [Abbott, 1990; Hunter, 1996]. The implication of this anisotropic behaviour in a pavement layer is that the dominant direction of flow is likely to be in the horizontal plane. The extent to which the direction of flow affects the leaching process is not known. However, it may be possible that an increase in the residence times and tortuosity of the flow path in the vertical plane may increase leaching.

In order to accommodate the coarse graded materials a large-scale apparatus is required, as previously defined. However, to test the lower permeability materials in such apparatus, a very high hydraulic gradient would be required across the sample in order to maintain the flow rate previously described. Baldwin et al. (1997) encountered problems when testing low permeability materials, such as pulverised fuel ash, using a large (1m x 0.3m x 0.3m) horizontal permeameter. The sample did not saturate and, instead, preferential flow (and channelling) occurred at certain points around the sample.

Ideally, one type of advective leaching test would have been used for all materials. However, in order to accommodate the different material particle sizes, permeabilities and the corresponding requirements for hydraulic gradient, it was concluded that two scales of "flow-through" test would be required, defined as:

1. A large-scale leaching vessel (minimum dimension ~ 300mm) to which a low hydraulic head (up to 1m) could be applied for testing higher permeability, and coarse grained materials;
2. A smaller-scale leaching vessel to which a higher hydraulic head could be applied for testing low permeability and fine grained materials.

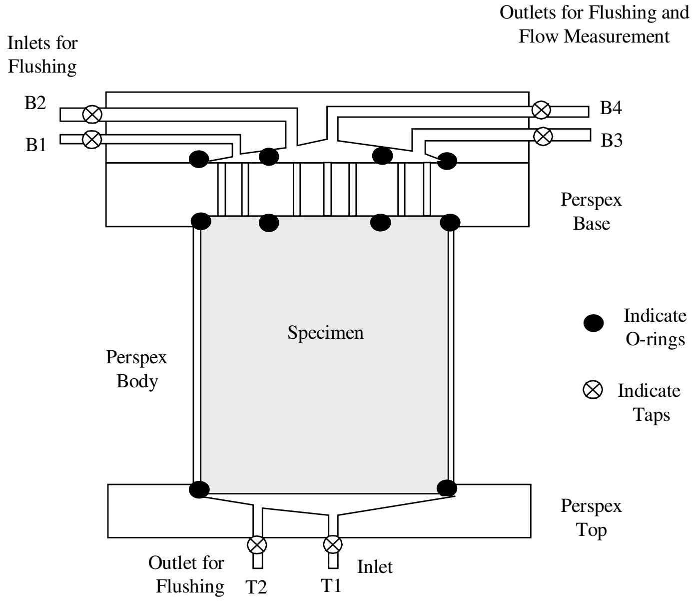
The large horizontal permeameter described in Table 5-1 was chosen for use with the coarse and permeable materials. A schematic diagram of this apparatus is shown in Figure 5-1. This equipment is as described in Department of Transport (1990) but with a cubic sample chamber 0.3m x 0.3m x 0.3m, as developed by Hunter (1996) and shown to behave similarly to the 1 metre length equipment.



SPECIMEN SIZE: 0.3m x 0.3m x 0.3m

Figure 5-1: Schematic diagram of the large horizontal permeameter [Hunter, 1996]

For the fine and lower permeability materials, the DoRLaP equipment described in Table 5-1 was selected for use. A schematic cross section of this apparatus is shown in Figure 5-2. This apparatus, developed by Birtwhistle (1997), has a cylindrical sample chamber 100mm in diameter with an adjustable height of between 50 and 150mm. This apparatus was selected because it enables peripheral flow to be assessed which may otherwise have been a cause of significant error at the high hydraulic gradients being applied to these test samples.



SPECIMEN SIZE: Diameter = 100mm, Length = 20 to 150mm

Figure 5-2: Simplified cross-section of the DoRLaP [Birtwhistle, 1997]

The advective leaching tests may be more simulative of any potential utilisation scenario than that illustrated by the rapid leaching characterisation tests in Chapter 3, but there are still many differences between the test scenario and the conditions that are experienced in a real application environment. Some important differences include:

a) The wider environmental conditions of the in-situ environment, such as temperature and biological action, are not illustrated;
b) The flow rates used in the leaching tests will be much higher than those found within a real construction due to the high hydraulic heads that will be used.

5.2 Advective Leaching Test Conditions

The two pieces of apparatus that were used for the advective leaching tests were the large horizontal permeameter and the Double Ring Laboratory Permeameter (DoRLaP). These apparatus were originally developed for determining the permeability of granular aggregate materials [Department of Transport, 1990] and clays [Birtwhistle, 1997] in the laboratory. The permeameter test methodologies were modified to enable the apparatus to be used as a leaching test. The test conditions are summarised in Table 5-2, followed by a discussion of the chosen methodologies.

Table 5-2: Advective leaching test conditions

Test Conditions	Large Horizontal Permeameter	DoRLaP
Apparatus	- Fixed-wall leaching vessel - Galvanised steel	- Fixed-wall leaching vessel - Perspex
Leachant	- Tap water	- Distilled water
Material Sample	- Compacted to >95% MDD at OMC - Dimensions: 0.3m x 0.3m x 0.3m	- Compacted to >95% MDD at OMC - Dimensions: diameter 100mm, height 150mm
Leachant Flow Rate	- 1 bed volume per 15 minutes	- 1 bed volume per 15 minutes
Leaching Duration	- 20 bed volumes	- 20 bed volumes

The leachant used in the DoRLaP tests was distilled water (5<pH<7, conductivity <0.5mS/m) as used previously in the rapid leaching characterisation tests and tank-leaching tests. The large horizontal permeameter tests used many litres of water each hour and such volumes of distilled water were not practical to prepare. For these tests tap water was used as the leachate and adjustments were made for any species that were present in this leachant.

In both apparatus, the material samples were compacted at their optimum moisture contents (OMC) to at least 95% of the maximum dry density. The materials were compacted into the leaching vessels in layers with a depth of approximately 50mm using a vibrating hammer with a square (for large horizontal permeameter) or circular (for DoRLaP) compaction foot.

The leachate flow rate was defined earlier as a fixed bed volume flow rate. To determine the rate of flow to be used in the tests, preliminary trials were performed in which the pH and conductivity of the leachates was monitored. A flow rate of 1 bed volume every 15 minutes was chosen because:

- All preliminary tests could sustain this flow rate;
- Rapidly decreasing pH and conductivity measurements indicated advective leaching was occurring;
- A 20 bed volume test could be completed within a working day.

The preliminary trials were also used to establish the test duration. In these tests, measures of pH and conductivity generally steadied after 5 to 30 bed volumes of leachate had passed through the sample. It was concluded that the test should run over a minimum of 20 bed volumes in order that equilibrium conditions would be achieved in most tests and, on a more practical level, that the test would be complete in one working day. In each test, each bed volume of leachate was collected separately and the pH and conductivity were measured. A full chemical analysis of the leachate was carried out on bed volumes 2, 5, 10 and 20.

5.3 Advective Leaching Tests Using the Large Horizontal Permeameter Apparatus

The test method for the advective leaching tests using the large horizontal permeameter, is based on the method described in the Department of Transport (1990) "A permeameter for road drainage layers". The method was adjusted to enable leachate sampling to be carried out, but the principles of the sample preparation and testing still apply. The compacted test sample is retained within the test box (which is about 150mm longer than the sample) by a grid at either end of the sample. During testing, a differential head of water is maintained across the sample by an upstream and downstream weir. This can be achieved by varying the length of pipe attached to the weir or by lifting the permeameter at one end. A schematic diagram of the apparatus is shown in Figure 5-1.

5.3.1 Large Horizontal Permeameter Test Methodology

The methodology developed for the sample preparation for the large horizontal permeameter is described in Box 5-1 below. The test procedure is summarised in Box 5-2. The sample preparation and test methodology is based on the procedure described in Department of Transport (1990) with modifications to enable it to be suitable for use as a leaching test. It was not possible for the samples to be cured for 90 days in a similar way to the compacted tank-leaching test samples because only one set of apparatus was available for use.

The leachates sampled from bed volumes 2, 5, 10 and 20 were analysed by the British Geological Survey for the complete range of 37 determinants that are listed in Table 3-9. As with all the chemical analyses, where the analytical method allowed, the majority of determinants were measured for each leachate.

Box 5-1: Sample preparation methodology for the advective leaching tests using the large horizontal permeameter

1) *Determine the maximum dry density (MDD) and the optimum moisture content (OMC)* of the test material using the methods described in BS 812.
2) *Determine the specific gravity* of the test material using the gas jar method described in BS 1377: Part 2 (1990), Section 8.2.
3) *Obtain a representative sub-sample* of the test material (laboratory sample) by coning and quartering [BS812]. The mass should be approximately 1.5 times the mass of material required to fill the sample mould (0.3m^3).
4) *Determine the residual moisture content* of the laboratory sample using the procedure described in BS812 Part 109.
5) *Mix the laboratory sample to the OMC for compaction* using tap water (the test leachant). Mix the sample in a clean concrete mixer and determine the as mixed moisture content of the material.
6) *Prepare the permeameter* by thoroughly washing the test box, lid, retaining grids and pipe work with leachant.
7) *Compact the test specimen* in the leaching vessel in 50mm layers to at least 95% of the MDD, using an electric vibrating hammer [BS 1377, 1990] fitted with a square tamping foot with dimensions of approximately 125mm. Scour the surface of the compacted material between layers to improve the join.
8) *Seal the test box* by topping the compacted sample with a sheet of tightly fitting sealed cell (impermeable) foam. A plastic-based foam was used to minimise any heavy metal contamination. The gasket and box lid are tightened in place with a series of nuts and bolts. The seal is tested by plugging the weirs with rubber bungs and applying a vacuum through the lid of the test box. To ensure there is no leak in the system, a vacuum of at least 7m water (20inches mercury) must be maintained.

Box 5-2: Methodology for the advective leaching tests using the large horizontal permeameter

1) *Calculate the bed volume* for the material being tested, using the volume and mass of the compacted sample and the specific gravity of the material (Box 5-1, Step 2). This volume of leachate represents one bed volume.
2) *Saturate the test sample* with de-aired water by applying the vacuum.
3) *Start the permeameter test* immediately after saturating the sample, to minimise diffusive leaching. Leachate is collected from the downstream weir in a large (~20 litre), clean, plastic container, placed on an electronic balance. Each bed volume of leachate is sampled independently. The test begins when the downstream weir first overflows into the leachate collection vessel.

4) *Obtain the required flow rate* by adjusting the hydraulic gradient. Calculate the required volumetric flow rate from the size of the bed volume and a flow rate of 1 bed volume every 15 minutes. Generally the required flow rate can be attained within the first few bed volumes.

5) *Leachate collection* is continuous and a new collection vessel should be used for each bed volume. The time taken to collect each bed volume of leachate should be recorded and the flow rate confirmed.

6) *Hydraulic gradient* is measured using manometers between the upstream and downstream weirs. The hydraulic gradient may have to be adjusted to maintain the required flow rate. Any change in hydraulic gradient should be noted.

7) *Leachate temperature* is measured at the upstream and downstream weirs throughout the test because the dynamic viscosity of water varies with temperature.

8) *Leachate sampling* is carried out for each bed volume of leachate. A sub-sample (100ml) is filtered through a 0.45μm filter paper. The pH and conductivity of the sample is measured and recorded. A full chemical analysis of the leachate is carried out on bed volume numbers 2, 5, 10 and 20 where three samples of the leachate (of at least 20ml) are retained for analysis (1 x unacidified, 1 x 1% nitric acid, 1 x 1% hydrochloric acid, as previously described).

9) *On completing the test* the test box is opened and a record is made of any observations that may have adversely affected the results (e.g. evidence of piping, flow across the top of the sample or non-saturation).

5.4 Advective Leaching Tests Using the DoRLaP Apparatus

The test method for the advective leaching tests using the DoRLaP, is based on the method developed by Birtwhistle (1997), for assessing the permeability of compacted clays, and modified by Bentley (2000). The principles of the original sample preparation and testing procedures were retained. A simplified cross-section of the DoRLaP apparatus is illustrated in Figure 5-2.

The DoRLaP consists of a cylindrical Perspex tube, which is clamped between two sets of Perspex plates that contain the pipe work for supplying and removing the percolating fluid. A rubber ‘O’-ring seal clamped between the material sample and the Perspex plate separates any flow resulting from side-wall leakage from that which has percolated through the central core of the sample.

The hydraulic head is applied to the sample by means of a bladder system controlled using compressed air. The maximum pressure that can be applied is 200kPa, which is equivalent to a hydraulic gradient of about 126 over a sample height of 150mm.

5.4.1 DoRLaP Test Methodology

Bentley (2000) adapted Birtwhistle's (1997) sample preparation methodology, used for clays, to enable the apparatus to be used for testing granular materials. The sample preparation methodology that was used is described in Box 5-3. Similarly, Bentley (2000) modified the sample saturation procedure to improve the degree of saturation, minimise loss of material particles and prevent swelling of the granular material. The procedure for the sample saturation and testing in the DoRLaP apparatus is described in Box 5-4. For further details of the DoRLaP test methodology, the reader is referred to Bentley (2000) and Birtwhistle (1997).

The DoRLaP samples are compacted into sections of Perspex tubing which were incorporated into the DoRLaP apparatus for testing. The availability of the tubing enabled the compacted samples to be cured for 90 days prior to testing.

Box 5-3: Sample preparation methodology for the advective leaching tests using the DoRLaP

1) *Determine the maximum dry density (MDD) and the optimum moisture content (OMC)* of the test material using the method described in BS 812.
2) *Determine the specific gravity* of the test material using the gas jar method described in BS 1377: Part 2 (1990), Section 8.2.
3) *Obtain a representative sub-sample* of the test material (laboratory sample) by coning and quartering [BS 812]. The mass should be approximately 6 times the mass of material required to fill the sample mould (100mm ∅ x 150mm).
4) *Determine the residual moisture content* of the material in the laboratory sample using the procedure described in BS812: Part 109.
5) *Mix the laboratory sample to the OMC for compaction* using distilled water (the test leachant).
6) *Prepare the Perspex sample moulds* by washing in leachant then dry, weigh and label each mould. Prepare circles of plastic sheeting to fit inside the moulds.
7) *Compact the test specimens* into the Perspex moulds (supported by the compaction apparatus and placed on a circular sheet of plastic) in 50mm layers to at least 95% of the MDD, using an electric vibrating hammer [BS 1377, 1990] fitted with a circular tamping foot. Weigh the required mass of material for the layer being compacted and scour the surface of the compacted material between layers. Level off the completed sample with the top of the mould. Use the first sample to determine the moisture content of the compacted specimen. Prepare 3 more compacted test samples for testing.
8) *Cure the compacted samples* in a sealed plastic bag for 90 days, turning over periodically. The same curing period was used in the compacted tank-leaching tests (see Section 4.5).

Box 5-4: Methodology for the advective leaching tests using the DoRLaP

1) *Calculate the size of the bed volume* for the material being tested, by using the volume and mass of the sample and the specific gravity of the material (Box 5-3, Step 2). This volume of leachate is divided into the inner and outer collection areas.
2) *Saturate the sample* with de-aired leachant. The saturation apparatus containing the material sample is place on a plastic support stand in a leachant bath. The leachant level should be above the top of the sample but below the tap in the top plate. The sample is saturated for 18 hours.
3) *Determine the degree of saturation* by removing the saturation assembly from the leachant bath (with all taps closed), wiping off any excess water and weighing the assembly. Calculate the degree of saturation, which must be greater than 85%.
4) *Set up the DoRLaP apparatus* to begin the advective leaching test. De-air and seal the apparatus whilst in the leachant bath. Remove from the bath, with all taps closed, and orientate so that leachate will flow vertically up through the sample.
5) *Start the DoRLaP leaching test* by attaching the leachant supply pipe to the tap in the base of the DoRLaP. Apply a low water pressure and open the leachant supply taps. Open the taps in the top plate that direct the leachate from the inner and outer areas to suitably calibrated leachate collection vessels (e.g. measuring cylinders) which, if possible, should sit on an electronic balance for the accurate determination of leachate mass. The test begins when leachate first flows from the apparatus.
6) *Obtain the required flow rate* by adjusting the hydraulic gradient. Calculate the required volumetric flow rate from the size of the bed volume and a flow rate of 1 bed volume every 15 minutes. Generally the required flow rate can be attained within the first few bed volumes.
7) *Leachate collection* is continuous and new collection vessels should be used for each bed volume. The time taken to collect each bed volume of leachate is recorded and the flow rate confirmed.
8) *Hydraulic gradient* is measured from the pressure gauge readings. The pressure may have to be adjusted to maintain the required flow rate. Any change in hydraulic gradient should be noted.
9) *The leachant supply vessel* periodically needs refilling. During this process the flow has to be switch off for a few minutes. This may affect the quality of the leachate and these bed volumes should not be used for complete chemical analysis.
10) *Leachate sampling* was carried out as described in Box 5-2, Step 8. All sample analysis was carried out on the leachate collected from the inner sampling area.
11) *During and after the test* the sample should be examined for any evidence of non-uniform flow, such as piping.

The leachates were analysed by the British Geological Survey as described for the leachates from the large horizontal permeameter.

5.5 The Advective Leaching Tests

The advective leaching tests were carried out on the materials listed in Table 5-3. The materials were selected to cover the range of gradings and types of materials included in the research. They also reflect the materials tested in the diffusive tank-leaching tests.

The choice of test method (i.e. large horizontal permeameter or DoRLaP) was based on the material particle size and expected permeability. The coarse graded and high permeability materials were tested in the large horizontal permeameter and the finer graded and lower permeability materials tested in the DoRLaP. When selecting the test methodology, consideration was made as to whether the materials were to be tested in a treated/bound condition later in the research programme (See Chapter 6) and the potential effect on permeability of that treatment. Where possible, the unbound and bound material should be tested in the same apparatus for ease of comparison.

Table 5-3: Summary of the advective leaching tests on materials at realistic gradings and compaction

Material	Test Abbreviation	Apparatus
Blast furnace slag	BFS-P	Large horizontal perm.
China clay sand	CCS-D	DoRLaP
Crumbed rubber (2-8mm)	CR-P	Large horizontal perm.
Foundry sand (alkali phenolic)	FSB-D	DoRLaP
Limestone	LS-P	Large horizontal perm.
MSW incinerator ash (New incinerator)	MSW(N)-P	Large horizontal perm.

Blank advective leaching tests were carried out on both the large horizontal permeameter and the DoRLaP apparatus. The tests were used to quantitatively determine whether the test apparatus and procedure result in any cross-contamination of the leachant. The blank leaching tests used the described testing procedures but without having a compacted material sample in the leaching vessel. To take account of the much larger bed volume of the empty leaching vessels, the leachant residence time in the control tests was increased to 45 minutes. The blank tests were run over 3 bed volumes. The pH and conductivity of each bed volume was measured and a full chemical analysis was carried out on bed volume 2.

A number of problems were encountered with maintaining the flow rate within the advective leaching tests. In many of the tests the hydraulic gradient had to be increased through the test in order to maintain the flow rate. This was particularly apparent in the test on the MSW incinerator ash, where the test was terminated after 13 bed volumes because the required flow rate could not be sustained. It is thought that the high fines content of the MSW incinerator ash resulted in a low permeability material despite the presence of a coarse fraction. A different problem was encountered with the DoRLaP test on the foundry sand (phenolic, brass) where the minimum hydraulic gradient was too high for the required flow rate. For this material the flow rate was about 1 bed volume every 7.5 minutes.

5.6 Results

The composition of the leachates obtained from the blank permeameter tests are given in Table 5-4 along with the detectable components of the tap water leachant used in the large horizontal permeameter and distilled water leachant (previously reported in Chapter 3 and 4) used in the DoRLaP apparatus. Any species that were listed in Table 3-9 but are not listed in Table 5-4 may exist but are at concentrations below the limits of analytical detection.

Table 5-4: Species detected in the blank permeameter test leachates and leachants

Test	**Sample Time**	**pH**	**Cond.**	**Concentration of determinants, mg/l**					
			µS/cm	**<0.01**	**0.01- <0.1**	**0.1- <1**	**1- <10**	**10- <100**	**>100**
ControlP	**BV2**	7.8	576		Br Ba	NO_2 F P Si Sr Cu Zn	K NO_3	Ca Mg Na Cl SO_4 TOC S	HCO_3 TIC
Tap water leachant		7.9	580	Mn	Br Ba	NO_2 F P Si Sr Cu Zn	K NO_3	Ca Mg Na Cl SO_4 TOC S	HCO_3 TIC
ControlD	**BV2**	6.1	19	Ba Sr Mn	Ni Cu	Na K Cl SO_4 S Si Zn	Ca TOC		
Distilled water leachant		5.2	7	Se	Ba Zn	Ca Na K Cl TIC Si	TOC		

Control P = Large horizontal permeameter control test

Control D = DoRLaP control test

BV = bed volume sampled

It can be seen from Table 5-4 that, in general, those species detected in the leachates from the control permeameter tests are present at similar concentrations in the leachant used in each test. The large horizontal permeameter does not appear to contribute detectable quantities of any chemical species over and above those found in the tap water leachant. The DoRLaP apparatus, however, contributes detectable quantities of calcium, sulphate, total sulphur, zinc, nickel and copper to the distilled water leachant. It can be seen from Table 5-4 that the concentrations of many of the chemical species are greater in the leachates from the large horizontal permeameter than the DoRLaP. This difference is the result of the different leachants in use in the two types of apparatus rather than leaching from the test apparatus itself.

The blank test data was used to quantitatively adjust the concentrations of the species detected in the leachates from the permeameter tests on the different materials, in order to account for those species that are, thus, deemed to result from the test method or leachant. Adjustments were made for all of the species listed in Table 5-4 with the exception of those species detected at concentrations less than 0.01mg/l, which were considered to be too close to the limits of analytical detection to guarantee the accuracy.

The concentrations of the species in the leachates from the permeameter tests were quantitatively adjusted for those species that resulted from the test method or leachant, as described above. A summary of the results of the permeameter tests are given in Table 5-5 where, for each test, the species detected in the bed volumes of leachate that were sampled are tabulated within concentration ranges. Those species that are listed in Table 3-9 that are not listed in Table 5-5 may be present in that leachate but are at concentrations below the limits of analytical detection.

The physical properties of the test materials and the conditions established within the permeameter tests are summarised in Table 5-6, whilst the calculated material permeabilities at specific bed volumes are given in Table 5-7.

Table 5-5: Summary of permeameter test data (concentrations in leachate)

Test	BV	pH	Cond. (µS/cm)	Concentration in leachate (mg/l)						
				<0.01	0.01-0.1	0.1-1	1-10	10-100	100-1000	>1000
BFS-P	**2**	8.3	1914		Zn	Br NO_2 Sr Mn Li	Mg NO_3 F B	Na K Cl Si	Ca S	SO_4
	5	8.2	1342		Zn	Mg Sr Mn Li B	Na F NH_4	K Cl Si	Ca SO_4 S	
	10	8.2	1172		Zn	F Sr Mn Li B	Na Cl NO_2 NH_4	K Si	Ca SO_4 S	
	20	8.2	1063		Li	NO_2 F Sr Mn B	Si	K	Ca SO_4 S	
CCS-D	**2**	8.7	88	Ba Mn Li	Sr Al	Cl P S	Ca Mg Na SO_4 F TIC Si	K HCO_3		
	5	8.9	51	Ba Mn Li	P Sr Al	Mg F Si	Ca K TIC	HCO_3		
	10	8.8	45	Sr	F Al	Mg Si	Ca K TIC	HCO_3		
	20	7.9	36	Sr Mn		Mg Si	Ca TIC	HCO_3		
CR-P	**2**	8.1	27		F Mn Fe Co Li	S Zn B	K Cl SO_4 Si	NO_3		
	5	7.9	9		F P Mn Fe Li	K Cl Zn B	NO_3 NH_4 Si			
	10	7.8	0	Mn	F Fe	Cl P Zn B	Si	NO_3		
	20	7.7	0	Mn	Cl Fe	K F P Zn B	Si	NO_3		
FSB-D	**2**	10.5	449	Ba Sr Mn	NO_2 P Fe Cu	Cl SO_4 F S Al	Si	Na TOC TIC	K HCO_3	
	5	10.8	241		NO_2 P Fe	Cl NO_3 F TOC Si Al	Na SO_4	K HCO_3 TIC		
	10	10.4	143	Ba	Fe	Si Al	Na TIC	K HCO_3		
	20	10.0	78	Mn	Fe Al	Cl Si	Na TIC	K HCO_3		
	40	9.5	44			Na Si	TIC	K HCO_3		
LS-P	**2**	8.0	40		Sr Zn Li	F B	K Cl NH_4 Si	NO_3		
	5	7.9	30	Mn	Sr Fe Li B	F Zn	K Cl NO_3 NH_4 Si			
	10	7.9	2	Mn Fe	Sr Li B	F Zn	K Cl NH_4 Si	NO_3		
	20	7.7	1	Mn Li	Fe Zn	F B	Cl Si	NO_3		
MSW(N)-P	**2**	8.5	193		Mo B		SO_4 S NH_4 Si Al	Na K Cl NO_3		
	5	8.3	75	Mn	F B		K SO_4 Si Al	Na Cl NO_3		
	10	8.1	59	Mn	F B	Al	Na K SO_4 Si	Cl NO_3		

Table 5-6: Summary of the physical properties and test conditions for the permeameter tests on materials at realistic grading and compaction

Material Code	MDD (Kg/m^3)	OMC (%)	Specific Gravity (Mg/m^3)	Mass of Test Sample (Kg)	Void Ratio	Approximate Flow Rate (Mins/BV)
BFS-P	2.0	6	2.86	55.39	0.29	15
CCS-D	1.9	10	2.70	2.194	0.37	15
CRR-P	0.6	-	1.12	16.12	0.47	15
FSB-D	1.9	10	2.64	1.901	0.39	7.5
LS-P	2.2	8	2.69	57.90	0.19	15
MSW(N)-P	1.6	12	2.56	48.35	0.28	15

Table 5-7: Summary of the calculated material permeabilities

Material Code	Permeability (m/s)			
	BV 2	BV 5	BV 10	BV 20
BFS-P	1.0E-04	8.9E-05	8.2E-05	5.2E-05
CCS-D	3.7E-06	3.2E-06	2.9E-06	4.5E-06
CRR-P	6.1E-03	3.8E-03	7.5E-03	5.8E-03
FSB-D	3.6E-04	4.2E-04	4.8E-04	4.9E-04
LS-P	4.2E-03	4.0E-03	4.2E-03	4.7E-03
MSW(N)-P	1.5E-04	1.3E-04	9.4E-05	2.9E-05

From Table 5-5 it can be clearly seen that the concentrations of species within the test leachates vary over a range of more than 5 orders of magnitude, which is the same range as that shown in the tank-leaching tests described earlier in Chapter 4. Observations from Table 5-5 compared with the results from the tank-leaching tests on compacted materials shown in Table 4-10, generally indicate that the leachates from each material contained the same detectable species in both types of test. A more detailed comparison of the results from the two types of leaching test is provided in Section 5.7.

The general patterns of leaching behaviour are illustrated by the pH and conductivity of the test leachates as shown in Figure 5-3 and Figure 5-4. It can be seen from Figure 5-3 that in general the leachate pH decreases slightly as more bed volumes of leachant are passed through the material sample. A similar but more pronounced trend it shown by the conductivity, as illustrated in Figure 5-4. The relationship between the declining measured conductivity and the number of bed volumes passed can be approximated to an exponential function (range of R^2 between 0.88 and 0.92) for the materials presented in Figure 5-4. This indication of a decline in leached species with increasing number of bed volumes is the expected trend from an advective leaching test.

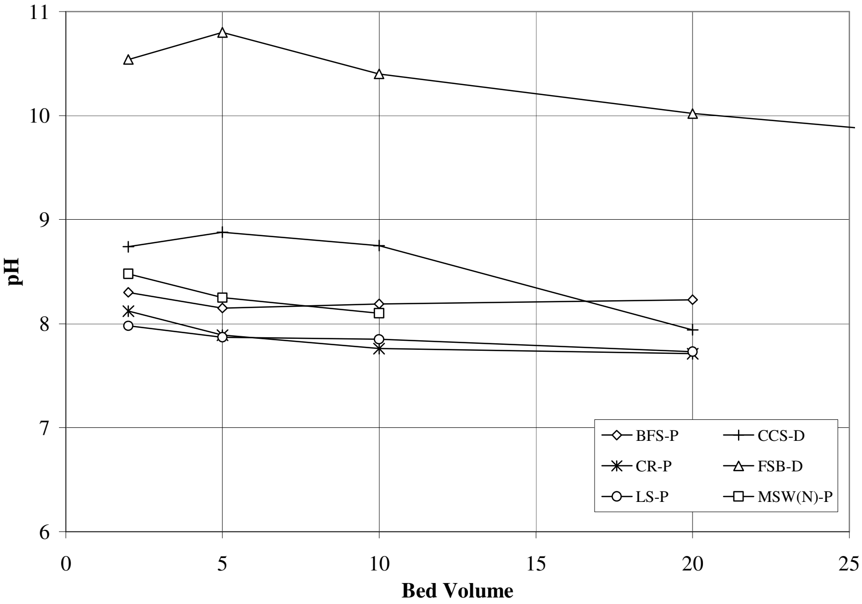


Figure 5-3: pH as a function of the number of bed volumes of leachate that have passed through the permeameter tests on compacted aggregates

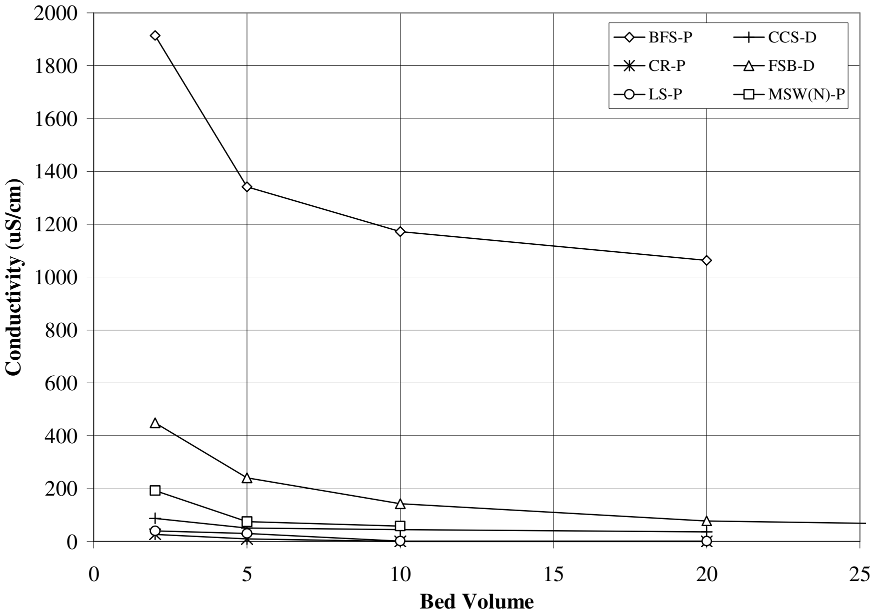


Figure 5-4: Conductivity as a function of the number of bed volumes that have passed through the permeameter tests on compacted aggregates

Many of the species measured in the leachates exhibit this expected trend of a decrease in species concentration with increasing number of bed volumes passed. These general trends are shown in Figure 5-5, which illustrates the concentrations of all species measured in the leachates from the foundry sand (phenolic, brass) sampled at bed volumes 2, 5, 10, 20 and 40. The rapid initial leaching shown by many of the species indicates the significant availability of those species on the surfaces of the individual material particles. This surface wash off provides an initial influx of species leached from the surface of the material particles, which then rapidly declines as this source is diminished.

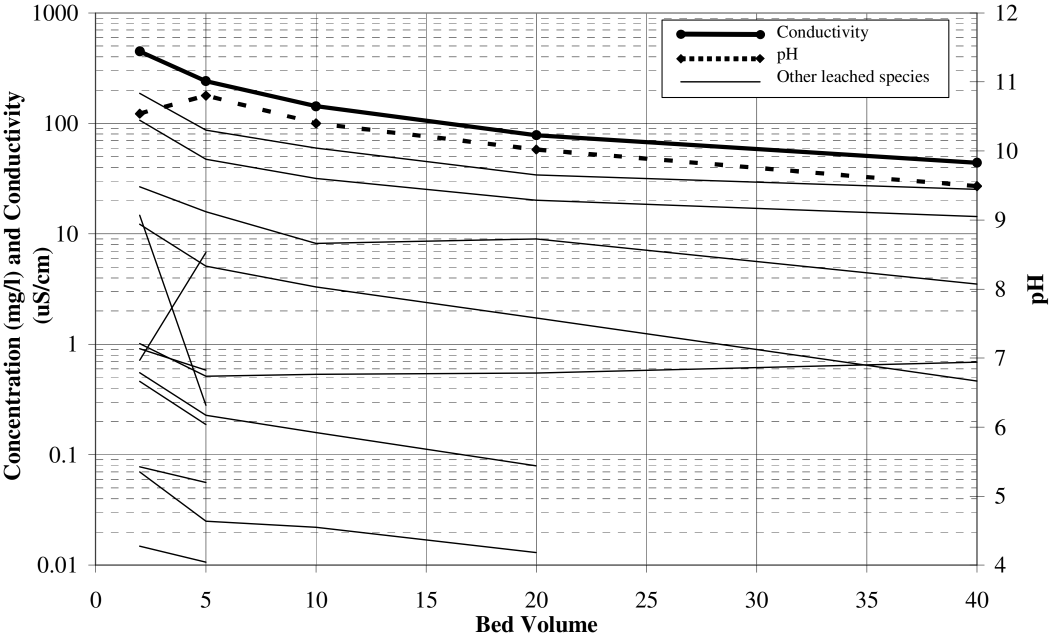


Figure 5-5: Concentration of species in the leachate as a function of the number of bed volumes of leachate that have passed in the permeameter test on foundry sand (phenolic, brass)

Some species appear to leach in the highest concentrations after a number of bed volumes of leachate have passed through the test sample. This apparent delay in leaching may result from a number of factors. It can be seen from Table 5-5 that the species present at the highest concentrations in the leachates are generally the most soluble species, particularly the alkali and alkali earth metals. The rapid leaching of such species may saturate the leachate and other less soluble components of the materials may only be able to leach once the surface source of soluble species is decreased.

The majority of species show the most rapid decline in species concentrations over the first 5 to 10 bed volumes of leachate passing through the test material. In the leachates from the latter part of the test the species concentrations are often relatively constant. This constant rate of leaching may be governed by the diffusive element of the leaching process as proposed by Gardner et al (2000).

5.7 Discussion

5.7.1 Normalisation of the Data from the Permeameter Leaching Tests

To obtain an indication of the relative toxicity of the species in the leachates from the different materials, the test data has again been normalised to the water quality maxima, using the method described in Section 3.4.6 for interpretation of the rapid leaching characterisation tests. The normalisation of the leaching test data against such criteria is, again, used here purely as a tool for interpretation.

Table 5-8 presents the normalised data for the permeameter tests on the compacted materials. The results represent the maximum concentration of each species from any of the bed volumes sampled in that test. It can be clearly seen from Table 5-8 that only a few species exceed the water quality maxima. No species exceed the water quality maxima by more than 100 times and ammonium is the only List II species to exceed the water quality maxima.

A comparison between the normalised permeameter test data and a similar representation of the tank-leaching test data on materials at realistic gradings and on compacted materials presents an interesting observation. If advection is the dominant leaching mechanism then at a given liquid to solid ratio the permeameter and tank-leaching tests should show equal leached quantities of each species. It has already been shown that, in general, the concentration of species in the leachates from the permeameter tests decline with an increasing number of bed volumes of leachate passed. If both of these factors are true, then the absolute concentration of species in the leachates from the early stages of the permeameter tests would be expected to be higher than the peak concentrations in the tank-leaching tests. However, the comparison of the normalised species concentrations from these tests, appears to show that absolute species concentrations are in fact lower in the leachates from the permeameter tests. This observation implies that diffusive as well as advective leaching processes are significant in the leaching shown in the tank-leaching tests. This comparison is discussed further in the following part of this chapter.

Table 5-8: Normalisation of permeameter tests data to arbitrary water quality maxima

Aggregate	Test	Maximum normalised concentration in leachate					
		Detected	1-10		10-100		>100
		<1	II	Other	II	Other	
BFS	**P**	Mg Na Cl NO_3 Zn Li B	NH_4	Ca K SO_4 F		NO_2 Mn	
CCS	**D**	Ca Mg Na Cl SO_4 F P Ba Mn Al Li		K			
CR	**P**	K Cl SO_4 NO_3 F P Fe Zn Li B	NH_4	Mn			
FSB	**D**	Ca Na Cl SO_4 NO_3 NO_2 F P Ba Mn Fe Cu		K Al			
LS	**P**	K Cl NO_3 F Mn Fe Zn Li B	NH_4				
MSW(N)	**P**	Na Cl SO_4 NO_3 F Mn Mo Li B	NH_4	K		Al	

P = Large horizontal permeameter on compacted material at realistic gradings

D = DoRLaP on compacted material at realistic gradings

5.7.2 Relationship between the Advective and Diffusive Components of Leaching

A general relationship between the advective and diffusive components of the leaching process has been proposed in the previous section. To enable a closer consideration of this relationship the pH and conductivity of the leachates from the permeameter tests were compared to those from the tank-leaching tests on the uncompacted and compacted materials. The pH of these leachates are shown for each material in Figure 5-6 and the equivalent representation of the conductivity is shown in Figure 5-7.

From Figure 5-6 it can be seen that the pH of the leachates from the blastfurnace slag, limestone and crumbed rubber materials are within an expected range given the results of the tank-leaching tests, the residence time in the permeameters and the higher pH of the tap water leachant compared to the distilled water. The china clay sand and foundry sand (phenolic, brass) that were tested in the DoRLaP apparatus exhibit a more significant change in pH than in the tank-leaching tests. This was not expected and will be considered when in interpreting the species concentrations.

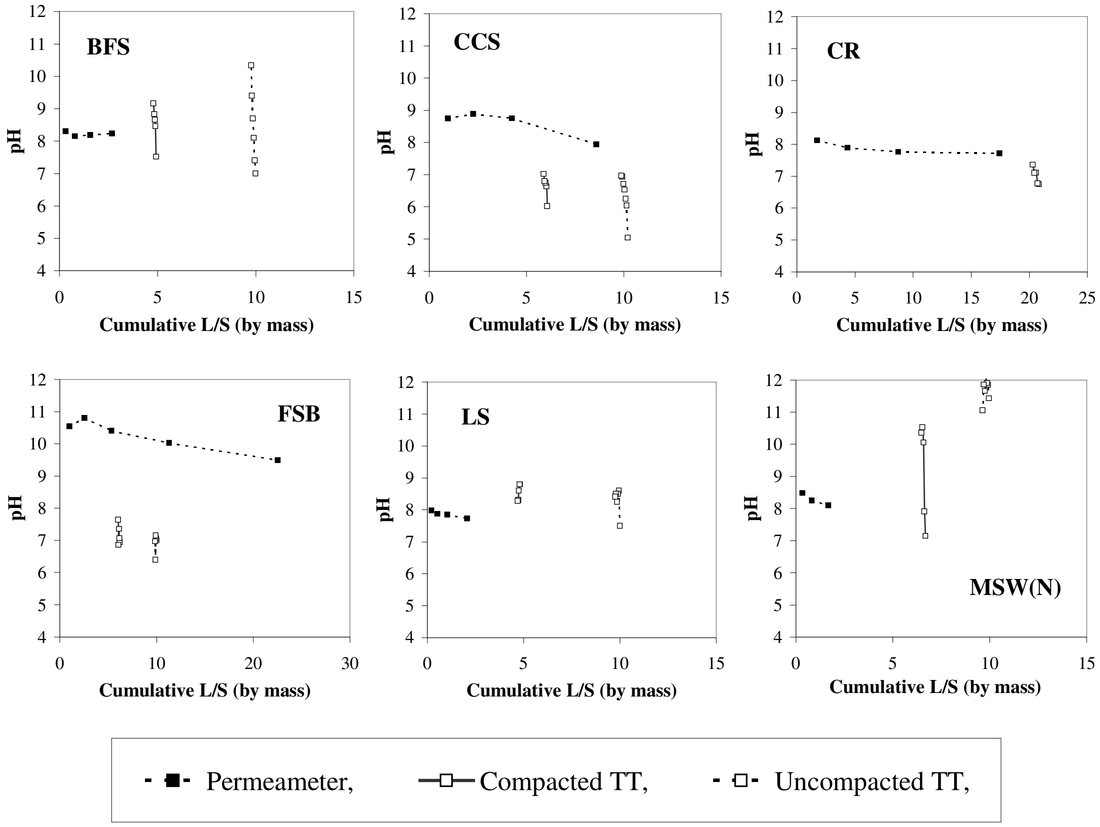


Figure 5-6: Comparison of the pH of the leachates from the permeameters and the tank-leaching tests on the uncompacted and compacted materials, plotted against liquid to solid ratio.

The conductivity exhibits differing patterns to that of the pH. The conductivity of the blastfurnace slag is as would have been expected, whilst the china clay sand is the only material to have a higher than expected conductivity. The remaining materials all exhibit a lower conductivity than would have been expected if leaching was mainly governed by advection. The problems with the permeameter test on the MSW incinerator ash are highlighted by the very low conductivity of these leachates compared to that shown in the tank-leaching tests.

The general trend of low conductivity is likely to be the result of the short residence times of the leachate within the material sample. To identify whether the patterns shown for the conductivity are reflected in the relationship between advective and diffusive leaching of the individual species, the absolute leached quantity of each species (mg/kg) from each material was determined for the permeameter and tank-leaching tests. For the tank-leaching tests the leached quantity was calculated directly from a known leachate volume, material mass and species concentration in the leachate. For the permeameter tests, an approximation to the cumulative release was calculated from available information on the:

- volume of leachate passing through the permeameter between each analysed sample of leachate;
- concentration of species in each analysed sample of leachate.

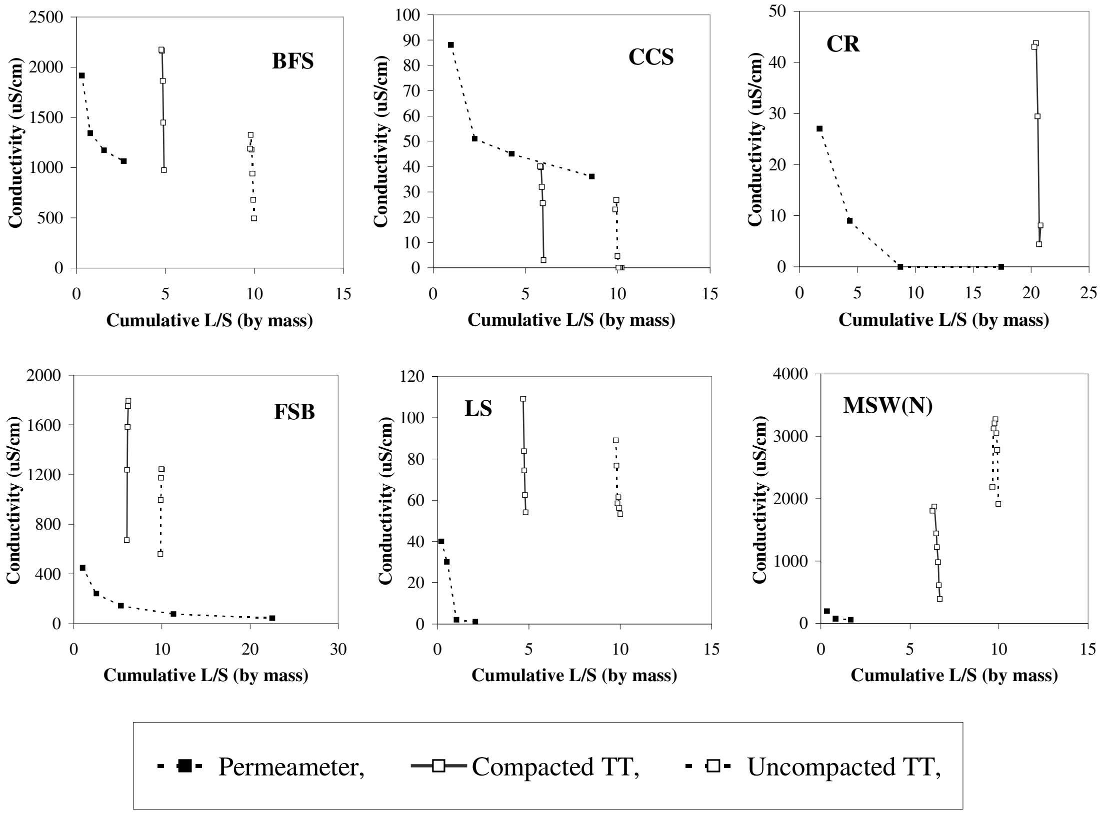


Figure 5-7: Comparison of the conductivity of the leachates from the permeameters and the tank-leaching tests on the uncompacted and compacted materials, plotted against liquid to solid ratio.

Figure 5-8 shows schematically how the total leached quantity was approximated. The species concentrations measured in the leachate samples were assumed to apply to half of the volume of leachate passed since the previous analysis and to half of the leachate passed before the next analysis.

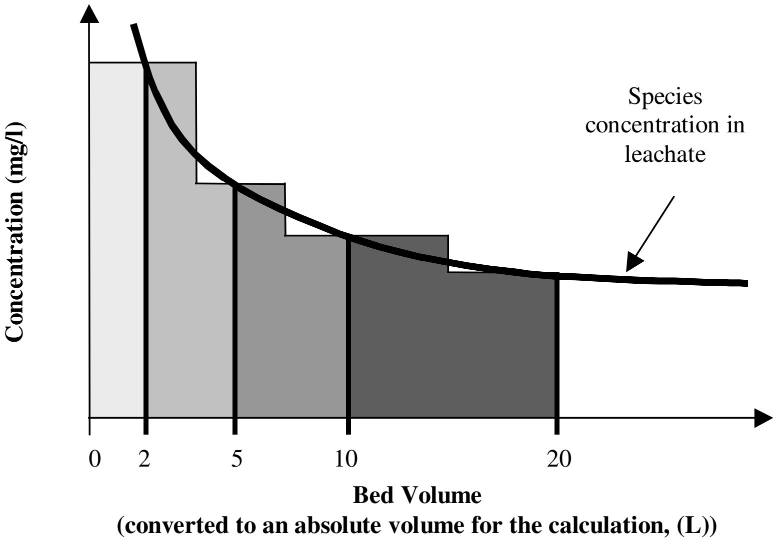


Figure 5-8: Schematic diagram to illustrate calculation of cumulative release from the permeameter tests using the Rectangular Rule

The absolute quantity of each species (mg/kg) released from each material in the permeameter and tank-leaching tests were compared in order to assess the relative importance of advective and diffusive leaching. Examples of the patterns are illustrated in Figure 5-9 for blastfurnace slag and Figure 5-10 for foundry sand (phenolic, brass). For a given liquid to solid ratio (by mass), the absolute quantity of species released from each material is generally less in the permeameters than in the tank-leaching tests. For many species the estimated quantities from the permeameter test are of the same order of magnitude as those shown in the tank-leaching tests but the permeameter results tend to be at the lower end of the range. This lower quantity of leaching is likely to be the result of the short contact time between the leachate and the material sample in the permeameter tests. This short contact duration does not allow for such significant diffusive leaching to occur. For a number of the species present at very low concentrations, the permeameters over predict the release from the tank-leaching tests because the low liquid to solid ratio of each bed volume in the permeameters allows the analytical detection of lower quantities of leached species.

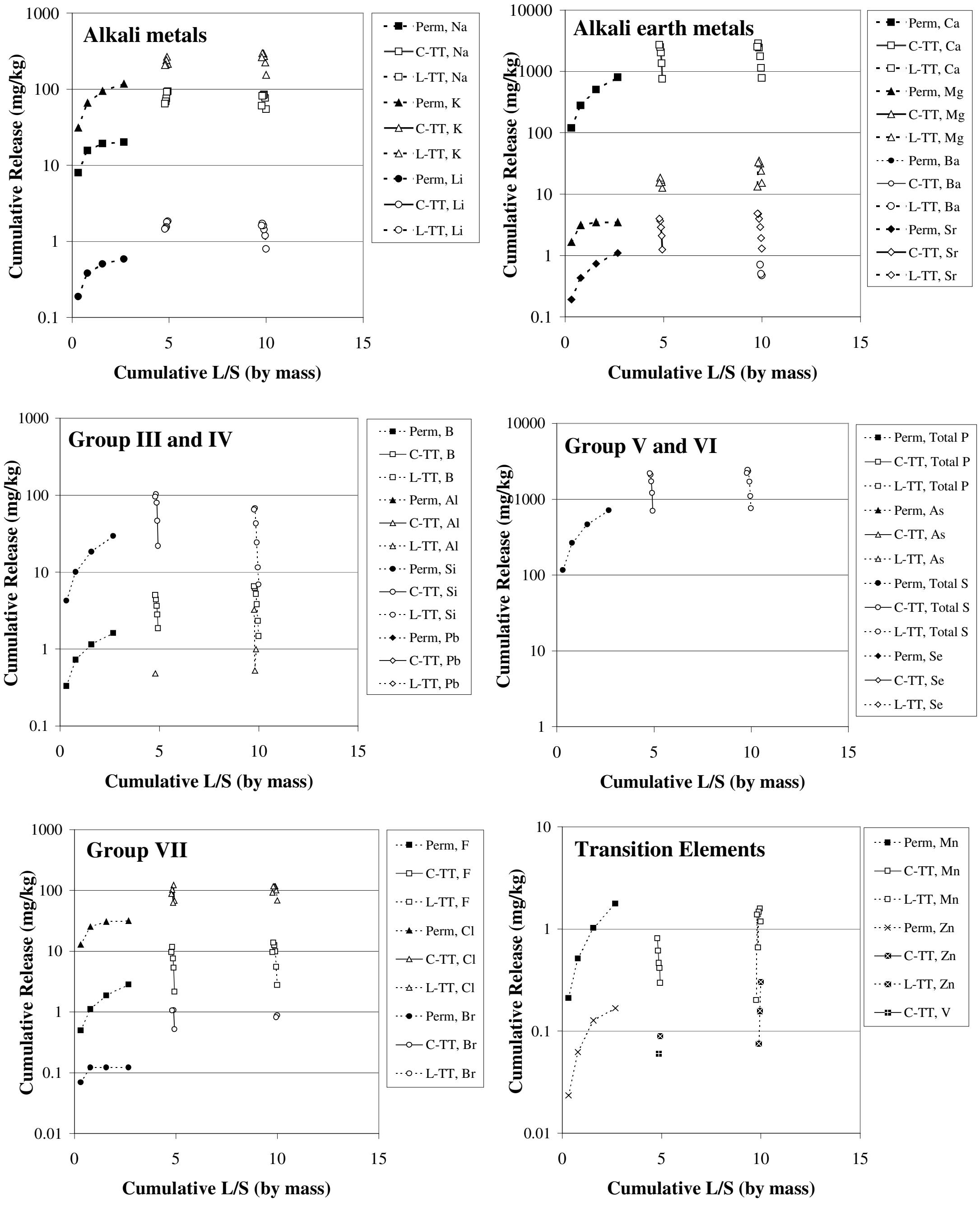


Figure 5-9: Comparison of the total leached species in the test leachates from the permeameter and tank-leaching tests on blastfurnace slag, plotted against liquid to solid ratio

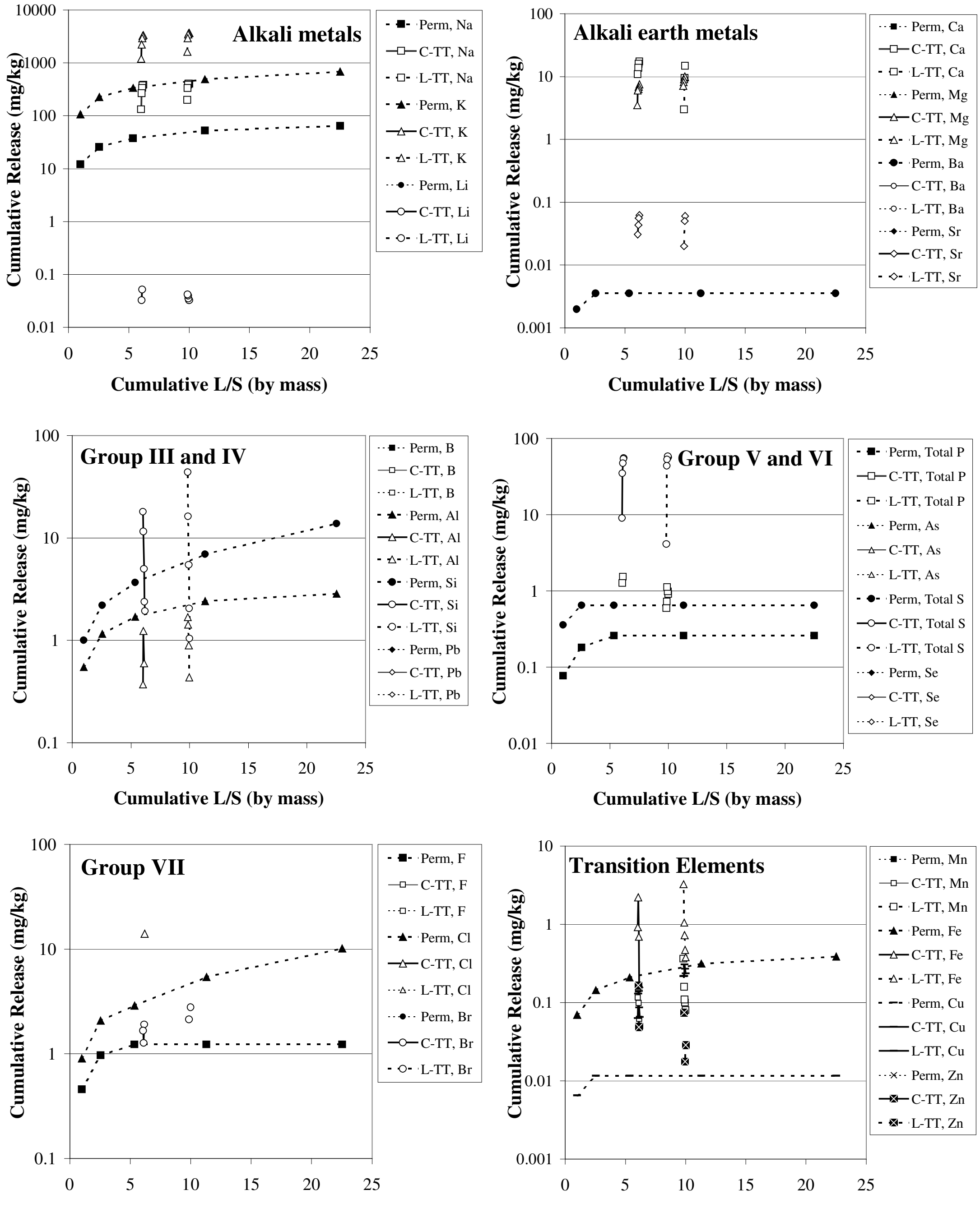


Figure 5-10: Comparison of the total leached species in the test leachates from the permeameter and tank-leaching tests on foundry sand (phenolic, brass), plotted against liquid to solid ratio

Consideration was made as to whether any of these leaching patterns could be the result of differing pH limiting conditions. However, plots of species concentrations against pH did not replicate any of the solubility curves previously discussed in

Chapter 3. It is therefore assumed that the leaching of most of the species is not controlled by solubility constraints.

5.8 Summary of Leaching by Advection

- A review of available flow-through test equipment and the parameters of the materials to be tested, revealed that two sizes of apparatus were required in order to be able to test the full range of material types. The test methodologies used were the large horizontal permeameter (Department of Transport, 1990) and the DoRLaP (Birtwhistle, 1997 as modified by Bentley, 2000). The test methodologies were adapted to enable the test to be performed and the leachate sampled in a way that was suitable for a leaching test, and would minimise any cross-contamination.

- The application of the test methodologies proved difficult for materials that exhibited certain properties. Notably, the MSW incinerator ash, which was tested in the large horizontal permeameter because of the large particles contained within the material, proved difficult to test in this apparatus because of the low permeability resulting from the high fines content.

- The general trend shown by the data was a decrease in species concentrations with an increasing number of bed volumes passing. This trend was as expected. The concentration of species in the leachate were, however, lower than would have been expected based on the data from the tank-leaching tests.

- It was generally concluded that the total quantity of each species leached during a permeameter test was less than the same material in a tank-leaching test for the same liquid to solid ratio. This phenomenon could not be attributed to pH controlling mechanisms and, thus, it was concluded that the relatively short contact time shown in the permeameters limited the contributions to leaching from diffusion in these tests.

5.9 Leaching by Advection and Diffusion - Summary

- Based on the outcome of the diffusive and advective leaching tests it would be apparent that materials require assessment in the form in which they are to be utilised, which in this case would represent the conditions of road construction or bulk fill. It is therefore proposed that any leaching test should be carried out on materials at realistic gradings where possible, and for those materials for which compaction is likely to have a significant effect, they should also be tested in compacted form.

6 Influence of Binder on Leaching

6.1 Introduction

The laboratory leaching tests described in Chapters 3, 4 and 5 involved the assessment of sole aggregate materials. The aim of this chapter is to identify what effect material treatment, binding or stabilisation has on the rate and quantity of leached species.

In practice, many alternative and conventional aggregate materials require some additional stabilisation in order to improve the mechanical properties of the material to enable their use in specific road construction applications. The addition of binders, and hence improved mechanical performance, may also enable raw materials of a lower quality to be used in higher specification applications, thus adding value to the material. Nunes (1997) studied the mechanical performance of various mixtures incorporating primary and alternative aggregates with conventional and alternative binders and observed improvements in mechanical performance characteristics, including resilient and permanent deformation, tensile strength, stiffness and resistance to fatigue.

The concept of material stabilisation has also been widely used in the hazardous waste management industry. Here, solidification and stabilisation processes are used as a method of containment for highly contaminated materials, to enable their safe disposal to land.

Binders can act in many different ways, but are generally thought to include a combination of physical and/or chemical barriers to the passage of both water and chemical species. Physical modification may include reducing material permeability and/or porosity or connected porosity, and coating the surface of the contaminated particles with an impermeable layer. Chemical barriers may include pH modification and other complex interrelated processes [Conner, 1990; Yousuf et al., 1995].

Van der Sloot, et al (1989) deduced that the leaching of potentially hazardous components of stabilised products was reduced by ensuring a high product alkalinity, a small surface-to-volume ratio and a high tortuosity, or low open porosity. Medici et al. (1989) also highlighted the effect of matrix porosity on leaching.

6.2 Selection of Binders and Mixtures

The binders and activators selected for study were summarised in Chapter 3. The composition of the solid fraction and the leaching characteristics of the binders, as described by the rapid leaching characterisation test, were also presented.

The treatment methods that were chosen for study are described in more detail in Section 6.4 of this chapter. The mixtures include a range of physical and chemical modification processes. The selected binders included hydraulic and pozzolanic cementitious binders with appropriate activators, and bituminous binders. The selection of mixtures was principally based upon the mechanically suitably mixtures proposed by Nunes (1997). The industrial partners in this research also recommended a number of mixtures, including the use of bituminous binders, based on current or potential usage in industry.

6.3 Characterisation of the Binders and Activators

As different leaching characteristics were anticipated for the bound mixtures compared to the unbound ones, it was desirable to know what chemical species occur as a consequence of the introduction of a binder and/or activator.

The results of the rapid leaching characterisation test on the hydraulic binders alone have already been described in Chapter 3. This section describes the tests performed on the activated binders. Since the activation of such binders only requires the addition of water (which would not contribute any additional contamination to the sample) a direct comparison could be made between the results of the tests on the activated and non-activated hydraulic binders.

Pozzolanic, or other binders that require chemical activators, were not assessed in this way because the combination of materials required to produce these stabilisation reactions would introduce complexities in the comparison, which were better addressed by the tank-leaching tests described in Section 6.4.

6.3.1 Methodology

Cured mortars of the cement and cement kiln dust were prepared for testing in the rapid leaching characterisation test. The dry materials were mixed with distilled water to form a smooth paste and then poured into plastic moulds, as used for the sample preparation of the compacted tank-leaching test samples. Each sample was sealed inside a plastic bag to prevent loss of moisture and cured for 90 days before testing. The cured samples were removed from the moulds and broken into manageable pieces before being further size reduced for testing, as described in Chapter 3.

6.3.2 Results

The chemical composition of the leachates from the rapid leaching characterisation tests on the cement and cement kiln dust and there mortars are summarised in Table 6-1 within specific concentration ranges.

It can be seen from Table 6-1, that the leachates from the cement mortar showed a slight increase in pH, but similar conductivity when compared to the test on the cement. For the cement kiln dust a similar increase in pH was shown, but a considerably higher conductivity resulted from the test on the mortar, which was not expected.

Table 6-1: Summary of concentration ranges for determinants detected in the leachates from the rapid leaching characterisation test on the hydraulic binders and their cured mortars

Material	**L/S**	**pH**	**Cond. µS/cm**	**Concentration in leachate (mg/l)**							
				<0.01	**0.01-0.1**	**0.1-1**	**1-10**	**10-100**	**100-1000**	**1000-10000**	**>10000**
ce	**2**	12.6	21557	Mn Ni Zn As	NH_4 Si Se	NO_2 Ba Cr	Br F TIC Li	Cl TOC Sr	Ca Na S	K HCO_3 SO_4	
	2-10	12.4	11681	Mn Fe Ni Zn Cd Pb V As Se	Cr	Si Al	Cl SO_4 F TIC Ba Li	Na TOC S Sr	K	Ca HCO_3	
ce-m	**2**	13.4	21357	TOC Mn Se	Br NO_2 Zn	Cl Si	SO_4 S NH_4 Ba Li	TIC Sr	Ca Na	K HCO_3	
	2-10	13.1	11491	TOC Mn Se	NO_2 Zn	Cl F Si Li	SO_4 S Ba Sr	Na TIC	Ca K	HCO_3	
ckd	**2**	12.9	56057	Ni Cd Pb	As	NH_4 Cu Se	NO_2 Ba Fe Al Cr Li	F TOC TIC Sr Zn	Ca Na Br	HCO_3 Cl SO_4 S	K
	2-10	12.4	15331	Ni Zn As Se		NH_4 Si Ba Cr	F TIC Sr Li	Na Br TOC	Ca Cl S	K HCO_3 SO_4	
ckd-m	**2**	13.6	71057	Se	Mn	Ba Fe Co Ni Cu Zn Cr V	Mg NO_2 F Si Sr Al Mo Pb Li B	Br TOC TIC P	Ca Na	Cl SO_4 S	K HCO_3
	2-10	13.0	17421	TOC Se	Zn	Si Ba Mo	NO_2 F P Sr Li	Br TIC	Ca Na Cl SO_4 S	K HCO_3	

Further observations from Table 6-1 reveal that hydration/activation of these binders has resulted in a significant (1 order of magnitude, or greater) reduction in the leached concentrations of a considerable number of species and the increase of only a limited few. For the cement, hydration and curing resulted in a reduction in the concentrations of calcium, total sulphur, chlorine, lithium and chromium in one or both phases of the test. Increased concentrations of barium and silicon were observed. Similarly for the cement kiln dust, the mortar showed significantly lower leached concentrations of bromine, fluorine, strontium, barium, aluminium, iron, zinc, chromium and selenium, but an increase in nickel and lead during the first phase of the test. Such observations were not possible in the second phase of the test since most of these species were below the limits of analytical detection.

6.3.3 Discussion

It is concluded that in the rapid leaching characterisation test a reduction in leached concentrations of a number of species occurred as a result of binder activation. Possible explanations for this reduction in leached species are:

- The cementitious reaction may have encapsulated the species either physically or chemically into the matrix;
- The larger size of the particles of mortar that were tested, compared to the fine-grained dry binder, may have reduced leaching from rapid surface wash-off.

It is apparent that any assessment of leachability from binders or mixtures should be carried out on the materials in the form in which they are to be utilised. Such an assessment will then incorporate the physical and chemical mechanisms of leaching attenuation resulting from the binding process.

6.4 Effect of Binder on Diffusive Leaching

Assessment of diffusive leaching from material mixtures was made using the tank-leaching test method previously described in Chapter 4. This test methodology was developed to enable bound mixtures of materials to be assessed using the same procedure as that described for the tank-leaching tests on compacted materials.

The 8 mixtures that were tested in a bound condition using the tank-leaching test are given in Table 6-2. The materials to be bound cover a range of leaching potentials:

- Relatively high leachability - pulverized-fuel ash and MSW incinerator ash;
- Mid-range leachability - blastfurnace slag;
- Relatively low leachability – china clay sand;
- Unusual leaching patterns - foundry sand (phenolic, brass).

A control mixture of limestone bound with cement was included to provide baseline data for a conventional mixture.

Table 6-2: Material mixtures tested in the tank-leaching tests on bound materials

Material Code	**Major Component**	**Binder / Activator**		
	% by weight	**% by weight**		
BFS+gbfs+li (1)	BFS 84	gbfs 15	li 1	
CCS+ckd+ce (1)	CCS 90	ckd 7	ce 3	
CCS+pfa+li+sc (1)	CCS 80	pfa 15.5	li 4	sc 0.5
FSB+ce	FSB 92	ce 8		
FSB+bi	FSB 95	bi 5		
LS+ce	LS 95	ce 5		
MSW+bi+filler	MSW 67	filler 28.5	bi 4.5	
PFA+fgd+li (1)	PFA 91	fgd 5	li 4	

KEY:

Major component:

BFS = blastfurnace slag
CCS = china clay sand
FSB =foundry sand (phenolic, brass)
LS =limestone
MSW = MSW incinerator ash
PFA = pulverized-fuel ash

Binders/Activators:

bi = bitumen (200pen)
ce = cement
ckd = cement kiln dust
fgd = flue-gas desulphurisation gypsum
filler = 20mm granite + limestone fines
gbfs = granulated blast furnace slag
li = quicklime
pfa = pulverized-fuel ash
sc = sodium carbonate

Note: The limestone aggregate in the LS+ce mix was graded to the specifications for a coarse CBM3 [Specification for Highway Works-Part 3, 1996]

(1) indicates mixture recommended by Nunes (1997) as having suitable mechanical performance properties

The MSW incinerator ash had been screened to remove the fines and ferrous and non-ferrous metals. The ash had then been crushed and graded with the addition of 20mm granite and limestone fines.

6.4.1 Methodology

The tank-leaching tests on bound materials were carried out using the procedure described in Chapter 4 for the compacted material samples. Details of the sample preparation methodology follow:

- For mixtures containing dry binders the minor components were mixed together prior to being added to the major material component;

- A 200-pen bitumen was used in the FSB+bi mixture. This bitumen was selected because it has a high viscosity which enables good mixing at low temperatures (mix prepared at 140°C). However, even at these temperatures it is possible that some, especially organic, contaminants may have volatilised. This grade of bitumen was also chosen because it is relatively inexpensive.
- The MSW+bi+filler mix was prepared in an asphalt batch mixing plant that was supplying this material mixture for commercial use. The tank-leaching test sample was prepared from the same batch of material that was supplied for the lysimeter test described later in Chapter 7. The MSW incinerator ash used in this mixture had been processed before use by screening out the ferrous and non-ferrous metals and the fine fraction of the material. The residual ash had been crushed and graded with the addition of granite fines. This processing is likely, in itself, to have reduced the leaching potential of the material and should be considered in the analysis of the results.

All of the prepared samples were cured for 90 days before testing in the same way as the compacted material samples. This duration of curing was chosen because, as shown by Nunes (1997), most mixtures will have gained the majority of their ultimate strength after such time. This implies that the reactions of the binding process are complete and that any modification to leaching, as a result of binding, will be optimised.

6.4.2 Results

The physical properties of the material mixtures and the conditions established within the tank-leaching tests on the bound materials are summarised in Table 6-3.

The pH of the leachates from the tank-leaching tests on the bound materials are illustrated in Figure 6-1. As expected, all of the mixtures containing cement or lime produce alkali leachates with a pH in the region of 12 to 13. For these mixtures it can be seen that the leachate pH is established rapidly within the first few hours of the test. In contrast, the pH of the leachates from the bitumen bound samples shows significantly less deviation from the pH neutral conditions of the initial leachant. It is thus concluded that the binder has a significant influence over the pH of the leachates.

Table 6-3: Summary of the physical properties and test conditions for the tank-leaching tests on bound materials

Material Code	MDD Kg/m^3	OMC %	Mass of Test Sample Kg	Test L/S by volume	Test L/S by mass
BFS+gbfs+li	2.1	7.0	2.420	10.0	5.12
CCS+ckd+ce	2.0	9.3	2.498	10.0	4.95
CCS+pfa+li+sc	2.0	9.2	2.500	10.0	5.73
FSB+bi	1.9	-	2.280	10.0	5.43
FSB+ce	1.8	9.8	2.220	10.0	5.57
LS+ce	2.2	8.0	2.598	10.0	4.76
MSW+bi+filler	1.6*	-	2.322	10.7	5.71
PFA+fgd+li	1.5	21.2	1.647	10.0	8.80

* sample density

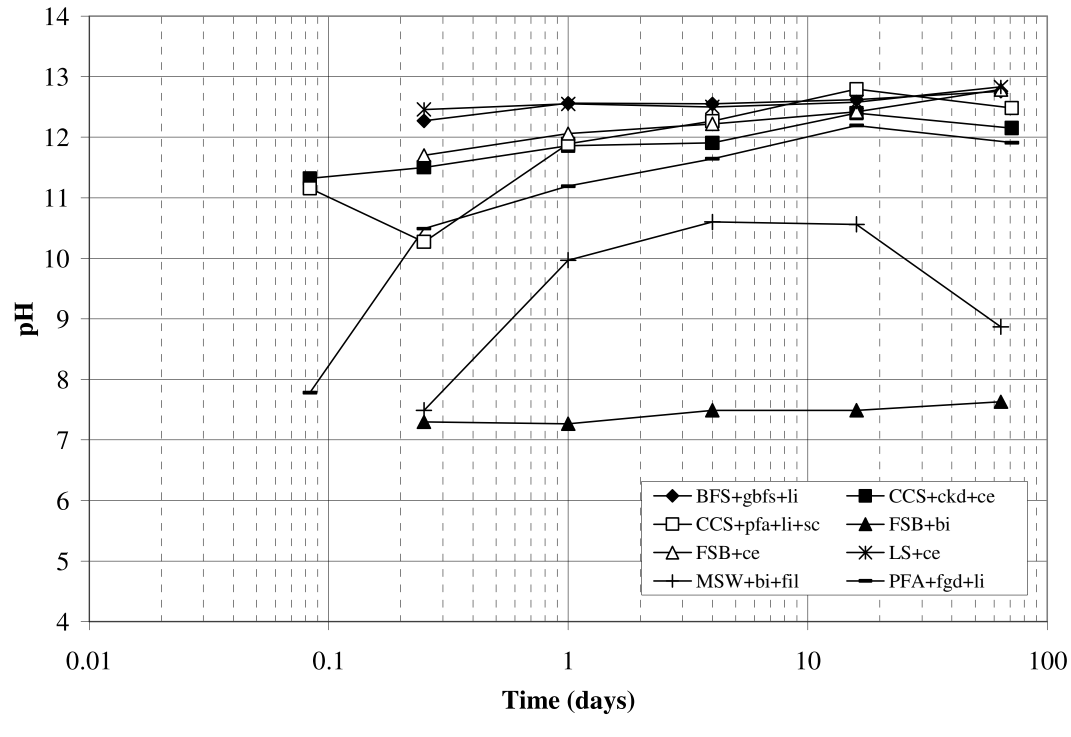


Figure 6-1: pH as a function of time for the tank-leaching tests on bound materials

Figure 6-2 illustrates the conductivity of the leachates from the tank-leaching tests on the bound materials. The influence of the binder is again shown by the lowest conductivity resulting from the bitumen bound materials. The most rapid initial increases in conductivity are shown in the leachates from the BFS+gbfs+li and LS+ce mixtures. Here it is possible that material permeability and/or porosity are playing a part in determining the rate of leaching. These mixtures both use coarse graded aggregates, which result in a matrix with a higher porosity compared to a material with the same mix proportions but a finer graded aggregate. The higher porosity may enable the leachant to permeate to the centre of the compacted sample more rapidly than in, say, a sand and cement mix, where the cement may fill some of the porosity.

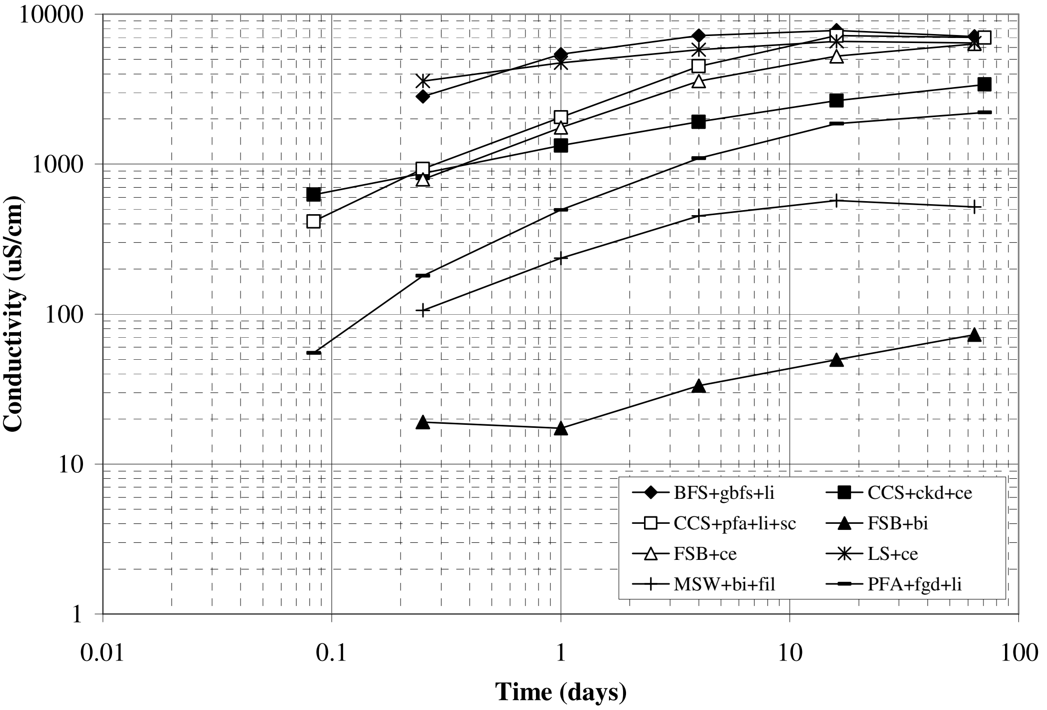


Figure 6-2: Conductivity as a function of time for the tank-leaching tests on bound materials

Figure 6-2 also shows an increasing conductivity in the leachates from all of the materials up until at least day 16 of the test, and in many cases until the completion of the test at day 64. This continued increase in conductivity is a possible indication that significant leaching is occurring throughout the duration of the test. In comparison to the tank-leaching tests on non-compacted and compacted materials, in which peak conductivity was generally observed in the leachates after 4 to 16 days of testing, it may illustrate an increased time to achieve equilibrium conditions. Such behaviour may be an indication of the binder acting to reduce the rate of leaching or that the binder is contributing significant leached species.

6.4.3 Discussion

To give some measure of the relative toxicity of the species in the different leachates from the tank-leaching tests on bound materials, the species concentrations have been normalised, arbitrarily, to the wqm as described in earlier chapters. The results of this comparison are given in Table 6-4, which shows that the majority of species have a normalised concentration of less than unity. No leachates exhibit List II species at normalised concentrations greater than 10, although potassium and/or aluminium are present at such concentrations in the leachates from a number of materials. All mixtures, except CCS+ckd+ce, contain a List II species with a normalised concentration of between 1 and 10.

Table 6-4: Normalisation of data from tank-leaching tests on bound materials to arbitrary water quality maxima

Material Code	Test L/S by mass	Maximum normalised concentration in leachate				
		<1	1-10		10-100	
			II	Other	II	Other
BFS+gbfs+li	5.12	Mg Na Cl NO_3 F P Ba Mn Fe Al Ni Cu Zn Mo Li B As	NH_4	Ca K SO_4 NO_2		
CCS+ckd+ce	4.95	Ca Mg Na Cl SO_4 NO_3 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B As		NO_2		K Al
CCS+pfa+li+sc	5.73	Ca Mg Cl SO_4 NO_3 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Cd Pb V Li B As	Mo	Na NO_2		K Al
FSB+bi	5.43	Ca Mg Na Cl SO_4 NO_3 NO_2 F P Ba Mn Fe Al Ni Cu Zn Cr Mo Pb Li B As	V	K		
FSB+ce	5.57	Mg Na Cl SO_4 NO_3 F P Mn Fe Ni Cu Zn Cr Mo Li B As	NH_4 Ba	Ca NO_2 Al		K
LS+ce	4.76	Mg Na Cl SO_4 NO_3 NO_2 F P Mn Fe Ni Cu Zn Mo Li B As	Ba	Ca K Al		
MSW+bi+filler	5.71	Ca Mg Na Cl SO_4 NO_3 NO_2 F P Ba Mn Fe Ni Cu Zn Cr Mo Pb V Li B As	NH_4	K		Al
PFA+fgd+li	8.80	Ca Mg Na Cl SO_4 NO_3 F P NH_4 Ba Mn Fe Ni Cu Zn Cr Cd Pb Li B As	Mo V	NO_2		K Al

II = List II substance [EEC Directive 76/464/EEC, 1976]

Other = Not List I or List II

To give an indication of the overall toxicity of the different leachates, the normalised species concentrations were summed for each leachate. Previously, in Chapters 3 and

4, this summation has been carried out based on the maximum possible concentrations in the leachates (i.e. <0.1 was taken to be 0.1). However, the materials bound with alkali binders, such as cement and lime, produced leachates with high concentrations of potassium and calcium. These species interfere with the analytical equipment and thus the leachates were quantitatively diluted before the analysis was performed with a consequential increase in the limits of analytical detection. A normalisation based on these higher limits of detection would give the impression that leaching has increased, when, in fact, it probably has not. Thus for the normalisation of this test data, minimum concentrations were used (i.e. <0.1 was taken as zero). The sum of all normalised concentrations is shown graphically in Figure 6-3 for each of the mixtures that were tested. Figure 6-4 shows an equivalent summation of the normalised List I and II species. The effect of normalising to the minimum, rather than the maximum, concentrations can be seen in Figure 6-4 by the apparently irregular patterns of leaching shown by some materials. The seemingly rapid rise and fall in concentrations is an indication of species falling just above and then below the limits of analytical detection. In general, significantly less than 50% of the total normalised concentration can be attributed to List I and II species.

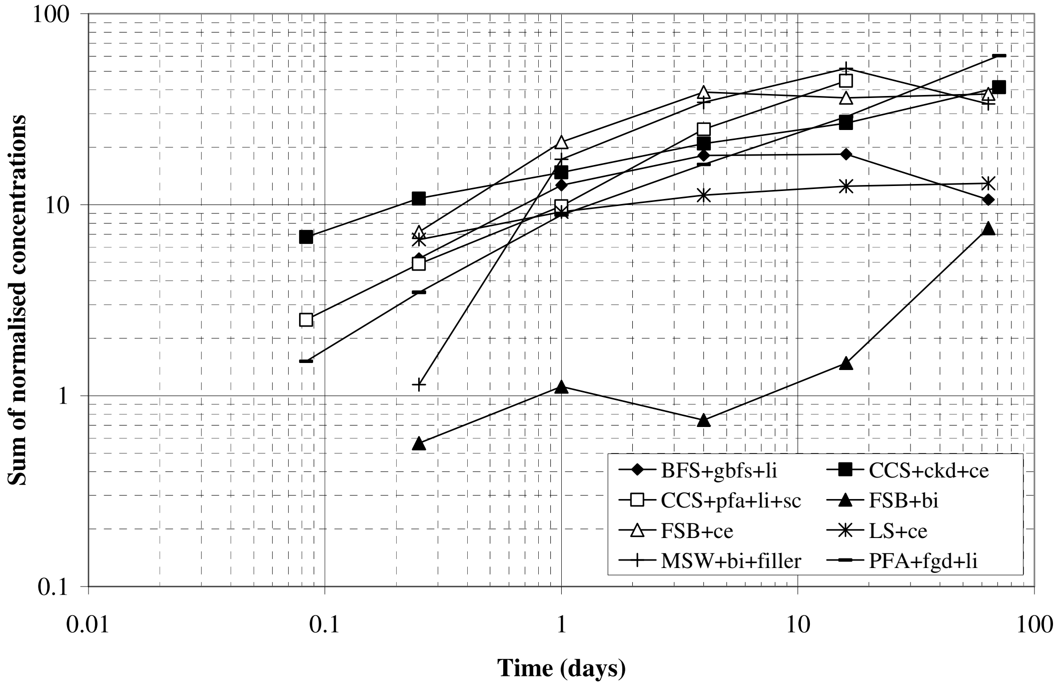


Figure 6-3: Sum of all normalised species concentrations from the tank-leaching tests on bound materials plotted against time

From Figure 6-3 it can be seen that most of the materials leach greater total normalised species than the LS+ce mix. The exceptions to this are the FSB+bi, which leaches significantly less, and the BFS+gbfs+li, which leaches to a similar degree. For the List I and II species, shown in Figure 6-4, the LS+ce appears to falls towards the higher end of the range, although the high detection limits may have artificially reduced the apparent concentrations in some of the leachates. As a traditionally utilised material, if the leaching of the LS+ce mix was used as baseline data to which to compare the leaching of the other materials, then many of these mixtures may be classed as acceptable for use. On a more element specific basis, as shown from Table 6-4, the LS+ce mix leached the highest quantities of the List II species barium. Normalised barium concentrations were in excess of 4 and thus it could be reasoned that if all List I or II species have normalised concentrations less than 4, then a material could be deemed acceptable.

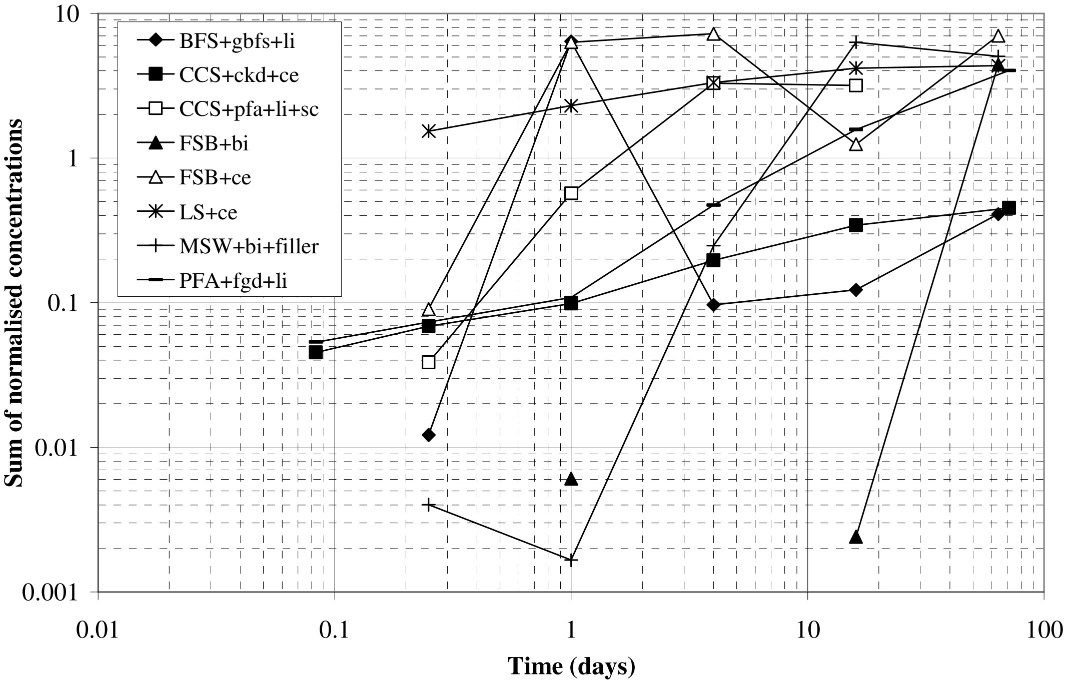


Figure 6-4: Sum of normalised List I and II species concentrations from the tank-leaching tests on bound materials plotted against time

Figure 6-5 shows the sum of the normalised concentrations of all leached species and List I and II species from the tank-leaching tests on the bound materials, compared to the leached species from the aggregate material tested solely in the compacted tank-leaching test.

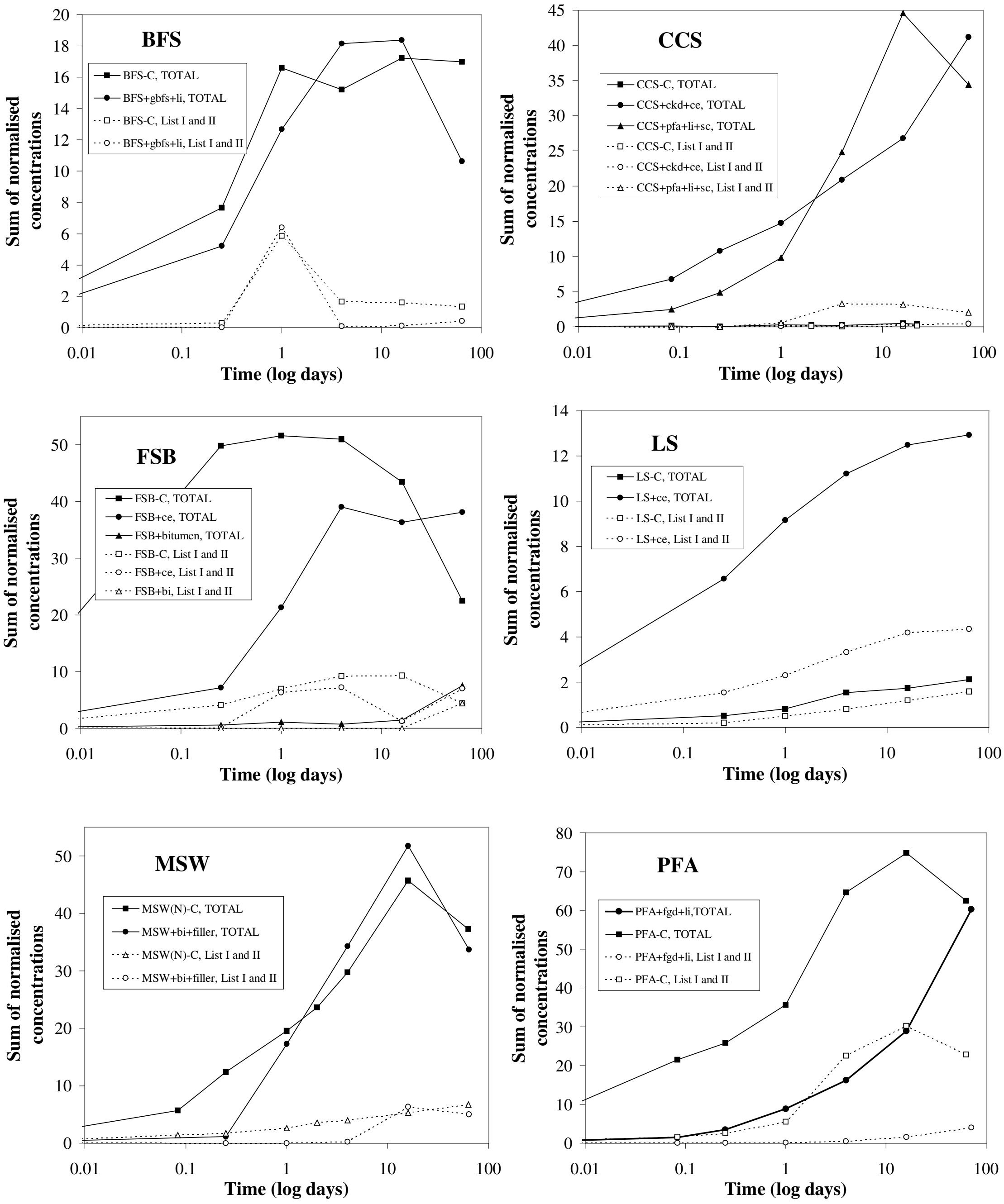


Figure 6-5: Comparison of the sum of normalised species concentrations from the tank-leaching tests on bound materials and the tank-leaching tests on compacted materials, plotted against time

It can be seen from Figure 6-5 that the stabilisation of the pulverized-fuel ash and foundry sand (phenolic, brass) resulted in the leaching of less than 50% of the total leached species, compared to the untreated material, during the early stages of the test. This is a significant indication that binding has decreased the rate of leaching from these materials. For the PFA+fgd+li and the FSB+ce, ultimate total leached species are about 20% lower than from their untreated aggregates and these concentrations were established over a longer period of time. A similar conclusion is drawn for the leaching of List I and II species which shows that both the rate and quantity of leaching have been reduced in these cases. For the FSB+bi, ultimate leached species only account for about 20% of the equivalent species leached from the compacted foundry sand (phenolic, brass). The List I and II species are also lower than from the untreated compacted foundry sand (phenolic, brass).

For the leaching of species from the MSW+bi+filler and the BFS+gbfs+li, Figure 6-5 again shows a reduced leached quantity of both the total and List I and II species, in the early stages of the test in comparison to the sole compacted aggregate material. Ultimate leached quantities are, however, similar.

For the limestone and china clay sand mixtures, total and List I and II leached quantities are significantly (2 to 20 times) higher than from the compacted aggregates when tested solely. For both of these materials, the contributions made to leaching from the sole materials are very small (see Chapter 4). In these mixtures, the binders have actually contributed significantly to the leached species. The CCS+pfa+li+sc and the CCS+ckd+ce exhibit similar total leached quantities, although the pfa+li+sc binder contributes more List I and II species than the ckd+ce stabilisation.

It is evident from the data that the apparent success of the binder in controlling the leaching of the chemical species depends upon the type and quantity of species that were available for release from the original aggregate.

The results of a more detailed examination of the effect of the binder on the cumulative release (mg/kg) of each chemical species in the tank-leaching tests at a given time is summarised in Table 6-5. The assessment shows the change in leached quantity resulting from the addition of the binder to the aggregate. The assessment uses the relationship between the quantity leached in the tank-leaching tests on the bound material and the tank-leaching test on the sole material at equal test durations to quantitatively determine whether the leached quantity has increased or decreased as a result of the addition of the binder. In Table 6-5 increases in leached quantity resulting from the addition of a binder are shown by a ‘+’ symbol and decreases by a ‘-’ symbol. The magnitude of the change is illustrated by the number of symbols, with a greater number indicating a greater change (See Table 6-5 – Key).

The material stabilisation appears to be most effective at reducing leaching from those materials that leached detectable quantities when tested solely in a compacted form. For such materials, the methods of stabilisation had varying degrees of success, but were overall beneficial to reducing leaching. For the limestone and china clay sand aggregates, which demonstrated few available species when tested solely, the stabilisation methods are actually shown to introduce leachable species. It would therefore be apparent that the decision to carry out stabilisation of aggregates for use in the road pavement should be made by consideration of both mechanical and environmental performance. It is also of upmost necessity for any assessment of leachability to be a reflection of the whole or stabilised product and not of the individual parts.

In order to be able to make recommendations for the appropriate stabilisation of any material, it is necessary to have an understanding of the mechanisms that have contributed to the leaching behaviour of the mixtures shown above. To examine whether pH modification, as a result of the binding process, reduced the leaching of certain species, the pH of the leachates from the tank-leaching tests on the bound mixtures was compared to that for the unbound materials, as illustrated in Figure 6-6. This figure also shows the conductivity of the leachates from both the tank-leaching tests on the compacted sole material and the bound mixture.

Table 6-5: Matrix summarising the effect on leached quantity of the addition of binders to aggregate materials, as observed from the tank-leaching tests on bound and compacted materials

Aggregate	**BFS**	**CCS**	**CCS**	**FSB**	**FSB**	**LS**	**MSW**	**PFA**
Binder	**+gbfs+li**	**+ckd+ce**	**+pfa+li**	**+bi**	**+ce**	**+ce**	**+bi+filler**	**+fgd+li**
pH	+	+	+	=	+	+	=	=
Cond.	++	+++ / ++++	++++	---	+ / ++	+++	--	+ / =
Ca	+	+++ / ++++	+++ / ++++	- / --?	+++ / ++++	+++	- / --	-
Mg	-?	-?	-?	--	-?	-?	= / -	-?
Na	= / -	+++	+++++	--- / ----	= / -	+++	--	- / --
K	=	++++	+++	---	-	+++	--	=
HCO_3	+?	++?	+ / ++?	---	+	+++	-	++ / +
Cl	= / -	++++	+++	- / +?	=?	= / -	---	--
SO_4	--	++	++	-- / ---	- / --	=	= / -	-- / ---
NO_3	+?	X	X	-?	-	--	+?	X
Br	-?	+?	+?	-?	--	X	--	--
NO_2	+ / ++	+?	+?	+?	+?	= / +	- / --	= / -
F	= / -	=	=	+?	X	=	--	- / --
TOC	= / +	+?	+?	- / --?	-	++?	--- / ----	=?
TIC	++	= / -	= / +	-- / ---	---	--	= / -	=
Total P	X	X	X	-?	-?	-?	--	=
Total S	-	++	++	---	--	= / +	=	-- / ---
NH_4	=	-?	= / +	- / --?	= / -	X	= / +	-?
Si	--- / ----	+ / ++	+ / ++	---	-	= / +	--	-
Ba	+?	+?	+?	X	+ / ++?	++	-?	--
Sr	+	+?	+?	-?	+++ / ++++	+++	--	--
Mn	---	-?	-?	-- / ---	-- / ---	-?	= / +	X
Total Fe	X	X	X	---	---	X	X	-?
Al	= / -	+++	++++	= / -	++	++	= / +	- / --
Co	X	X	X	+?	X	X	X	X
Ni	X	X	X	+?	-?	X	X	X
Cu	X	X	X	=	-?	X	----	X
Zn	+?	=?	=	=	-	+ / ++	-	- / --?
Cr	X	X	+?	X	X	X	X	-?
Mo	X	X	+?	+?	X	X	- / --	---
Cd	X	X	X	+?	X	X	X	X
Pb	X	X	+?	+?	X	X	X	X
V	-?	X	+?	+?	X	X	X	--
Li	-	+?	+?	=	+++	+?	- / --	- / --
B	--	X	+?	X	X	X	+ / ++?	- / --?
As	X	-?	+	X	X	X	-?	-- / ---
Se	X	++	++	X	X	X	-?	--

Key:

Symbol	+++++	++++	+++	++	+	=	-	--	---	----	-----
Cumulative release (mg/kg) from: bound material / unbound material	>1000	1000 to 100	100 to 10	10 to 2	2 to 1.25	1.25 to 0.8	0.8 to 0.5	0.5 to 0.1	0.1 to 0.01	0.01 to 0.001	<0.001

X	Effect of binder cannot be defined except that it does not cause a large increase in leaching
- ?	Binding results in a possible decrease in leaching
+ ?	Binding results in a possible increase in leaching
/	Denotes borderline values
=	Denotes no change

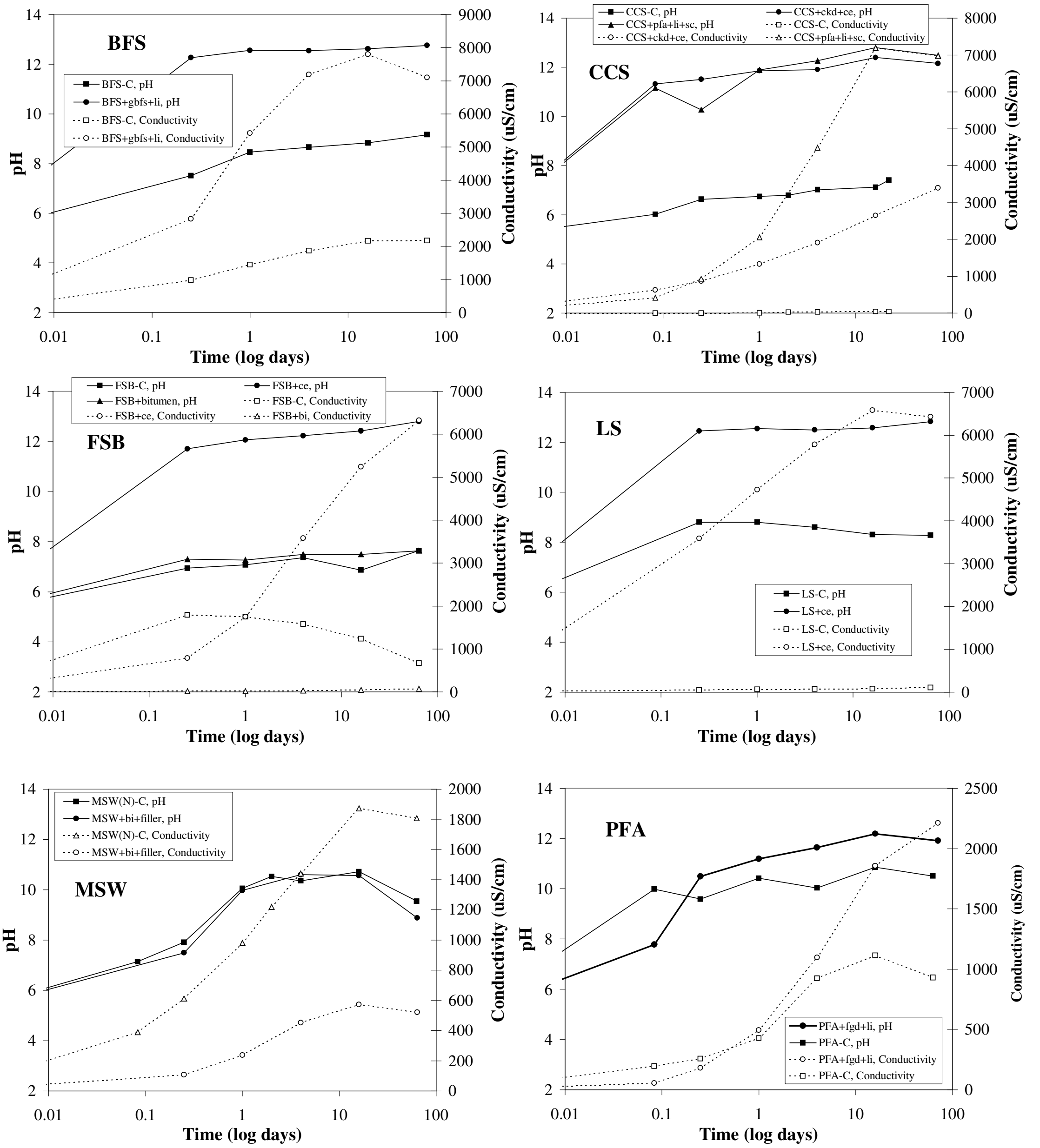


Figure 6-6: Comparison of the pH and conductivity of the leachates from the tank-leaching tests on bound materials and the tank-leaching tests on compacted materials, plotted against time

The mixtures for which pH modification occurred as a result of the binder were BFS+gbfs+li, CCS+ckd+ce, CCS+pfa+li+sc, FSB+ce and LS+ce. This was expected since at least one component of each of these binders is alkali in nature and thus caused the pH of the leachates from these mixtures to be alkali (in the pH range of 11 to 12). The lime in the PFA+fgd+li mixture generated leachates that were slightly more alkali than the sole pulverized-fuel ash which itself produced a leachate pH of nearly 11. The MSW incinerator ash and foundry sand (phenolic, brass) mixtures bound with bitumen, have a pH in the same region as the bulk material tested solely.

The evidence of whether pH modification has exerted control over leaching by altering species solubility is questionable. There is insufficient evidence to conclude either way although the alkali leachates generated by many of the binders are unlikely to have an adverse effect on the leaching of most species.

From Figure 6-6, it can also be observed that all of the mixtures that exhibit pH modification into the alkali region as a result of the binder also show substantial increases in the conductivity of those leachates. This results from the leaching of large quantities of the alkali and alkali earth metals from the binders (observed in Table 6-5). These species contribute to both pH modification and conductivity but generally have water quality criteria set at relatively high concentrations and therefore, do not substantially affect the sum of normalised species concentrations.

The bitumen binder appears to act as a waterproof boundary between the aggregate material and the leachant. This binder appears to reduce the leaching of the majority of the detectable species in the leachates from these materials, including the alkali and alkali earth metals that are so abundant in the alkali binders.

6.5 Influence of Binder on Advective Leaching

An assessment of advective leaching from the material mixtures was also made by use of the large horizontal permeameter and DoRLaP as described in Chapter 5. These permeameter tests were designed to enable the bound mixtures to be tested using the same procedures as those described for the compacted aggregates.

The bound mixtures tested in the permeameters are listed in Table 6-6. A total of 4 mixtures were tested. Fewer bound materials were tested in the permeameters than in the tank-leaching tests because only a single piece of large horizontal permeameter apparatus was available for use. Thus it was not possible to test samples that required curing in this apparatus.

Table 6-6: Material mixtures tested in the permeameters on bound materials

Material Code	Apparatus	Major Component		Binder / Activator			
		% by weight		% by weight			
CCS+ckd+ce (1)	DoRLaP	CCS	90	ckd	7	ce	3
FSB+bi	DoRLaP	FSB	95	bi	5		
FSB+ce	DoRLaP	FSB	92	ce	8		
MSW+bi+filler	Large horizontal perm	MSW	67	filler	28.5	bi	4.5

KEY:

Major component:

CCS = china clay sand
FSB =foundry sand (phenolic, brass)
MSW = MSW incinerator ash

Binders/Activators:

ce = cement
ckd = cement kiln dust
bi = bitumen (200pen)
filler = 20mm granite + limestone fines

Note: The MSW incinerator ash had been screened to remove the fines and ferrous and non-ferrous metals. The ash had then been crushed and graded with the addition of 20mm granite and limestone fines.

(1) indicates mixture recommended by Nunes (1997) as having suitable mechanical performance properties

6.5.1 Methodology

The permeameter tests on bound materials were carried out using the procedure described in Chapter 5 for the compacted material samples. Details of the sample preparation methodology follow:

- The MSW+bi+filler test specimen was prepared by reheating, remixing and compacting a sub-sample of the batch-mixed material that was used in the lysimeter (see Chapter 7).
- The remaining materials were prepared as described in Section 6.4.1 and compacted into the DoRLaP leaching vessel as described in Chapter 5 for the DoRLaP tests on the unbound materials. The DoRLaP samples were cured for 90 days before testing.

6.5.2 Results

The physical properties of the mixtures and the conditions established within the permeameter tests on the bound materials are summarised in Table 6-7. The calculated permeabilities of the materials are given in Table 6-8.

Table 6-7: Summary of the physical properties and test conditions for the permeameter tests on bound materials

Material Code	MDD Kg/m^3	OMC %	Specific Gravity Mg/m^3	Mass of Test Sample Kg	Void ratio	Flow rate Mins/bed volume
CCS+ckd+ce	2.0	9.3	2.70	2.366	0.25	15
FSB+bi	1.9	-	2.25	2.120	0.20	7.5
FSB+ce	1.8	9.8	2.63	2.083	0.33	7.5
MSW+bi+filler	2.1*	-	2.50	56.78	0.16	15

* sample density

Table 6-8: Summary of the calculated permeability of the mixtures

Material Code	Permeability (m/s)			
	BV 2	BV 5	BV 10	BV 20
CCS+ckd+ce	1.8E-05	2.4E-05	3.2E-05	5.8E-05
FS+ce	7.9E-06	1.0E-05	1.4E-05	2.8E-05
FS+bi	1.0E-04	9.8E-05	3.0E-05	5.8E-06
MSW+bi+filler	1.1E-03	1.1E-03	1.1E-03	3.0E-03

The pH of the leachates from the permeameter tests on the bound mixtures are shown in Figure 6-7. The cement treated mixtures again exhibit significantly alkali leachates with a pH in the region of 12 to 13, which is similar to that shown in the tank-leaching tests on these mixtures. Again, the materials bound with bitumen show a much less significant change in leachate pH from that shown by the sole aggregate materials. As the number of bed volumes of leachate passing through the sample increases, the pH of the leachate decreases marginally.

The conductivity of the leachates from the permeameter tests on the bound mixtures verses the number of bed volumes of leachate passed is illustrated in Figure 6-8. The cement treated mixtures result in leachates with the highest conductivity, probably as a result of the large quantities of the highly soluble alkali and alkali earth metals available in the cement. The measured conductivity diminishes with an increase in the number of bed volumes of leachate passing. Such a pronounced trend provides an indication of initial advective leaching with a rapid decline in the availability of these species, followed by slower elements of diffusive leaching.

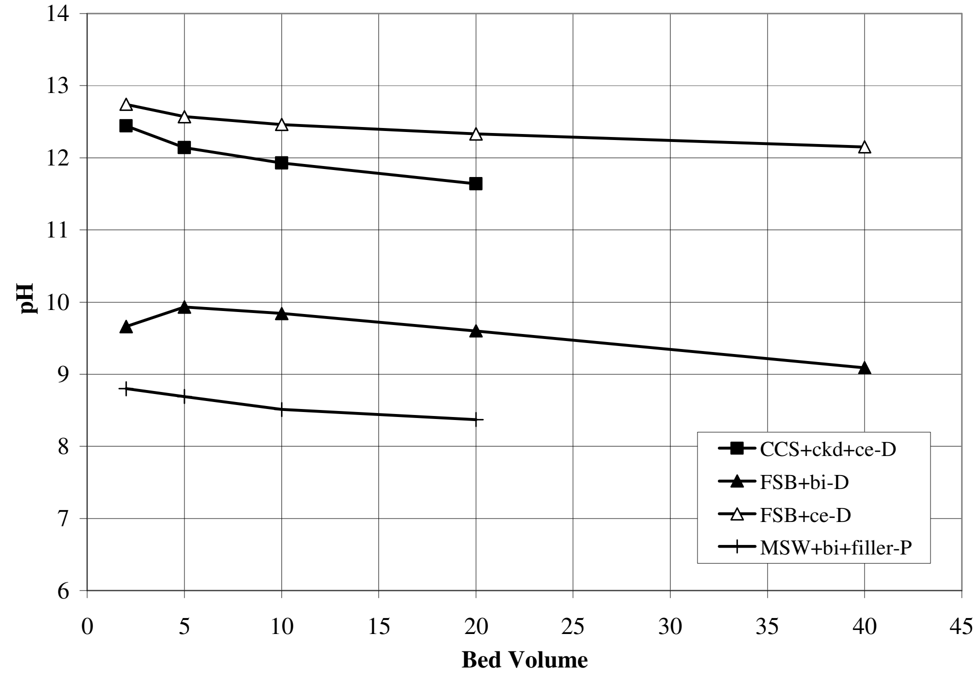


Figure 6-7: pH as a function of the number of bed volumes of leachate passed through the permeameter tests on bound materials

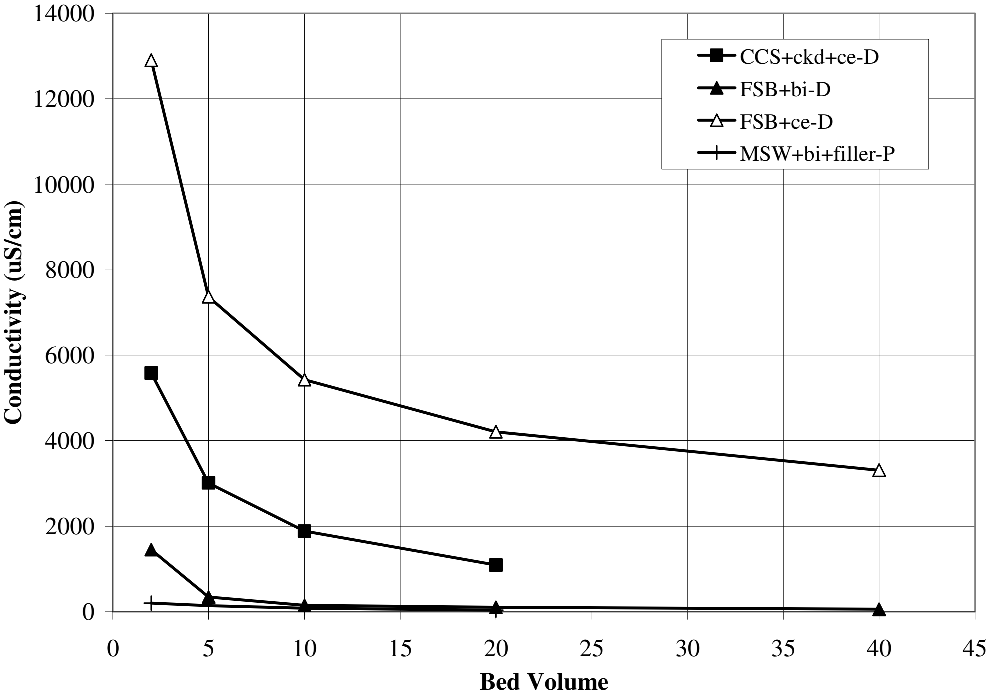


Figure 6-8: Conductivity as a function of the number of bed volumes of leachate passed through the permeameter tests on bound materials

6.5.3 Discussion

Once again, to give some measure of the relative toxicity of the species in the leachates from the permeameter tests on bound materials, the species concentrations have been normalised, arbitrarily, to the water quality maxima as first described in Chapter 3. The results of this comparison are given in Table 6-9.

From Table 6-9 it can be seen that the majority of chemical species have a normalised concentration less than unity. Those species which have a normalised concentration greater than unity are generally the same species that exhibited such leaching in the tank-leaching test on the same bound material.

Table 6-9: Normalisation of concentrations of chemical species in the leachates from permeameter tests on bound materials to arbitrary water quality maxima

Material Code	Test	Maximum normalised concentration in leachate					
		<1	1-10		10-100		>100
			II	Other	II	Other	
CCS+ckd+ce	D	Ca Na Cl SO_4 NO_3 NO_2 F P Ba Mn Fe Li	NH_4	Al		K	
FSB+bi	D	Mg Na Cl SO_4 P Ba Mn Fe Al Cu Li	NH_4			K	
FSB+ce	D	Cl SO_4 NO_3 Mn Fe Ni Li	Ba	Ca Na NO_2 Al	NH_4		K
MSW+bi+filler	P	Ca Na K Cl SO_4 NO_3 F Mn Ni Zn Li B		NO_2 Al			

The results of a more detailed examination of the effect of the binder on the cumulative release (mg/kg) of each chemical species in the permeameter tests at a given L/S ratio is summarised in Table 6-10. The method of assessment is the same as that used in the assessment of the relationship between the quantity leached in the tank-leaching tests on the bound material and the tank-leaching test on the sole material. The assessment uses the relationship between the quantity leached in the permeameter tests on the bound material and the permeameter test on the sole material at equal L/S ratios to quantitatively determine whether the leached quantity has increased or decreased as a result of the addition of the binder. In Table 6-10 increases in leached quantity resulting from the addition of a binder are shown by a '+' symbol and decreases by a '-' symbol. The magnitude of the change is illustrated by the number of symbols, with a greater number indicating a greater change (See Table 6-10 – Key).

Table 6-10: Matrix summarising the effect on leached quantity of the addition of binders to aggregate materials, as observed from the permeameter tests on bound and compacted materials

Aggregate	**CCS**	**FSB**	**FSB**	**MSW**
Binder	**+ckd+ce**	**+bi**	**+ce**	**+bi+filler**
pH	+	-	+	=
Conductivity	+++	--	+++	=
Ca	++ / +++	X	+ / ++?	+?
Mg	-?	+?	X	X
Na	+++	=	++	--
K	+++	=	++	-
HCO_3	+++	= / -	++ / +++	X
Cl	++++	- / --	++	-
SO_4	++ / +++	= / -	+ / ++	++
NO_3	+ / ++?	-?	- / --	=
Br	+ / ++?	+?	+ / ++?	-
NO_2	+?	-?	++	+?
F	-	- / --?	- / --?	+
TOC	+ / ++?	++	+++	X
TIC	= / -	= / -	---	X
Total P	++	= / -	- / --?	X
Total S	++ / +++	++	++	+++
NH_4	+?	+?	+ / ++?	- / --?
Si	= / -	--	= / +	= / +
Ba	+++	=	++++	X
Sr	+++	-	+++++	+?
Mn	= / -	-	=	+++
Total Fe	+?	-	--	X
Al	++ / +++	--	= / +	-
Co	X	X	X	X
Ni	X	X	+?	+?
Cu	X	+	-?	X
Zn	X	X	X	+?
Cr	X	X	X	X
Mo	X	X	X	-?
Cd	X	X	X	X
Pb	X	X	X	X
V	X	X	X	X
Li	+++	+?	+?	+
B	X	X	X	= / +
As	X	X	X	X
Se	X	X	X	X

Key:

Symbol	+++++	++++	+++	++	+	=	-	--	---	----	-----
Cumulative release (mg/kg) from: bound material / unbound material	>1000	1000 to 100	100 to 10	10 to 2	2 to 1.25	1.25 to 0.8	0.8 to 0.5	0.5 to 0.1	0.1 to 0.01	0.01 to 0.001	<0.001

X	Effect of binder cannot be defined except that it does not cause a large increase in leaching
- ?	Binding results in a possible decrease in leaching
+ ?	Binding results in a possible increase in leaching
/	Denotes borderline values
=	Denotes no change

Note: MSW+bi+filler comparison to MSW incinerator ash was generally inconclusive because the MSW incinerator ash test in the permeameter was largely unsuccessful because of the low permeability of this material when compacted.

The comparison between the leaching of the sole and bound materials in the permeameter tests is inhibited by the large number of chemical species observed at concentrations below the limits of analytical detection. The relatively short residence time of the leachate within the material sample in the permeameter tests is thought to be the reason for the lower concentrations. However, a number of observations can be made from Table 6-10.

- The cement based binders resulted in an increase in leachate pH. Conductivity increased by 1 to 2 orders of magnitude and the concentrations of most of the alkali and alkali earth metals increased by up to 3 orders of magnitude;
- The bitumen bound materials showed similar or reduced pH and conductivity compared to the unbound material. A slight increase in sulphur, and potentially manganese and copper was observed. All other species were leached at similar or lower amounts than in the unbound material.

6.6 Conclusions

The results of the all the laboratory leaching tests on the bound materials were compared to the results achieve from the same tests on the unbound aggregates. The results for the comparison based on the tank-leaching test results were given in Table 6-5 whilst the comparison based on the permeameter test results is presented in Table 6-10. In order to define the probable effect of binding on each aggregate material the results of these two assessments were compared. The results are contained in Table 6-11

6.7 Summary

- This chapter investigated the effect that treating aggregate materials with a range of binders had on the leaching properties of the material.

- The rapid leaching characterisation test was used to compare the leaching properties of unhydrated hydraulic binders with those of the cured mortars of these binders. The results showed significant reductions in the leaching of many species from the cured mortars when compared to the unhydrated binder. It was concluded that any assessment of leachability from binders or mixtures should be carried out on the materials in the form in which they are to be utilised.

Table 6-11: Matrix summarising the effect of the addition of binder treatments to aggregate materials on cumulative release (mg/kg), based on an assessment of tank-leaching test and permeameter test data

Aggregate	**BFS**	**CCS**	**CCS**	**LS**	**FSB**	**FSB**	**MSW**	**PFA**
Binder	**+gbfs+li**	**+ckd+ce**	**+pfa+li**	**+ce**	**+bi**	**+ce**	**+bi+filler**	**+fgd+li**
pH	+	+	+	+	= / -	+	=	=
Cond.	++	+++ / ++++	++++	+++	-- / ---	+ / +++	= / --	+ / =
Ca	+	++ / ++++	+++ / ++++	+++	-	+++ / ++++	- / --	-
Mg	-?	-?	-?	-?	--	-	= / -	-?
Na	= / -	+++	+++++	+++	= / ----	++ / -	--	- / --
K	=	+++ / ++++	+++	+++	= / ---	++ / -	- / --	=
HCO_3	+?	++ / +++	+ / ++?	+++	= / ---	+ / +++	-	++ / +
Cl	= / -	++++	+++	= / -	- / --	= / ++	- / ---	--
SO_4	--	++ / +++	++	=	= / ---	++ / --	++ / -	-- / ---
NO_3	+?	= / +	X	--	-	- / --	= / +	X
Br	-?	+?	+?	X	=	--	- / --	--
NO_2	+ / ++	+?	+?	= / +	=	++	- / --	= / -
F	= / -	= / -	=	=	=	-	+ / --	- / --
TOC	= / +	+?	+?	++?	++	+++ / -	--- / ----	=?
TIC	++	= / -	= / +	--	= / ---	---	= / -	=
Total P	X	= / +	X	-?	= / -	-	--	=
Total S	-	++ / +++	++	= / +	++ / ---	++ / --	+++ / =	-- / ---
NH_4	=	=	= / +	X	= / -	= / -	=	-?
Si	--- / ----	= / ++	+ / ++	= / +	-- / ---	=	+ / --	-
Ba	+?	+++	+?	++	=	++++	-	--
Sr	+	+++	+?	+++	-	+++ / +++++	--	--
Mn	---	= / -	-?	-?	- / ---	= / ---	= / +++	X
Total Fe	X	+?	X	X	- / ---	-- / ---	X	-?
Al	= / -	++ / +++	++++	++	= / --	+	=	- / --
Co	X	X	X	X	= / +	X	X	X
Ni	X	X	X	X	= / +	=	= / +	X
Cu	X	X	X	X	= / +	-	----	X
Zn	+?	=	=	+ / ++	=	-	-	- / --?
Cr	X	X	+?	X	X	X	X	-?
Mo	X	X	+?	X	=	X	- / --	---
Cd	X	X	X	X	=	X	X	X
Pb	X	X	+?	X	=	X	X	X
V	-?	X	+?	X	=	X	X	--
Li	-	+++	+?	+?	=	+++	+ / --	- / --
B	--	X	+?	X	X	X	= / +	- / --?
As	X	-	+	X	X	X	-	-- / ---
Se	X	+	++	X	X	X	-	--

Key:

Symbol	+++++	++++	+++	++	+	=	-	--	---	----	-----
Cumulative release (mg/kg) from: bound material / unbound material	>1000	1000 to 100	100 to 10	10 to 2	2 to 1.25	1.25 to 0.8	0.8 to 0.5	0.5 to 0.1	0.1 to 0.01	0.01 to 0.001	<0.001

X	Effect of binder cannot be defined except that it does not cause a large increase in leaching
- ?	Binding results in a possible decrease in leaching
+ ?	Binding results in a possible increase in leaching
/	Denotes range of values
=	Denotes no change

- Tank-leaching tests were used to assess diffusive leaching from bound materials. 8 material mixtures were investigated incorporating hydraulic, pozzolanic and bituminous binders. The tank-leaching tests proved simple to perform on the bound materials.

- The results of the tank-leaching tests on the bound mixtures were compared to those of the tank-leaching tests on the sole aggregate. The aggregates bound with the cement and lime based binders produced leachates with a high pH and increased conductivity and alkali and alkali earth metals. Materials bound with bitumen showed similar or lower pH and conductivity than that shown by the unbound material and a reduction in the leaching of many other chemical species.

- Permeameter tests were used to assess advective leaching from bound materials. 4 material mixtures were investigated incorporating hydraulic and bituminous binders. The low permeability of some of the bound materials made it difficult to maintain the required flow rates through some of the materials.

- The results from the permeameter testing showed that aggregates bound with the cement and lime based binders produced leachates with a high pH, in the order of 11 or 12. Compared with the unbound aggregate binding with cement or lime produced increases in conductivity and the leached quantity of most of the alkali and alkali earth metals. Materials bound with bitumen showed similar or lower pH and conductivity than that shown by the unbound material.

- The results from the tank-leaching tests and permeameter tests on the bound materials were compared with the results of the tests on the unbound aggregates. In many cases, binding reduces the quantity and concentration of chemical species in the leachate. However, for materials that leach minimal quantities of chemical species when tested in the unbound form, binding can actually significantly increase the quantity of leached species.

- The relationship between the results of the unbound and bound material tests was used to produce a matrix illustrating the effect of binding on the leaching of each chemical species from the mixtures tested. This matrix can be used to suggest appropriate binder treatments to use on aggregate materials that leach an excess of a particular chemical species in order to reduce the level of leaching to acceptable levels.

7 Large-Scale Simulation

7.1 Introduction

Chapters 3, to 6 of this thesis described the details of laboratory scale testing of alternative aggregate materials and the interpretation of the results of these tests. The effect of treating a range of materials with selected binders was examined in Chapter 6. This bench-top laboratory testing, carried out under controlled conditions, enabled the investigation of the effects of physical properties of the materials on the leaching process.

The research presented in this chapter examines leaching from aggregates under conditions replicating the in-situ environment of a road construction or bulk fill. This chapter describes the development of a set of large-scale lysimeter tests and discusses the results attained. The results from this large-scale testing are also used later in Chapter 8 as a tool to examine whether the laboratory tests provide a simulative representation of the real leaching environment.

The laboratory tests applied in Chapters 3 to 6 were specifically selected and modified to produce what was thought to be a closer representation of the in-situ construction scenario than that presented by many commonly used leaching test methodologies. However, in theory the laboratory tests may not provide a complete representation of the real case because:

a) The test samples have smaller physical dimensions compared to that of a road construction layer or bulk fill application;
b) External environmental considerations, such as temperature, biological action and periodic wetting and drying of the material, are not represented;
c) The duration of the tests is relatively short compared to the life of a construction in the field and do not, therefore, replicate any possible ageing and deterioration of the materials over time;
d) The leachant flow rate and/or liquid to solid ratio is high compared to that expected in the field, given the probable in-situ permeability of compacted aggregates and the surfacing of the complete road structure.

7.2 Development of the Large-Scale Simulations

In the development of the large-scale simulations the following physical and environmental factors were considered to be important requirements:

- The materials and mixtures should be tested at gradings and levels of compaction that are realistic for the material as placed in a road construction;
- The dimensions of the test sample should be of sufficient size to represent a road construction layer;
- The natural environmental conditions of the in-situ scenario should be represented, including:
  - periodic wetting and drying of materials resulting from precipitation events;
  - changes in temperature;
  - biological action;
  - material degradation and ageing with time;
- The likely hydraulic regime should be represented;
- The results of the large-scale simulation should be comparable to the laboratory leaching tests.

Ideally, a large-scale simulation to determine how alternative aggregate materials leach in the in-situ environment would involve monitoring the leachate generated from real roads constructed with such materials. However, the development of such a simulation is hindered due to:

- The large inputs of time and resources required to construct the simulations;
- The limit on the number of materials that it would therefore be possible to examine in this way;
- The long duration of testing (potentially many years) expected to be required in order to achieve sufficient through-flow of leachate;
- The limited control over instrumentation and sampling from such simulations.

It was therefore decided that, for this research, a large-scale test, or lysimeter, was more appropriate for examining in-situ leaching performance than the monitoring of real road constructions.

Three hydraulic scenarios were considered for simulation in the lysimeters, as described and illustrated in Figure 7-1.

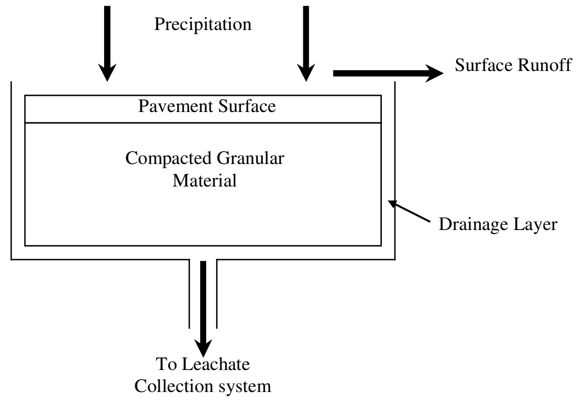
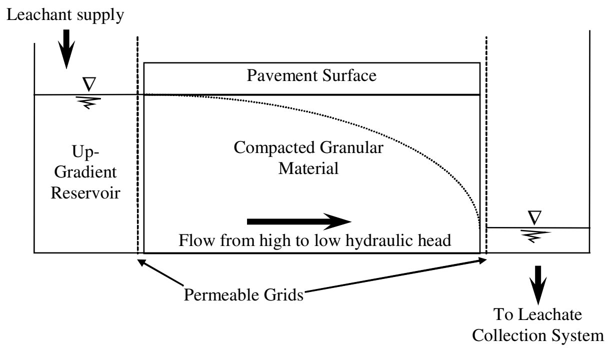
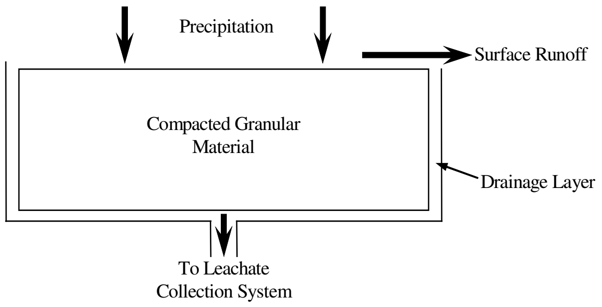
1. Infiltration through the paved surface of a typical road construction during the life of the pavement	
2. Groundwater flow through a typical road construction during the life of the pavement	
3. Infiltration into the granular materials of a typical road during pavement construction or at end-of-life (deteriorated pavement surface)	

Figure 7-1: Hydraulic scenarios considered for lysimeter tests

It was decided that each lysimeter test should be performed using the same leaching scenario because this would enable the largest number of materials and mixtures to be tested and would also enable all results to be compared directly. Hydraulic scenario 3 (see Figure 7-1) was selected for representation in the lysimeter tests. This scenario represents an unsurfaced compacted aggregate exposed to natural environmental conditions during the construction phase of a pavement and also the end-of-life pavement where the road surfacing is degraded and cracked. This scenario was considered favourable since:

- It represents the 'worst case' scenario in terms of the quantity of precipitation that would permeate through the test material, and thus it was considered likely that permeate would be collected during the test period;
- It could be established outdoors with leachant supplied by precipitation rather than artificial watering;
- It would not introduce additional variables associated with the practicality of constructing a consistent road surfacing on top of each compacted material (as would be required for scenario 1);
- The construction and monitoring of the scenario would be relatively simple and the degree of comparability between tests would therefore be high.

7.3 Design and Construction of the Lysimeters

The lysimeters were designed to contain a 350mm depth of compacted aggregate material, which was selected to be typical of the depth of sub-base used in the UK. The exposed surface of the compacted aggregate was selected as $1m^2$ (i.e. unit area) for simplicity and to provide a reasonable surface area of compacted aggregate.

The lysimeters were fabricated from a series of stacking inspection chamber square sections that were manufactured from recycled plastic. Each lysimeter was waterproofed by lining with polythene sheeting. The drainage system in each cell consisted of a concrete base that sloped gently towards a central channel. The channel was formed from half a plastic pipe that ran across the cell and passed through the lysimeter wall into a pipe that carried the leachate to the collection vessel. The concrete base in each cell was covered with waterproof sheeting, a layer of gravel and a sheet of geotextile before compacting the aggregate material in layers above. The top surface of the compacted material was covered in a layer of gravel to prevent spatter.

A number of schematic drawings showing the construction details of the lysimeters are given in Appendix B. Further details of the construction and operation of the lysimeters are given below.

7.3.1 Aggregate Materials and Mixtures

Nine aggregate materials and mixtures were selected for testing within the lysimeters, as summarised in Table 7-1. The materials and mixtures were selected to cover a range of gradings, material types and mixtures as tested in the laboratory leaching tests in Chapters 3, 4, 5 and 6.

Table 7-1: Summary of the aggregate materials and mixtures tested in the lysimeters

Material Code	**Major Component**		**Binder / Activator**			
	% by weight		**% by weight**			
Aggregates						
BFS	BFS	100				
CRR	CR	100				
FSB	FSB	100				
LS	LS	100				
MSW(N)	MSW(N)	100				
Mixtures						
BFS+gbfs+li	BFS	84	gbfs	15	li	1
CCS+ckd+ce	CCS	90	ckd	7	ce	3
LS+ce	LS	95	ce	5		
MSW+bi	MSW	67	filler	28.5	bi	4.5

KEY:

Major component:

BFS = blastfurnace slag
FSB =foundry sand (phenolic, brass)
MSW(N) = MSW incinerator ash (new)
CCS = china clay sand
LS =limestone
CRR = crumbed rubber

Binders/Activators:

bi = bitumen (200pen)
ckd = cement kiln dust
gbfs = granulated blast furnace slag
ce = cement
filler = 20mm granite + limestone fines
li = quicklime

Note: The limestone aggregate in the LS+ce mix was graded to the specifications for a coarse CBM3 [Specification for Highway Works-Part 3, 1996]

The MSW incinerator ash had been screened to remove the fines and ferrous and non-ferrous metals. The ash had then been crushed and graded with the addition of 20mm granite and limestone fines.

7.3.2 Lysimeter Site

The lysimeter field was located at the Sutton Bonington Campus of the University of Nottingham. The site was on level ground that was easy to excavate with access to an electricity and water supply, which was required during the construction. A ground survey for underground services was carried out prior to any excavation.

7.3.3 Lysimeter Base

A construction platform was built to support the lysimeter cells by excavating a trench approximately 17m x 1.7m x 0.21m deep and backfilling with 150mm of compacted aggregate (crushed concrete and brick) using a vibrating plate compactor. The compacted aggregate was overlaid with a 30mm layer of compacted sand to be used as a bedding layer for the lysimeter cells. A second trench (13.9m x 0.5m x 0.3m deep) was excavated parallel to the first to hold the leachate collection vessels.

7.3.4 Lysimeter Cells

Each lysimeter cell was formed from four square, stackable inspection chamber sections. The internal dimensions of each section were 1m x 1m x 0.15m high. The base section of each lysimeter was manufacture with a 70mm diameter hole through the centre of one side of the section to enable the leachate collection pipe to pass from the inside to the outside of the lysimeter cell and into the leachate collection vessel. The 9 base sections (and an additional plain section to form a storage area) were located onto the construction platform and levelled into the sand layer. A leachate collection pipe was positioned across each base section, through the hole in the side of the section and into the leachate collection vessel. The leachate collection pipe was manufactured from a single length of 68mm diameter plastic piping. The pipe was cut to form a gutter when inside the lysimeter cell but remaining as a complete pipe when outside the section. The pipes were set with a cross-fall of approximately 1 in 7, to allow the leachate to drain from the lysimeter cell to the collection vessel.

To hold the base sections in position and to provide a hard-standing for working around the cells, the area around the base sections was overlaid with pre-mixed concrete (approximately 70mm thick). In the base of each lysimeter a sloping layer of concrete was placed to channel the percolating leachate into the collection pipe. The leachate collection pipe drained into the leachate collection vessels which were manufactured from high density polyethylene containers, with lids, and were recessed into the ground.

7.3.5 Lysimeter Lining and Drainage

The lysimeters were waterproofed by lining with heavy duty polythene sheeting. The polythene sheeting was overlaid with 6mm closed-cell polyethylene foam that was designed to take on the shape of the aggregate material sample as it was compacted inside the cell and thus reduce the peripheral flow down the sides of the cell.

A drainage layer comprising 40mm and 20mm gravel was constructed in the base of each lysimeter cell. The gravel was covered with a sheet of geotextile (Terram 500) to reduce the migration of fines into the drainage layer from the material sample compacted on top.

7.3.6 Placement of Materials in Lysimeters

The test materials and mixtures were compacted in 7 layers of 50mm, to produce the overall thickness of 350mm. The optimum moisture content and maximum dry density of each material and mixture was determined in the laboratory and used to calculate the mass of aggregate, binder and water required for each mix. The materials were mixed in a standard cement mixer, placed into the lysimeter cell and compacted in layers using three passes of a Kango vibrating hammer with a 150 x 150mm square foot. The thickness of each layer was determined by measurement and used to ensure that a good degree of compaction was achieved. The surface of each layer was scarified before placement of the next layer to improve the adhesion between the layers. A summary of the preparation of each material is given in Table 7-2. Each lysimeter was topped with a layer of 20mm river gravel to prevent spatter and control vegetation growth. The lysimeter field was opened to receive rainfall on 7 September 1999. The completed lysimeter field is shown in Figure 7-2.

Figure 7-2: Constructed lysimeter field

Table 7-2: Lysimeter sample preparation

Material or Mix	Total Material Mass (kg)	Optimum Moisture Content (%)	Maximum Dry Density (Mg/m^3)	Mixing Method
Aggregates				
BFS	BFS (100%) **TOTAL=698.5kg**	6	2.01	The BFS was added to the mixer and the water was added gradually.
CR (2-8mm)	CR (100%) **TOTAL=189.1kg**	-	0.6	The CR was not mixed. Compacted using the Kango hammer on top of a wooden board.
FSB	FSB (100%) **TOTAL=594.9kg**	10	1.8	Blocks (up to 0.5m) of FSB were broken up and blended with loose FSB in the mixer before adding the water.
LS	LS (100%) **TOTAL=776.8kg**	8	2.2	The LS was added to the mixer and the water was added gradually.
MSW(N)	MSW(N) (100%) **TOTAL=615.9kg**	12	1.62	The MSW(N) was added to the mixer and the water was added gradually.
Mixtures				
BFS (84%) gbfs (15%) li (1%)	BFS = 623.28 kg gbfs = 111.405 kg li = 7.288 kg **TOTAL=742.0kg**	7	2.08	The li was mixed with some gbfs then mixed with the remaining gbfs before adding the BFS and then gradually adding the water.
CCS (90%) ckd (7%) ce (3%)	CCS = 639.57 kg ckd = 49.742 kg ce = 21.322 kg **TOTAL=710.6kg**	9.3	2.03	The ce, ckd and some CCS were mixed before adding the rest of the CCS and then gradually adding the water.
LS (95%) ce (5%)	LS = 718.28 kg ce = 35.75 kg **TOTAL=754.0kg**	8	2.2	The ce was mixed with a small amount of LS before adding the rest of the LS and then gradually adding the water.
MSW (67%) filler (28.5%) bi (4.5%)	**TOTAL=701.3kg** MSW = 469.884 kg	As supplied	2.4	The hot mix was supplied by Darleston Coated Stone. The material was compacted as supplied

7.4 Data Collection

7.4.1 Leachate Sampling

The lysimeters were monitored and sampled for a period of approximately 1 year. The leachate collection vessels were usually emptied every 2 to 3 days with the volume of leachate being recorded and a running total of the cumulative L/S ratio calculated for each lysimeter. A sample of the leachate from each lysimeter was sent for analysis for the determinands listed in Table 3-9 on the following dates:

- 21 October 1999
- 1 November 1999
- 24 November 1999
- 19 January 2000
- 7 February 2000
- 28 February 2000
- 24 March 2000
- 10 April 2000
- 26 September 2000

Ideally, every sample of leachate would have been analysed, however, financial constraints limited the analysis to a series of spot samples. Patterns of cumulative release (mg/kg) against time and L/S ratio were calculated using the Rectangular Rule as described in Section 5.7 and as used in the analysis of the permeameter data.

7.4.2 Meteorological Data

The Sutton Bonington Site of the University of Nottingham has a meteorological station that reports directly to the UK Met Office. Data is recorded every minute and can be summarised into hourly or daily totals. The data from this station was used to monitor the weather conditions to which the lysimeters were exposed.

7.4.3 Rainwater Composition

To quantitatively determine whether the leachate sampling and storage procedure resulted in any cross-contamination of the leachate, a sample of rainwater was collected and analysed. The composition of the rainwater was used to quantitatively adjust the concentrations of the chemical species detected in the leachates from the lysimeters.

7.5 Results

7.5.1 Leachate Quantity

The quantity of leachate that passed through each lysimeter is summarised in Table 7-3. The table shows the cumulative quantity of leachate (litres) passed at the time when samples were collected for analysis. The table also presents the cumulative precipitation (litres/m^2) at the time of leachate sampling, which was calculated from hourly precipitation figures obtained from the Sutton Bonington Meteorological Station. In the table the precipitation figures are compared to the volume of leachate to determine the percentage of the precipitation that percolated through each lysimeter.

During the first year each lysimeter cell received approximately 572mm of precipitation with a total during the monitoring period of 641.4mm (7 September 1999 to 26 September 2000). The average annual rainfall recorded at the Sutton Bonington meteorological station between 1916 and 1998 was 607mm per annum and therefore the year of testing represented a drier than average year by approximately 5%.

Table 7-3: Cumulative quantity of leachate passing through each lysimeter compared to the cumulative rainfall

	Cumulative quantity of leachate passed through each lysimeter litres (% of the cumulative precipitation)									**Cumulative precipitation**
Date	**BFS**	**CR (2-8mm)**	**FSB**	**LS**	**MSW(N)**	**BFS+ gbfs+li**	**CCS+ ckd+ce**	**LS+ce**	**MSW+ bi+filler**	**litres/m^2**
21/10/99	81.0 (78.5%)	83.9 (81.3%)	64.1 (62.1%)	77.1 (74.7%)	89.9 (87.1%)	73.7 (71.4%)	96.2 (93.2%)	83.8 (81.2%)	73.0 (70.7%)	**103.2**
01/11/99	110.0 (85.5%)	107.6 (83.6%)	95.0 (73.9%)	106.0 (82.4%)	121.8 (94.7%)	97.4 (75.7%)	111.4 (86.6%)	111.7 (86.9%)	95.7 (74.4%)	**128.6**
24/11/99	128.2 (85.2%)	126.1 (83.8%)	113.5 (75.5%)	124.9 (83.0%)	139.7 (92.9%)	115.6 (76.9%)	118.4 (78.7%)	129.9 (86.4%)	114.5 (76.1%)	**150.4**
19/01/00	200.1 (89.2%)	185.6 (82.7%)	193.0 (86.0%)	195.5 (87.1%)	215.2 (95.9%)	186.3 (83.0%)	171.6 (76.5%)	201.1 (89.6%)	181.5 (80.9%)	**224.4**
07/02/00	203.6 (87.5%)	190.4 (81.8%)	198.5 (85.3%)	199.0 (85.5%)	218.9 (94.0%)	190.4 (81.8%)	175.2 (75.3%)	205.8 (88.4%)	185.3 (79.6%)	**232.8**
28/02/00	255.2 (90.2%)	244.0 (86.3%)	236.1 (83.5%)	247.2 (87.4%)	272.0 (96.2%)	237.7 (84.1%)	199.1 (70.4%)	259.2 (91.7%)	239.2 (84.6%)	**282.8**
24/03/00	272.1 (90.0%)	260.2 (86.0%)	257.0 (85.0%)	261.0 (86.3%)	287.7 (95.1%)	253.8 (83.9%)	214.2 (70.8%)	276.1 (91.3%)	255.6 (84.5%)	**302.4**
10/04/00	296.9 (89.1%)	287.2 (86.2%)	281.2 (84.4%)	286.4 (86.0%)	312.6 (93.8%)	278.7 (83.6%)	236.2 (70.9%)	301.9 (90.6%)	281.2 (84.4%)	**333.2**
26/09/00	539.9 (84.2%)	543.8 (84.8%)	528.5 (82.4%)	535.4 (83.5%)	575.6 (89.7%)	519.5 (81.0%)	421.6 (65.7%)	566.8 (88.4%)	551.2 (85.9%)	**641.4**

During the period of testing the quantity of leachate collected from each lysimeter was between 422 and 576 litres, as shown in Table 7-3. This volume represents 65.7% to 89.7% of the total precipitation, therefore runoff and evaporation losses and storage within the test materials accounted for between 34.3% and 10.3% of the total rainfall. The lowest percentage of precipitation passed as leachate was shown in lysimeter 1 containing the CCS+ckd+ce. Observations of this lysimeter in the field showed pooling of rainwater on the surface of the material in this cell. It is likely that this pooling resulted in increased evaporation losses when compared to the other cells.

The complete set of data for the percentage of precipitation that percolated through the lysimeters is shown in Figure 7-3, plotted against time. The figure shows that each lysimeter tends to exhibit very similar patterns of behaviour although the actual percentage passing varies. It is likely therefore that the actual percentage of rainfall passing is a function of the properties of the material, particularly the permeability, whereas the pattern of percentage of rainfall passing is a function of the rainfall. In Figure 7-3 two functions of the rainfall data are plotted alongside the percentage of rainfall passing. The two functions of rainfall are:

1. Rainfall since the previous sample - the sum of all rain fallen since the previous date when the leachate collection vessels were emptied;
2. Rainfall event intensity - the sum of all the rain fallen since the previous date when the leachate collection vessels were emptied divided by the number of hours in which rainfall occurred.

These functions of rainfall are plotted on Figure 7-3 as the negative of the calculated values. There is some evidence to suggest that the higher the rainfall event intensity, the lower the percentage of precipitation passing. This would be a reasonable conclusion because during high intensity rainfall water may run off (overflow) or pool on the surface of the lysimeters and be available for evaporation, whereas in lower intensity events the rain would have time to permeate directly into the lysimeter materials.

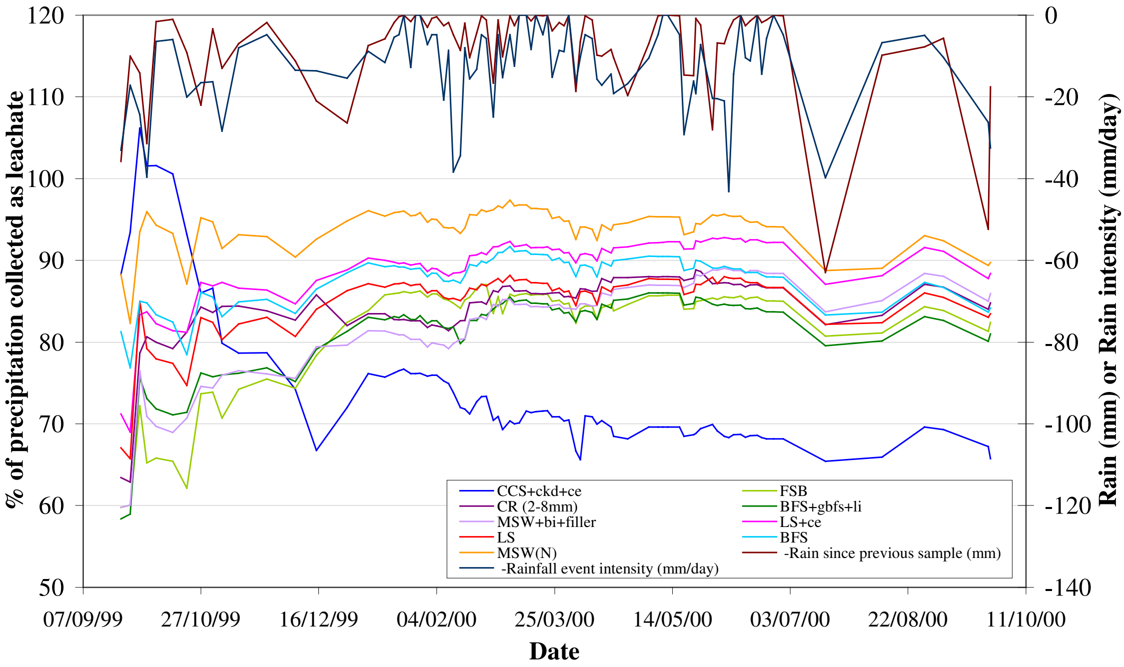


Figure 7-3: Percentage of precipitation collected as leachate plotted with functions of total rainfall and rainfall intensity, against time in the lysimeter tests

7.5.2 Leachate Quality

The composition of the rainwater sample is summarised in Table 7-4 where all detected chemical species are listed. Those species listed in Table 3-9 that are not listed in Table 7-4 may exist but at concentrations below the limits of analytical detection.

Table 7-4: Detectable determinants in the rainwater sample

Sample	pH	Cond. µS/cm	Concentration of detectable determinants (mg/l)				
			0.001-0.01	0.01-0.1	0.1-1	1-10	10-100
Rainwater	5.3	10.1	Sr	Zn	Ca Na Cl Total S Si	K SO_4 NO_3 TIC	TOC

The composition data given in Table 7-4 was examined to ascertain whether the detected species are typical for rainwater, or whether they have resulted from the sample collection or storage process. The data was compared to published rainfall composition data (US Geological Survey, 2000), blank test and distilled water data from the laboratory tests and the concentration of species in the lysimeter leachates.

The only corrections to be applied to the lysimeter leachate data are those that are deemed to originate from the sample collection and storage process. No correction was to be made for the actual chemistry of the rain since the lysimeters are designed to mimic real field conditions in which the rain, and associated chemistry, will form part of the leachant. Bearing this in mind, the lysimeter data was only corrected for zinc, which was present at similar concentrations in the analysis of the distilled water used in the laboratory tests and is likely to have originated from the cap of the Sterylin tubes used to store the leachate samples. The remaining species were generally either at concentrations within the range shown by the US Geological Survey (2000) and assumed to be ‘typical’, or were small (more than 1 order of magnitude less) when compared to the concentrations found in the lysimeter leachates. No correction was therefore applied for these species in the lysimeter leachates.

The concentration of species in the lysimeter leachates were quantitatively adjusted for the zinc, as described above. A summary of the lysimeter test data, giving the maximum and minimum concentrations measured for each chemical species in the leachate, is given in Table 7-5. Those species that are listed in Table 3-9 that are not listed in Table 7-5 may be present but are at concentrations below the limits of analytical detection.

From Table 7-5 it can be seen that the concentrations of different species in the leachates from the lysimeter tests vary over more than 6 orders of magnitude. A comparison of this data to that from the laboratory leaching tests indicates that in general, the species that are present in the leachates from the laboratory tests are also present in the lysimeter leachates for the individual materials. However, the actual species concentrations are often higher in the leachates from the lysimeters than in the laboratory tests and many of the lysimeter leachates contain addition species, particularly trace elements, which were not identified in the laboratory tests. The reason for these observations is most likely to be the relatively low liquid to solid ratio shown in the lysimeter tests. This low ratio means that small quantities of leached species (mg/kg) will result in higher concentrations in the leachates that will therefore be detectable.

Table 7-5: Summary of the lysimeter test data (maximum and minimum concentrations in the leachate)

Material Code	Max/ Min	pH	Cond. µS/cm	Concentration in leachate (mg/l)						
				<0.01	0.01-0.1	0.1-1	1-10	10-100	100-1000	>1000
BFS	Max.	9.0	3930		Ba Ni Cu Zn Mo V	NO_2 P Mn Fe Al	NO_3 Br F NH_4 Sr Li B	Mg Si	Ca Na K Cl S	SO_4
	Min.	7.8	2370	Zn	Ba Mn	Li	Mg Na Cl F Sr B	K Si	Ca S	SO_4
CR (2-8mm)	Max.	7.7	260	Li	Ba Fe Co Cu B	F Sr Mn	Mg Na K Br NO_2 P Si Zn	Ca Cl SO_4 NO_3 S NH_4		
	Min.	6.7	109		F Ba Sr Mn B	Br NO_2 P NH_4 Si Zn	Mg Na K Cl NO_3 S	Ca SO_4		
FSB	Max.	9.4	23500		Br Mn	NO_3 NO_2 Ba Sr Co Ni Zn Mo V Li	P NH_4 Fe Cu	Ca Mg Cl F S Si	SO_4	Na K
	Min.	7.8	610		Ba Sr Zn Li	P Fe	Ca Cl SO_4 S Cu	Na Si	K	
LS	Max.	8.3	490	Mn Cu Li	P Al Co Zn Mo V	Br Ba Sr Fe B	K NO_2 Si	Ca Mg Na Cl F S NH_4	SO_4 NO_3	
	Min.	7.8	190		NO_2 Ba Zn	Na Cl Si Sr	Mg K NO_3 F S	Ca SO_4		
MSW(N)	Max.	12.1	25300		Ba Mn Fe Cd V Li	Mg F P Sr Ni Zn Cr Pb B As	NO_3 NO_2 Si Cu Mo	Ca Br NH_4	S Al	Na K Cl SO_4
	Min.	9.0	1714	Sr	Ba Zn	Br NO_2 NH_4 Si Cu Mo	Ca Al	S	Na K Cl	SO_4
BFS+gbfs+li	Max.	12.9	12090		Mn Ni Cu Zn Mo	F P Al B	Br NO_2 NH_4 Si Ba Sr Fe Li	NO_3	Ca Na Cl	K SO_4 S
	Min.	11.8	740	Zn	Br Ba B	NO_2 NH_4 Si Li	Ca Na Cl NO_3 Sr	K SO_4	S	
CCS+ckd+ce	Max.	12.0	11160		Ba Sr Mn Cu Zn Mo	Mg Fe Ni Cr V Li B	Ca NO3 NO2 F P NH4 Al	Br Si	Na Cl SO4 S	K
	Min.	9.3	722	Sr	Fe Li	Br F P Al	Ca S Si	Na Cl SO4	K	
LS+ce	Max.	12.8	8890	Mn	Mg Br Fe Ni Cu Zn Cr Mo V	F P Al	Cl NO_3 NO_2 S NH_4 Si Ba Sr Li	SO_4	Ca Na K	
	Min.	9.3	1301	Zn	NO_3 Ba	NO_2 F Si Sr Al Li	Ca Na Cl SO_4 S	K		
MSW+bi+filler	Max.	8.8	859	Mn Zn Li	Ba Fe Cu Mo	Br NO_2 F P Sr Al B	Mg NH_4 Si	Ca Na K NO_3 S	Cl SO_4	
	Min.	7.6	220	Ba Mn	NO_2 Sr Al B	Mg Br F Si	K NO_3	Ca Na Cl SO_4 S		

The general patterns of leaching behaviour are illustrated by the pH and conductivity of the leachates, which are shown in Figure 7-4 and Figure 7-5. The figures show that both the pH and conductivity vary considerably between individual leachate samples. The conductivity data shows similar trends between materials, which is particularly evident for the leachates from 28 February 2000 which all show a significant reduction in conductivity compared to the leachates from the previous set of samples. This significant variation between consecutive samples was also observed by Fällman (1997) in her lysimeter leaching tests.

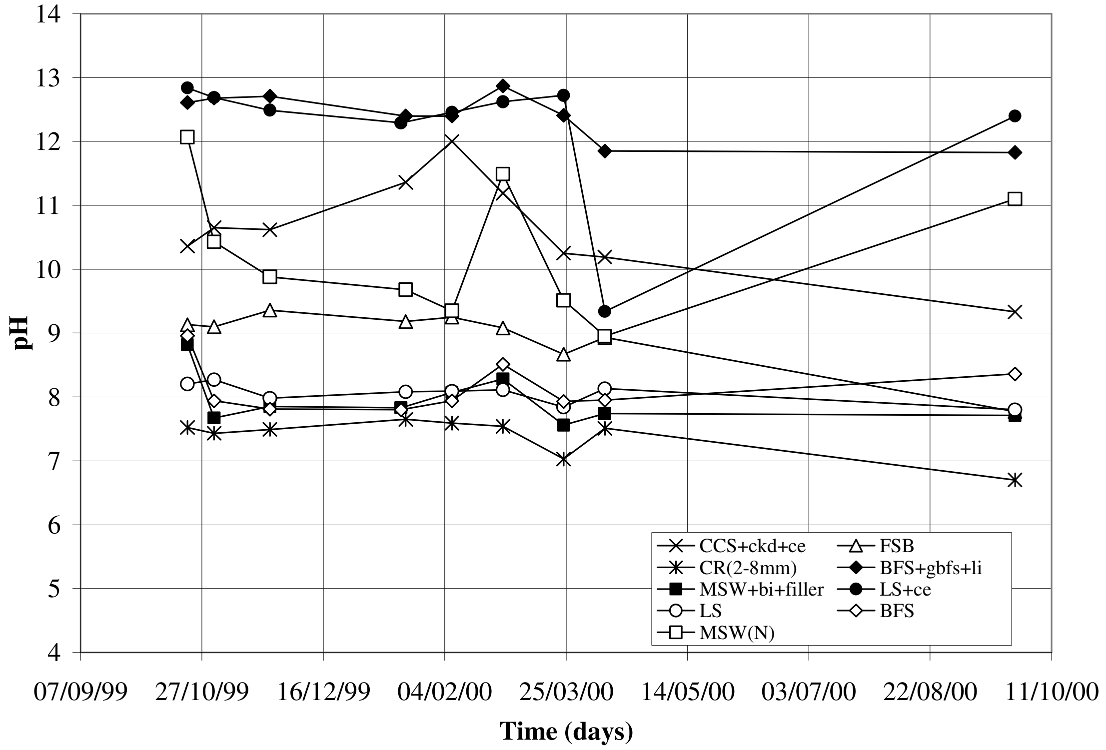


Figure 7-4: pH as a function of time for the lysimeter tests

The relationship between pH and conductivity in the lysimeters is uncertain. For some lysimeters pH and conductivity show similar patterns whereas in others the patterns appear to be the inverse of each other. In most lysimeters the relationship between the two parameters is inconsistent and may indicate different governing factors for the two parameters.

In general, the leaching of individual species fluctuates but shows some decline with an increase in time and L/S ratio. The fluctuation of species concentrations between consecutive samples is usually less than one order of magnitude. For many of the species the patterns of concentration in the leachate closely reflect the conductivity of that leachate. This correlation is particularly noted for the alkali and alkali earth metals and is illustrated in Figure 7-6 for the MSW+bi+filler lysimeter. A similar pattern is noted in many cases for the leaching of chlorine, sulphur, silicon and boron amongst others. This correlation is not shown by the BFS+gbfs+li and LS+ce lysimeters and for some individual species in a number of other lysimeters. However, in general, the species of the same chemical groups show similar patterns of leaching even when these are not the same as the conductivity.

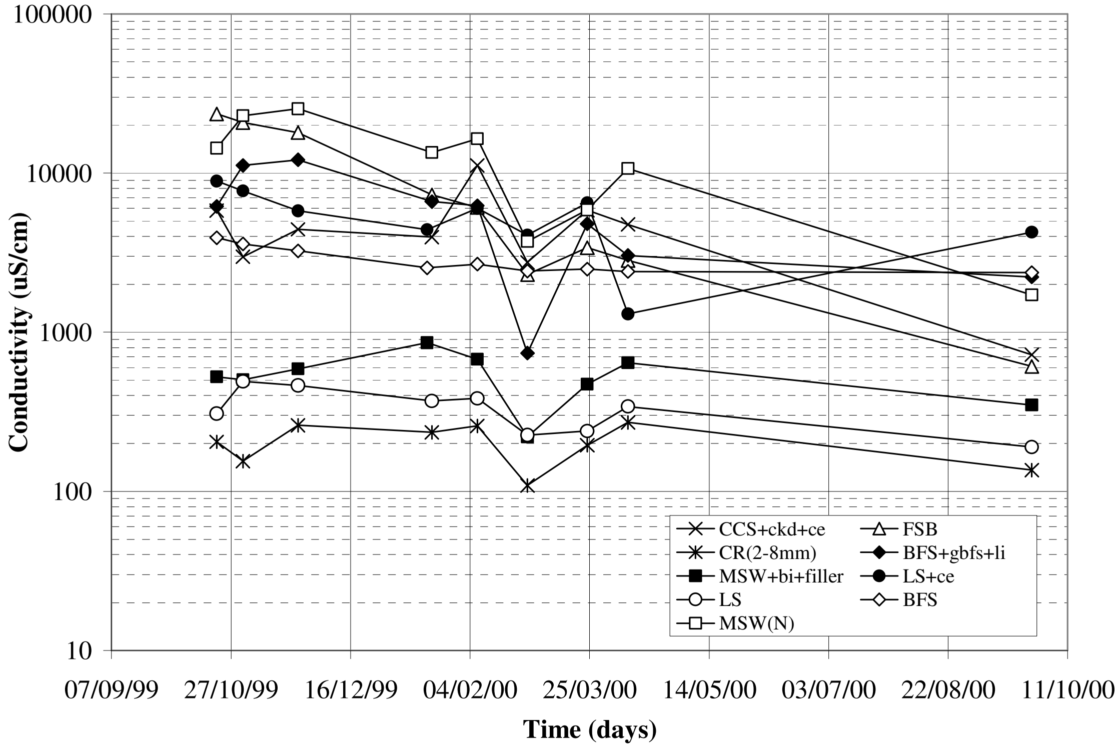


Figure 7-5: Conductivity as a function of time for the lysimeter tests

A correlation between the concentration of leached species and the leachate pH is not observed. However, the transition metals, for which the pH is likely to have most significant effect, are often below the limits of analytical detection and therefore the patterns of leaching are unknown. There is limited evidence that the leaching of aluminium from the materials in the lysimeters may be influenced by the pH of the leachate.

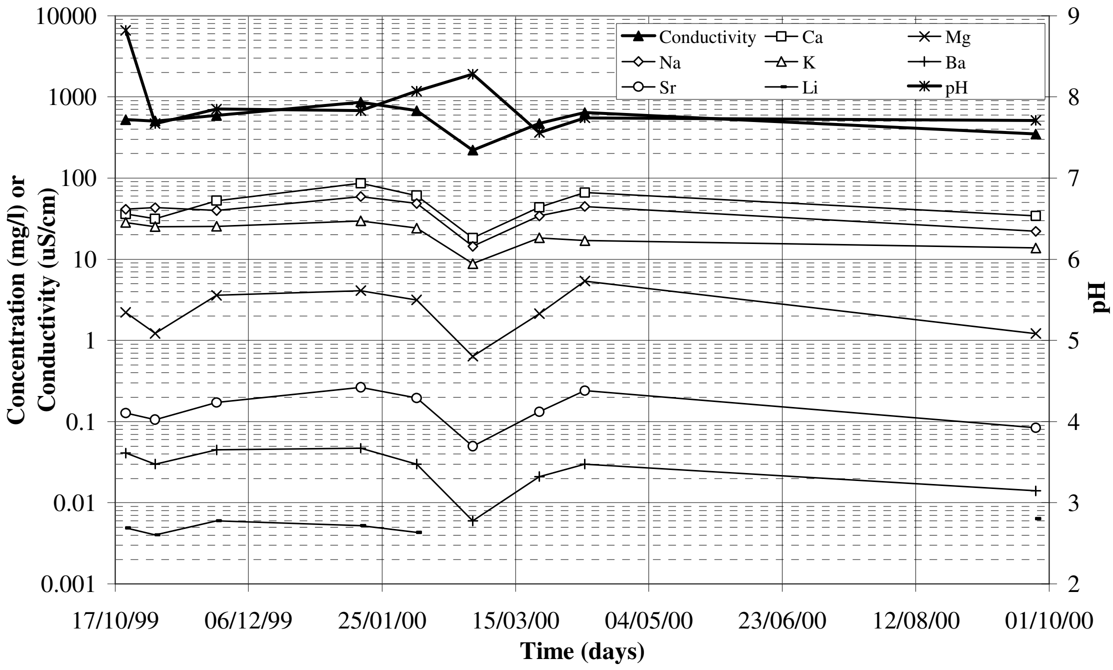


Figure 7-6: Concentration of alkali and alkali earth metals, conductivity and pH in the leachates from the MSW+bi+filler lysimeter as a function of time

The leachate quality data was examined for a relationship to a function of the rainfall. The data was normalised to different functions of the rainfall such as quantity, duration, intensity, event intensity and time since previous rainfall event. Although many of these functions show similar patterns to those of the species concentrations in the leachate, and therefore present some evidence that the patterns of leaching are dependent upon a function of the rainfall, it was not possible to determine the exact function, or combination of functions, that were the controlling factors. It is likely that a combination of effects is in operation.

The effect of the air temperature on the leaching process was also examined. Ilać et al. (2000), amongst others, observed that leaching is effected by temperature and that the rate of leaching is increased at higher temperatures. Figure 7-7 shows the maximum, minimum and mean air temperature during the period over which each sample was collected for analysis. Again, there is some evidence to suggest that temperature had an effect on leaching in the lysimeter tests. However, the likely combinations of effects contributing to the leaching outcome make it difficult to confirm the effect of temperature.

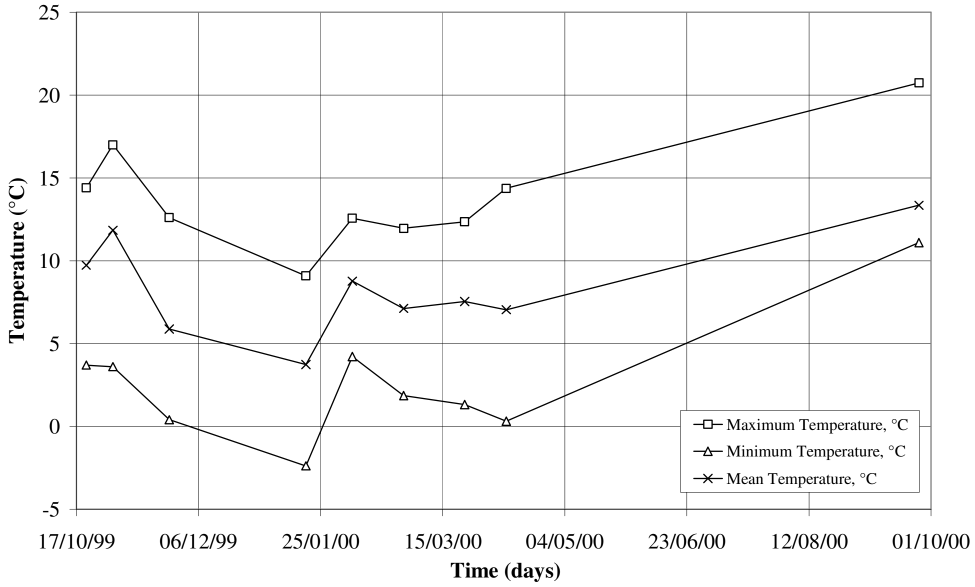


Figure 7-7: Average temperature during each sampling period plotted against time for the lysimeter tests

7.6 Discussion

7.6.1 Cumulative release (mg/kg)

The available information on the volume of leachate passed and the concentration of species in each analysed sample of leachate were used in the calculation of the cumulative leached quantity of each species (mg/kg) from each lysimeter at a given time or L/S ratio. The calculation was carried out using the Rectangular Rule as used on the permeameter test results and described in Figure 5-8 and Section 5.5.

In each case the cumulative release (mg/kg) of chemical species from the lysimeters increases with increasing time and L/S ratio, as expected. For the majority of chemical species the cumulative release (mg/kg) initially rises relatively steeply. With increasing time or L/S ratio the gradient of the line generally reduces and the cumulative release (mg/kg) approximately tends to an asymptote. This asymptote may be almost horizontal, indicating that all of the available chemical species has been leached from the material, or may have a shallow inclination that indicates that leaching is controlled by release rate, or diffusion. An example of the cumulative release (mg/kg) plotted against L/S ratio is given in Figure 7-8 for leaching of all chemical species from foundry sand (phenolic, brass).

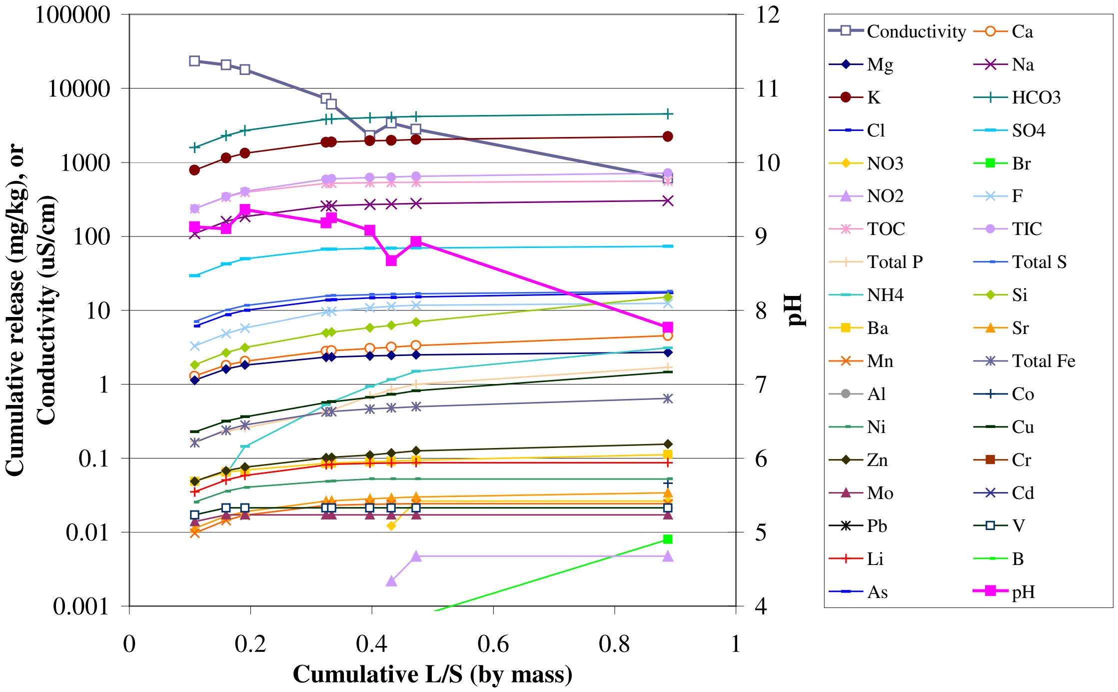


Figure 7-8: Cumulative release (mg/kg) of all chemical species from the FSB lysimeter plotted against cumulative L/S ratio

This cumulative release data will be used further in Chapter 8 of this thesis to enable a comparison to be made between the lysimeter tests and the laboratory leaching tests.

7.6.2 Normalisation of the Data from the Lysimeter Tests

To give an indication of the relative toxicity of the species in the leachates from the different materials in the lysimeter tests, the test data has again been normalised to the arbitrary water quality maxima, as listed in Table 3-13 and described in Section 3.4.6 and used in the interpretation of the laboratory leaching tests. The normalisation of the lysimeter data against such criteria is, again, used here purely as a tool for interpretation.

Table 7-6 presents the maximum and minimum normalised data for the lysimeter tests. It can be seen from Table 7-6 that all materials leached List II and other species in excess of the water quality maxima. The leachates from six materials contained List II species at between 10 and 100 times the water quality maxima. However, no materials leached List II substances in excess of 100 times the water quality maxima although potassium and/or aluminium were leached at this level for the foundry sand (phenolic, brass), MSW incinerator ash and CCS+ckd+ce.

Table 7-6: Normalisation of concentrations of chemical species in the leachates from the lysimeter tests to arbitrary water quality maxima

Material Code	Max/ Min	Maximum normalised concentration in leachate					
		<1	1-10		10-100		>100
			I & II	Other	II	Other	Other
BFS	Max	Mg Na Cl NO_3 P Ba Fe Ni Cu Zn Cr Mo Cd Pb Li As	F NH_4 V	Ca SO_4 NO_2 Mn Al B		K	
	Min	NO_2 NH_4 Mn Al V	F	Ca K SO_4 B			
CR (2-8mm)	Max	Ca Mg Na K Cl SO_4 F P Ba Fe Al Ni Cu Cr Mo Cd Pb V Li B As	Zn	NO_3 Mn	NH_4	NO_2	
	Min	NO_3 NH_4 Mn Zn		NO_2			
FSB	Max	Ca Mg Cl NO_3 Ba Al Cu Zn Cr Cd Pb Li B As	P Ni Mo V	Na SO_4 NO_2 Mn Fe	F NH_4		K
	Min	Na SO_4 NO_2 F P NH_4 Mn Ni Mo V		Fe		K	
LS	Max	Ca Na K Cl P Ba Mn Al Ni Cu Zn Cr Mo Cd Pb V Li B As	F	Mg SO_4 NO_3 Fe	NH_4	NO_2	
	Min	Mg SO_4 NO_3 NO_2 F NH_4 Fe					
MSW(N)	Max	Ca Mg NO_3 F P Ba Mn Fe Zn V Li B	Ni Cu Cr Cd As	SO_4	NH_4 Mo Pb	Na Cl NO_2	K Al
	Min	Cl SO_4 NH_4 Ni Cu Cr Cd Pb As	Mo	Na NO_2		K Al	
BFS+gbfs+li	Max	Mg NO_3 F P Mn Ni Cu Zn Cr Mo Cd Pb V Li B As	Ba	Ca Na Cl SO_4 Al	NH_4	K NO_2 Fe	
	Min	Ca Na Cl SO_4 NO_2 NH_4 Ba Fe Al		K			
CCS+ckd+ce	Max	Ca Mg NO_3 Ba Mn Fe Cu Zn Cd Pb Li B As	F P NH_4 Ni Cr Mo	Na Cl SO_4	V	NO_2 Al	K
	Min	Na Cl SO_4 NO_2 F P NH_4 Al Ni Cr Mo V				K	
LS+ce	Max	Mg Cl SO_4 NO_3 F P Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B As	NH_4 Ba	Ca Na Al		K NO_2	
	Min	Ca Na NH_4 Ba Al		K NO_2			
MSW+bi+filler	Max	Ca Mg Na Cl SO_4 NO_3 F P Ba Mn Fe Ni Cu Zn Cr Mo Cd Pb V Li B As	NH_4	K NO_2 Al			
	Min	K NO_2 NH_4 Al					

To give an indication of the overall 'toxicity' of the different leachates, the normalised species concentrations were summed for each leachate. The minimum possible concentrations (i.e. <0.1 taken as 0) were used in this calculation because the limits of analytical detection vary between materials and leachate samples, as discussed previously. Figure 7-9 shows the sum of all normalised concentrations and Figure 7-10 shows the sum of normalised List I and II species for each lysimeter material.

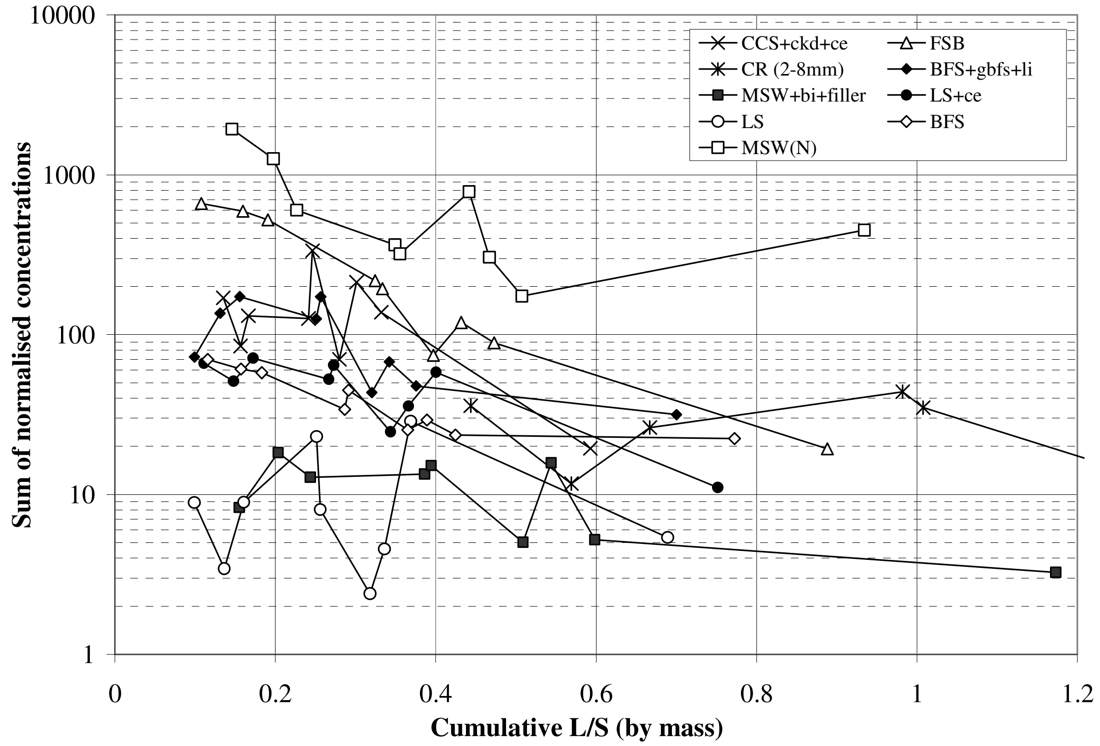


Figure 7-9: Sum of all normalised species concentrations from the lysimeter tests against cumulative liquid to solid ratio (minimum normalised concentrations)

The sum of normalised species concentrations generally shows an overall reduction with increasing L/S ratio and/or time. The rate of reduction is generally larger for materials exhibiting a higher sum of normalised concentrations, notably MSW incinerator ash and foundry sand (phenolic, brass). Limestone and MSW+bi+filler show relatively constant sum of normalised species, however the actual sum is low compared to other materials. Although these general trends appear to be present, there are also large fluctuations of up to 1 order of magnitude shown between consecutive samples in some of the lysimeters. The size of the fluctuations is generally larger for those materials with a smaller sum of normalised concentrations. These large fluctuations are likely to be the result of chemical species concentrations falling just above and then below the limits of analytical detection.

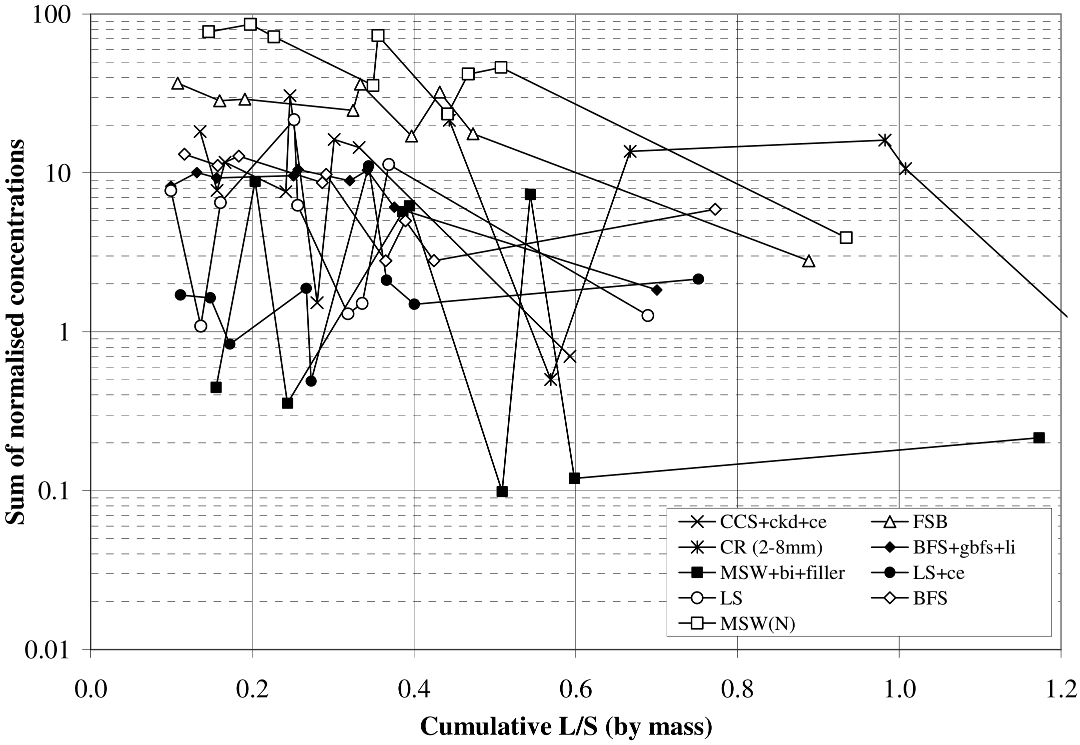


Figure 7-10: Sum of normalised concentrations of List I and II species from the lysimeter tests against cumulative liquid to solid ratio (minimum normalised concentrations)

For the majority of lysimeter materials the sum of normalised concentrations of List I and II species also show a general decline with increasing L/S ratio. However, LS+ce, MSW+bi+filler and limestone show more constant values for the sum of normalised List I and II species. These materials also exhibit the lowest values of theses normalised concentrations. The fluctuations between consecutive values of normalised List I and II species vary by up to 1.7 orders of magnitude. Again this is thought to result from the concentrations of chemical species moving from above to below the limits of analytical detection.

7.6.3 Effect of Binding on Leaching in the Lysimeters

To give an indication of the effect of material binding on species leachability the properties of the leachates from the sole materials and their mixtures have been compared. Figure 7-11 shows the pH and conductivity of the leachates from these tests. It can be seen that binding the blastfurnace slag with gbfs and lime increased the pH and the initial conductivity of the leachates. A similar effect was noted for the limestone when bound with cement. Binding the MSW incinerator ash (new incinerator) with bitumen had the opposite effect and significantly reduced both the pH and conductivity of the leachates throughout the test duration.

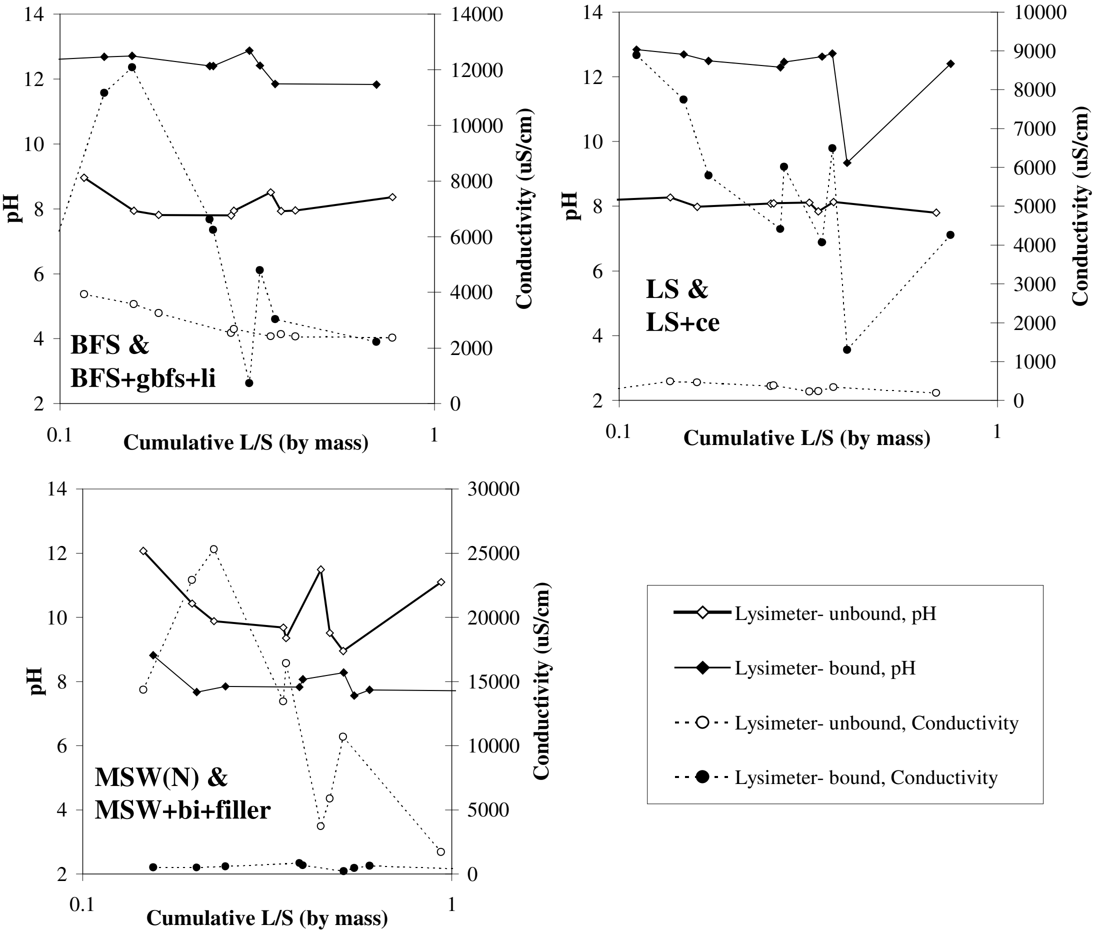


Figure 7-11: Comparison of the pH and conductivity of the leachates from the lysimeter tests on unbound and bound materials, plotted against cumulative L/S

The effects noted from the lysimeter tests on binding the blastfurnace slag and limestone are similar both in a generic and quantitative nature to that shown in the tank-leaching tests on these materials and mixtures. The reduction in conductivity from binding the MSW incinerator ash was also noted in the tank-leaching tests, although the pH remained unchanged on binding.

Figure 7-12 compares the sum of the total normalised species concentrations and the sum of the List I and II normalised species concentrations for the unbound and bound materials in the lysimeter tests. The graphs represent the minimum concentrations (i.e. <0.1 taken as 0) in the calculations because of the varying limits of analytical detection.

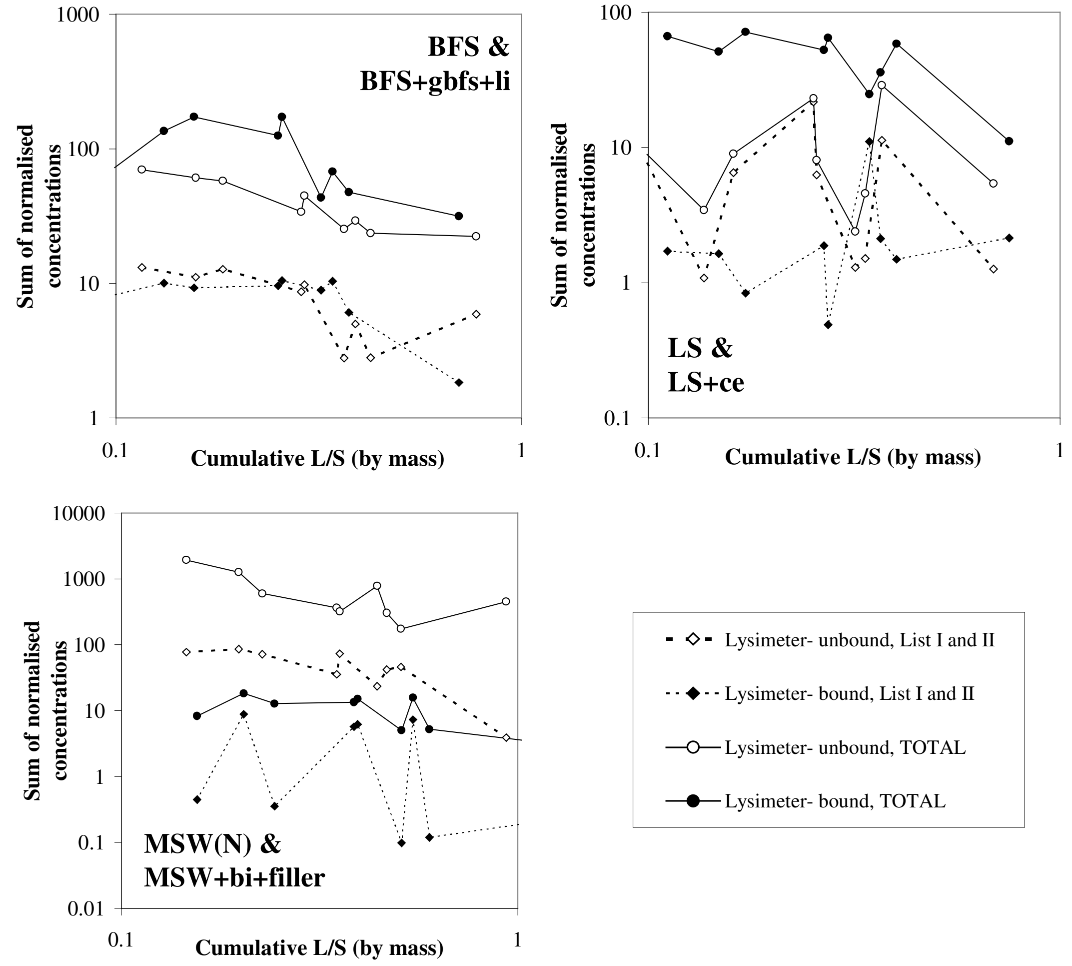


Figure 7-12: Comparison of the sum of normalised species concentrations in the leachates from the lysimeter tests on unbound and bound materials, plotted against cumulative L/S ratio

From Figure 7-12 it can be observed that binding the blastfurnace slag with granulated blastfurnace slag and lime increased the total normalised concentration but had little effect on the sum of List I and II species. The addition of cement to the limestone again increased the total normalised concentrations but slightly reduced the List I and II species in the leachate, whilst binding the MSW incinerator ash with bitumen significantly reduced both the total and List I and II normalised species concentrations in the leachate. The relationship shown between the blastfurnace slag and the BFS+gbfs+li mix is similar to that observed in the tank-leaching tests on these materials. The tank-leaching tests on the compacted limestone and the LS+ce mix showed an increase in the total normalised concentrations but also in the List I and II species. The reduction in both total and List I and II normalised species concentrations shown for the MSW incinerator ash when bound with bitumen in the lysimeters was also shown in the tank-leaching tests on these materials, although in these laboratory tests this reduction was less significant.

A detailed examination of the effect of binding on leaching was carried out by comparing the cumulative release (mg/kg) of each chemical species from the bound and unbound lysimeters for the same periods in time. The assessment uses the same methodology as that applied in the assessment of the effect of binding in the tank-leaching tests and permeameters discussed in Chapter 6. The results of the assessment of the lysimeter data are given in Table 7-7. Significant reductions in leached quantity as a consequence of binding are noted for many chemical species in all of the mixtures tested. Increases in conductivity and the leached quantity of the alkali and alkali metals (except magnesium) are shown in the mixtures incorporating cement or lime binders. The MSW+bi+filler material showed reductions or no increase in the leached quantity of the majority of chemical species.

7.7 Summary and Conclusions

- Large-scale lysimeter tests were designed and constructed to simulate the physical dimensions, material properties and environmental conditions of aggregate materials within a road construction or bulk fill.

- Nine materials and mixtures were tested in the large-scale lysimeters.

- The leachate collected from each lysimeter cell was quantitatively and qualitatively assessed for a period of over 1 year.

Table 7-7: Matrix summarising the effect on leached quantity of the addition of binders to aggregate materials, as observed in the lysimeters

Aggregate	**BFS**	**LS**	**MSW**
Binder	**+gbfs+li,**	**+ce**	**+bi+filler**
pH	+	+	-
Conductivity	+/++	++/+++	---
Ca	-	++	++
Mg	-/--?	-----	+++
Na	+/++	+++	---
K	+	++++	---/----
HCO_3	+++	+++	---
Cl	+/++	-/--	---
SO_4	--	--	--
NO_3	+++	--	+++
Br	+	---	---
NO_2	+++	++/+++	--
F	---/----	--	++
TOC	+/++	++	---
TIC	+/++	+	--
Total P	--	+?	--
Total S	-	--	--
NH_4	+	--	---
Si	---	+	-
Ba	++	+++	=/+
Sr	+/++	++	=
Mn	----	+/++	-
Total Fe	-/+?	+/++	+
Al	--/---	++++	----
Co	X	-?	X
Ni	--/---	+?	-?
Cu	-	-/=/+?	----
Zn	--	+/++	-----
Cr	X	+?	-?
Mo	--	+/++	---/----
Cd	X	X	-?
Pb	X	X	-?
V	-?	+?	-?
Li	=/+	++++	=/-
B	---	-?	--
As	X	X	-?
Se	X	X	X

Key:

Symbol	+++++	++++	+++	++	+	=	-	--	---	----	-----
Cumulative release (mg/kg) from: bound material / unbound material	>1000	1000 to 100	100 to 10	10 to 2	2 to 1.25	1.25 to 0.8	0.8 to 0.5	0.5 to 0.1	0.1 to 0.01	0.01 to 0.001	<0.001

X	Effect of binder cannot be defined except that it does not cause a large increase in leaching
- ?	Binding results in a possible decrease in leaching
+ ?	Binding results in a possible increase in leaching
/	Denotes borderline values /
=	Denotes no change

- Each lysimeter received 641mm of precipitation during the period of assessment. The quantity of leachate collected from each lysimeter represented between 65.7 and 88.4% of this precipitation. The remainder was lost to evaporation and runoff.

- The absolute concentrations of chemical species within the leachates from the lysimeter tests were generally higher than those observed in the laboratory tests. This is probably due to the lower L/S ratios in the lysimeters resulting in less dilution of the leached species.

- The higher concentrations observed in the lysimeter tests enabled many chemical species that were below limits of analytical detection in the laboratory tests to be detected and quantified.

- The concentrations of chemical species in leachates often form the basis of an assessment of environmental acceptance. Thus the higher concentrations observed at lower L/S ratios in the lysimeters could have a significant impact on what is deemed to be acceptable data for use in assessments. The use of flux rate, which considers the volume of leachate as well as the concentration of the chemical species, is likely to be more appropriate than simply concentrations.

- The concentrations of chemical species in the leachates from the lysimeters generally reduced with increasing time and L/S ratio. It can thus be concluded that the source of chemical species from aggregate materials in road construction is likely to decline and that leaching will improve over time (i.e. lower concentrations).

- The patterns of leaching shown by different chemical species were often similar for a particular material, although the absolute concentrations differed.

- The effect of binding on leaching was determined for three mixtures by comparison to the observed leaching from the sole aggregate. In general, binding resulted in a decrease in the cumulative release (mg/kg) of many of the chemical species.

8 Relationship between Laboratory Tests and Large Scale Simulations

8.1 Introduction

Chapters 3, 4 and 5 presented the methodology and results from the programme of laboratory characterisation and leaching tests performed on a range of alternative aggregate materials. The tiered testing programme was designed to identify the effect of different physical conditions on the leaching process. The various tests involved agitated leaching, unagitated leaching, size reduced material, material at realistic grading and material compaction and examined the processes of diffusive and advective leaching. Each chapter described the test methods being applied and the data that was attained.

Chapter 6 applied the same testing regime to a selection of alternative aggregate materials stabilised with a variety of binder modifications and reviewed the leaching behaviour in comparison to that of the unbound material. Chapter 7 described and discussed the lysimeter trials that were large-scale field simulations designed to be indicative of the likely in-situ leaching environment of a road construction or bulk fill.

In this chapter the relationship between the large-scale lysimeter trials and the more fundamental laboratory scale leaching tests is discussed

8.1.1 Need for Interpretation

The purpose of performing the lysimeter trials was to examine how the alternative aggregate materials leach in the uncontrolled conditions of the real environment. The trials were successful in a research environment and provided valuable data on the leaching properties of the materials that were tested. It would however not be practical for industry to perform a lysimeter trial on every new type of material, mixture or each supplier or as a test to confirm the leaching properties of a particular batch of material. The lysimeter trials are too time consuming for use in such circumstances.

The laboratory scale tests are a much more appropriate tool for use by industry as a means of determining material leachability. However, it is necessary to be able to use a laboratory test to determine how a material will perform in the real situation. This chapter aims to compare the leaching patterns observed in the laboratory tests with those shown in the lysimeter tests. It also aims to determine whether in-situ leaching can be estimated from laboratory leaching tests, which tests provide the most simulative data and how that data should be used and interpreted.

This discussion is divided into two main sections:

1. The data from all leaching tests and lysimeters are used to describe similar patterns of leaching;
2. An empirical relationship based on all test data is proposed for predicting cumulative release (mg/kg) and concentrations (mg/l) of chemical species in the leachate for any L/S ratio.

8.1.2 Basis of Interpretation

A series of plots have been used as the basis for the interpretation of the leaching test data and comparison of the lysimeters with the laboratory tests. Each plot contains the data from all tests performed on a particular sole material or mix and each type of plot was completed for each chemical species determined in the leachate.

The plots that were used to examine the data from all the laboratory and field leaching tests are as follows:

- pH verses leached species concentration (mg/l);
- L/S ratio verses leached species concentration (mg/l), pH and conductivity;
- L/S ratio verses cumulative release (mg/kg) of leached species.

The plots for each chemical species from each of the 9 material tested in the lysimeters are contained in Appendix C, D and E respectively.

It is important to note that there are two different types of tests being considered:

- batch leaching tests (rapid leaching characterisation test, tank-leaching tests), and
- flow-through leaching tests (permeameters, lysimeters).

In the batch tests all leached species are contained within the whole sample of test leachate. In the flow-through tests, however, the leachate is continually renewed and the leached species are contained within the leachate that is within the sample at a given time. In theory, if two tests produce comparable leaching performance, the concentration of chemical species in the leachate of a batch leaching test at a given time or L/S will not equal those obtained from a flow-through test at the same time or L/S. Instead the concentrations in the batch test should equal the average concentration of chemical species in the leachate of a flow-through test for samples generated between a time or L/S ratio of zero and the time or L/S ratio of the batch leaching test. Interpreting this theory for the cumulative or total leached quantity of a chemical species suggests that after a specific time or at a given L/S the quantity leached in a batch leaching test will be equal to that leached in the flow-through test. If these patterns are not observed, then the rate and/or the quantity of leaching vary between the different test methods.

8.2 Observations of Relationships between Laboratory and Field Conditions

8.2.1 Leached Species verses pH

Figure 8-1 shows the pH of the leachates against L/S ratio for all tests on each of the 9 materials and mixtures that were examined in the lysimeter. It can be seen from Figure 8-1 that, for most materials, the pH of the leachates generated from the different tests varies by between 1 and 3 units of pH, thus indicating a reasonable degree of consistency. The exceptions to this are:

1. MSW incinerator ash that shows variations of up to 5 pH units between tests and a large range of pH in individual tests; and
2. Foundry sand (phenolic, brass) that shows a range of 4 pH units between tests but generally shows little variation within a single test.

The plots of concentrations (mg/l) of chemical species in the leachates against pH are given in Appendix C for each of the materials included in the lysimeter tests. Observations of these plots show little evidence for pH controlled leaching. This may be for a number of reasons:

1. The pH does not vary significantly between tests and thus solubility limits are relatively constant in the different tests;

2. The chemical species with solubility limits that are most susceptible to changes in pH are often at concentrations (mg/l) below the limits of analytical detection and thus, cannot be fully assessed.

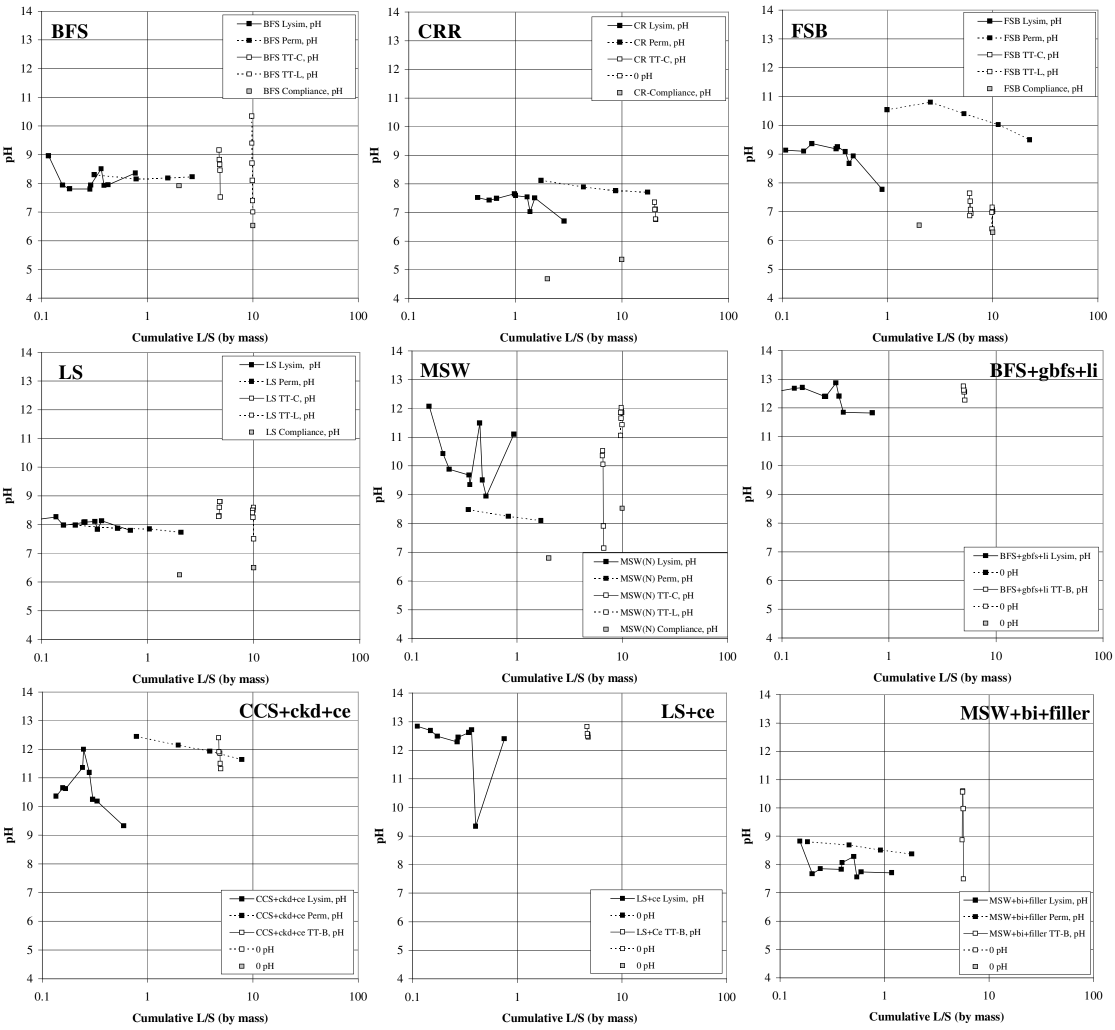


Figure 8-1: pH verses L/S ratio for all laboratory and lysimeter tests

8.2.2 Leached Species verses L/S Ratio

The leached species have been considered both in terms of concentrations (mg/l) and cumulative release (mg/kg) verses L/S ratio. The results of all tests have been plotted for each chemical species for each of the 9 materials tested in the lysimeters. The plots are given in Appendix D and E respectively.

A detailed description of the different patterns of leaching shown by the individual chemical species in the leachates from all tests for the 9 materials tested in the lysimeters is given in Appendix F. A number of the key patterns and observations are summarised below.

The graphs of leached species concentrations (mg/l) against L/S ratio show 3 main patterns of leaching. These patterns are illustrated in Figure 8-2, Figure 8-3 and Figure 8-4 described below.

Figure 8-2 shows the concentrations of the alkali metals sodium, potassium and lithium in the leachates from the tests on foundry sand (phenolic, brass). This figure illustrates the pattern of rapid decline in species concentrations with increasing L/S ratio. The lysimeter and permeameter results show a continued decline in concentrations with increasing L/S ratio and the batch tests (TT-L, TT-C and the rapid leaching characterisation test) show an approximate average of the concentrations of all L/S ratios between zero and the L/S ratio of the particular test.

The rapid decline in leached species concentrations with increasing L/S ratio is likely to indicate initial rapid leaching via surface wash-off, followed by release rate dependant leaching.

Figure 8-3 shows the concentrations of the Group III and IV chemical species boron, aluminium, silicon and lead in the leachates from the tests on MSW+bi+filler. This figure illustrates a pattern of relatively constant species concentrations with increasing L/S ratio. In general, the leachates from all tests show similar concentrations of the same chemical species. This pattern may imply that leaching is controlled by solubility constraints or by the rate at which species can be leached from the solid matrix.

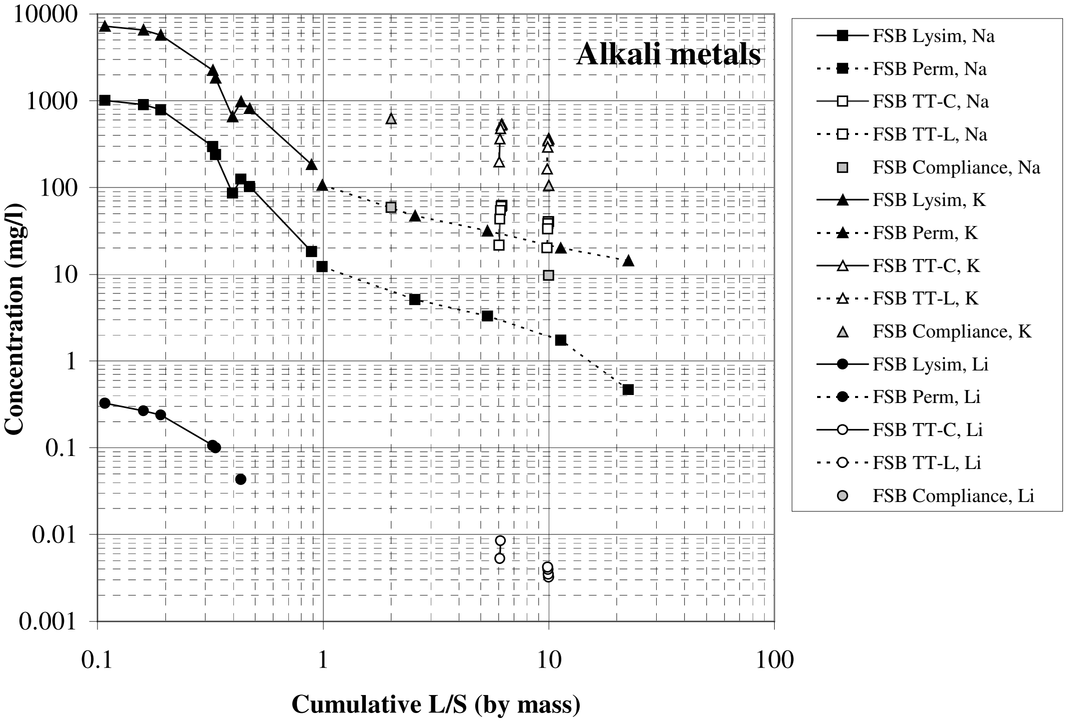


Figure 8-2: Concentrations of alkali metals from foundry sand (phenolic, brass)

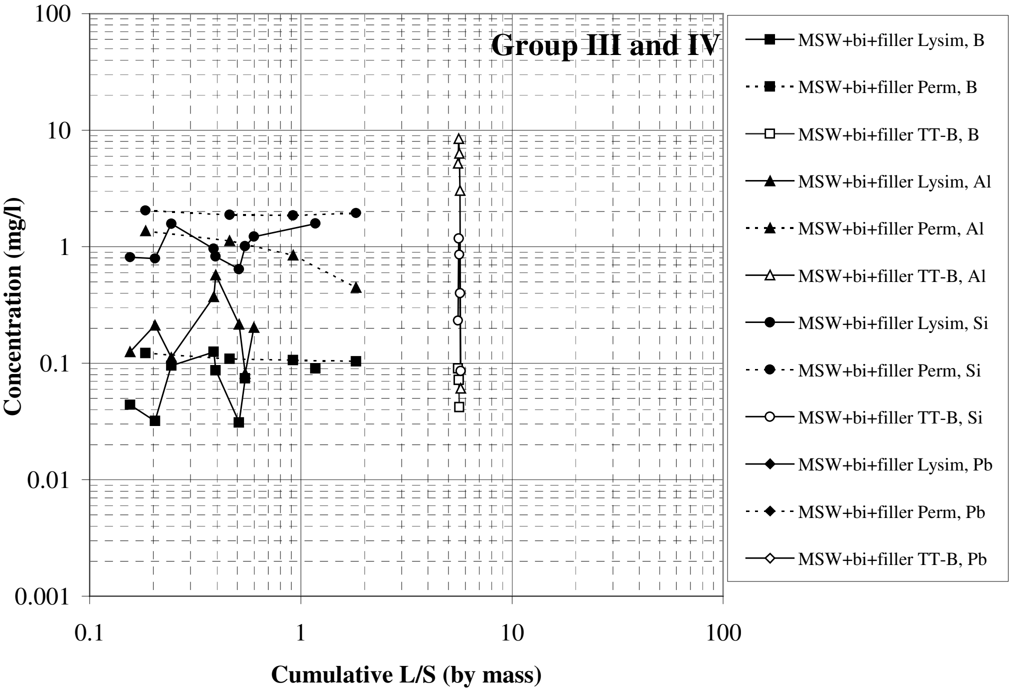


Figure 8-3: Concentrations of Group III and IV species from MSW+bi+filler

Figure 8-4 shows the concentrations of alkali earth metals calcium, magnesium, barium and strontium in the leachates from the tests on blastfurnace slag. This figure illustrates a pattern of leaching with a gentle decline in leached species concentrations with increasing L/S ratio. The concentrations observed in the batch leaching tests are usually similar or slightly higher than those shown in the flow-through tests at the same L/S ratio. This trend implies a range of mechanisms are controlling the leaching process.

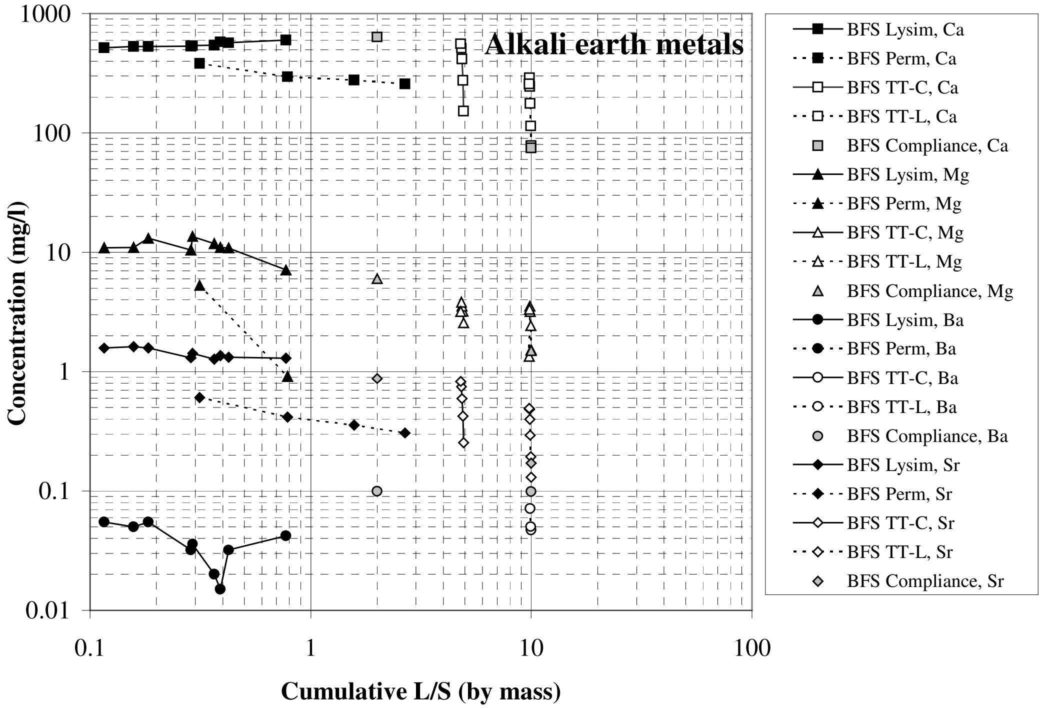


Figure 8-4: Concentrations of alkali earth metals from blastfurnace slag

In generally, many of the chemical species from all materials exhibit leaching patterns in the range between the two extremes of behaviour illustrated in Figure 8-2 and Figure 8-3. As such, it is likely that a range of processes operate to affect the leaching performance. There is also considerable scatter in the data sets as a whole, with different tests showing varying relationships to each other. It was thus not considered possible to categorise the leaching of individual chemical species from each material into specific groups.

Observations of the cumulative release (mg/kg) of chemical species against L/S ratio again show varying patterns. Figure 8-5 shows the relationship between the cumulative release (mg/kg) of the alkali metals sodium, potassium and lithium against L/S ratio for tests on CCS+ckd+ce. This figure shows excellent correlation between L/S ratio and cumulative release (mg/kg) with all tests showing similar values at the same L/S ratio.

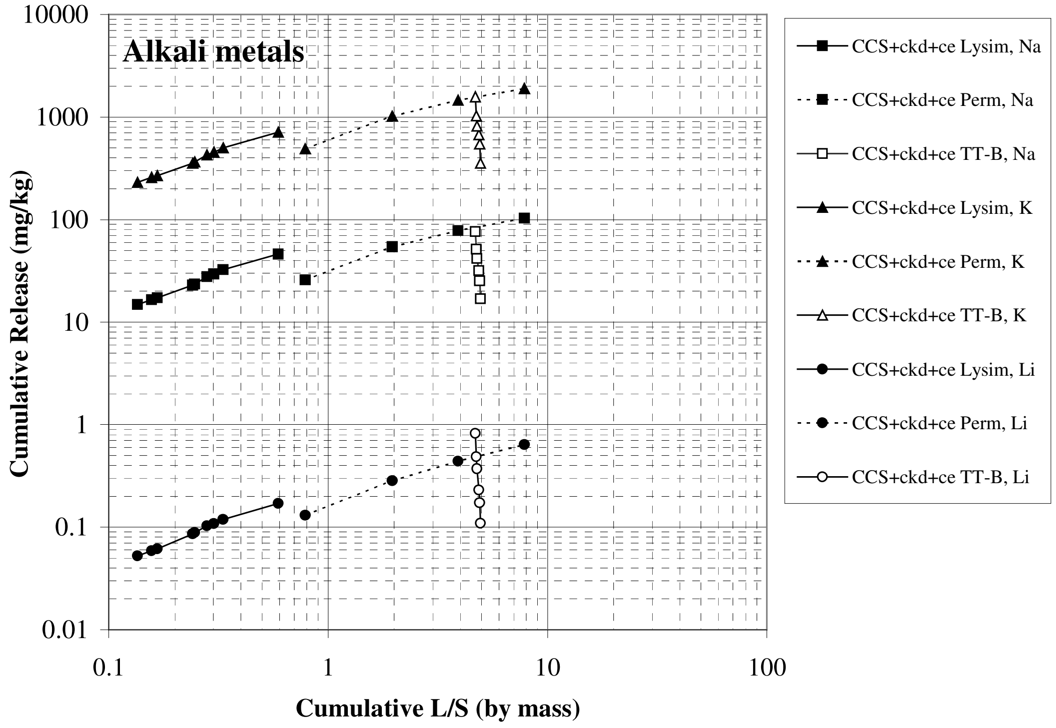


Figure 8-5: Cumulative release (mg/kg) of alkali metals from CCS+ckd+ce

However, a more common pattern is illustrated in Figure 8-6, which shows the cumulative release (mg/kg) of the Group VII species fluorine, chlorine and bromine against L/S ratio from the MSW incinerator ash. Here it can be seen that the lysimeter, tank-leaching test and rapid leaching characterisation test results show similar cumulative release (mg/kg) at the same L/S ratio. However, the permeameter tests appear to significantly under predict the observed cumulative release (mg/kg). This may be due to a number of reasons, including:

1. The short residence time of the leachate within the material sample during the permeameter test, and therefore a reduction in leaching by diffusion;
2. Errors in the method of calculation of the cumulative release (mg/kg) from this test data.

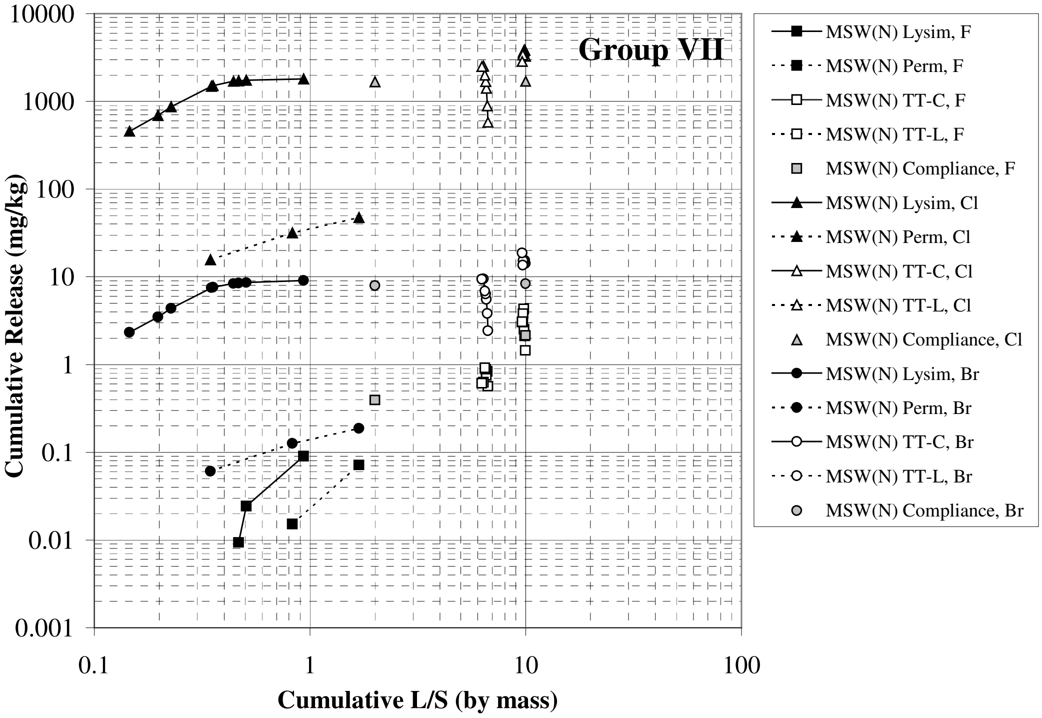


Figure 8-6: Cumulative release (mg/kg) of Group VII species from MSW incinerator ash

From the data presented here, it is thus concluded that in general the concentrations (mg/l) of chemical species in the leachate and the cumulative release (mg/kg) shown by the tank-leaching tests and rapid leaching characterisation test is largely illustrative (same order of magnitude) of the performance of the lysimeter tests at a given L/S ratio.

8.3 Empirical Relationship between Laboratory and Field Conditions

The relationships discussed in Section 8.2 describe observations of the patterns shown when the results of all laboratory tests and the large scale simulations are plotted together for an individual chemical species leaching from a particular material. Such observations are useful for identifying the potential mechanisms controlling the leaching process.

To enable a user to perform a laboratory leaching test and use the result to determine the likely cumulative release (mg/kg) and concentrations (mg/l) in the leachate at any L/S ratio, requires further consideration of the data. This section provides a methodology for obtaining a trend line that can be used to find a rough approximation of these parameters, for any L/S ratio.

8.3.1 Background

The data obtained directly from the laboratory leaching tests and lysimeters is reported as concentrations (mg/l) of chemical species in the leachate. Data reported in this way cannot be directly compared for all types of test. The differing hydraulic regimes shown in the individual tests mean that the flow-through tests (lysimeters and permeameters) show concentrations in the leachate at increasing L/S ratios whereas the batch leaching tests (rapid leaching characterisation test and all tank-leaching tests) show an ‘average’ concentration for all L/S ratios between zero and the maximum L/S ratio of the test.

The concentrations (mg/l) of chemical species in the leachate and the L/S ratio of the tests can be used to calculate the cumulative leached quantity or cumulative release (mg/kg) at a given L/S ratio, as used in previous chapters. The plot of cumulative release (mg/kg) against L/S ratio generally shows an increase in release with increasing L/S ratio. Values for cumulative release (mg/kg) can be compared directly for all the laboratory leaching tests and lysimeters. As such, this data was considered to be the most appropriate for use in identifying an empirical relationship between the laboratory leaching tests and lysimeters.

For a particular chemical species leaching from a specific material, the cumulative release (mg/kg) generally increases with L/S ratio in all tests to produce similar values at given L/S ratios. However, the actual values of cumulative release (mg/kg) shown by different chemical species differ. These different values for cumulative release (mg/kg) make it difficult to describe all relationships by a single trend.

To enable the leaching of different chemical species from all materials to be compared and a general trend to be defined, the data for all chemical species from all materials was shifted along the Y-axis relative to a defined point.

8.3.2 Application of the Shift Factor (F_s)

Each chemical species leached from all tests on a particular material were shifted along the Y-axis in a way that maintained the observed pattern, on the log-log plot, of L/S ratio against cumulative release (mg/kg). This was carried out by shifting all the data, for a particular chemical species, along the cumulative release axis by a logarithmic factor: *The Shift Factor (F_s).*

For each set of data, the cumulative release (mg/kg) shown by the rapid leaching characterisation test at L/S = 10 was used as the data point at which to quantify the logarithmic Shift Factor to normalise each data set. This data point was chosen since it was easy to define and the value shown at this point was generally fairly consistent with that shown by the tank-leaching tests and lysimeters. The main limitation of using this data point for the normalisation is that only unbound materials can be used in the assessment, since bound mixtures were not assessed in the rapid leaching characterisation test.

The Shift Factor (F_s) is the factor by which the data from all tests for a particular chemical species and material is shifted by in order to carry out the normalisation along the Y-axis. The value of the Shift Factor (F_s) is different for each chemical species and for each material. The Shift Factor (F_s) is determined from the location of the cumulative release (mg/kg) in the rapid leaching characterisation test at L/S=10 relative to the Point of Normalisation, arbitrarily chosen to be a cumulative release (mg/kg) of 1. The determination and use of the Shift Factor is illustrated in Figure 8-7. The value of the Shift Factor (F_s) is determined using Equation 8-1.

$$F_s = \log Y_d \qquad \textbf{Equation 8-1}$$

Where,

F_s = Shift Factor

Y_d = cumulative release (mg/kg) for particular chemical species in rapid leaching characterisation test at L/S=10

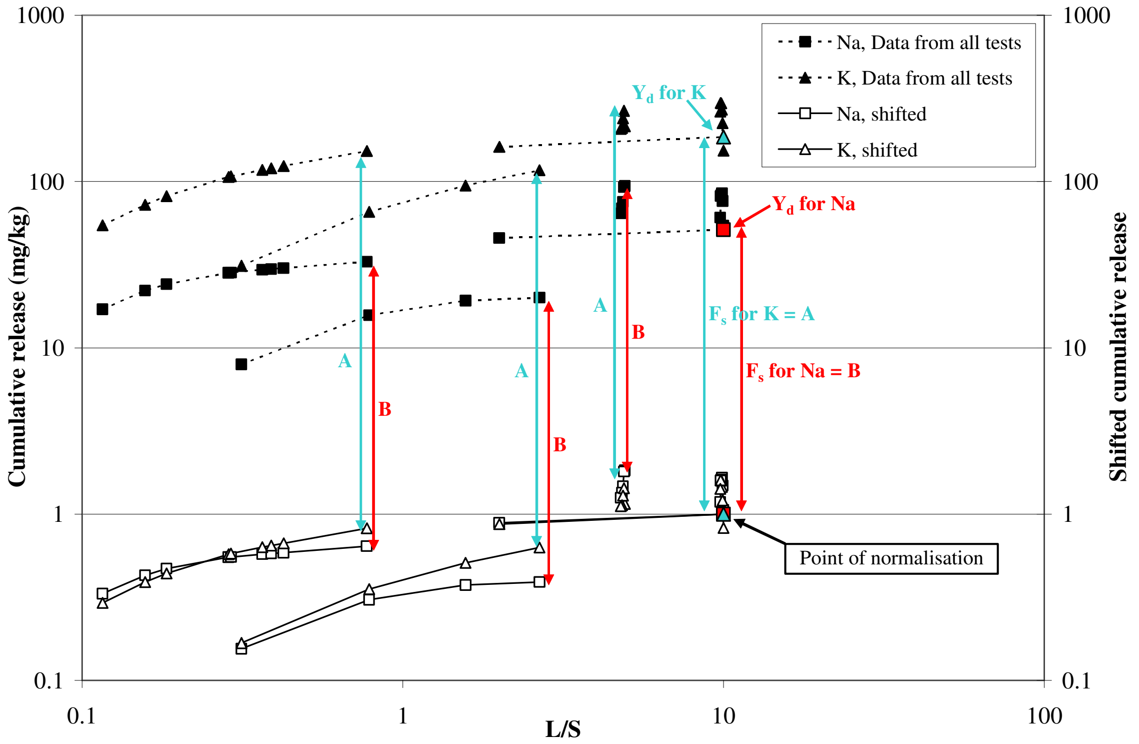


Figure 8-7: Normalisation of data using the Shift Factor, F_s – Example showing leaching of sodium (Na) and potassium (K) from blastfurnace slag

The Shift Factor (F_s) was used to normalise the cumulative release (mg/kg) of a particular chemical species, shown at each data point for all tests carried out on a specific material. The Shift Factor was applied to each data point by using Equation 8-2.

$$\log Y_s = \log Y_T - \log Y_d$$ **Equation 8-2**

Where,

Y_s = shifted cumulative release

Y_T = cumulative release (mg/kg) at a particular data point to be shifted

Y_d = cumulative release (mg/kg) in rapid leaching characterisation test at L/S=10

8.3.3 Fitting a Trend Line

The shifted data obtained by applying Equation 8-2 was plotted as log of L/S ratio on the x-axis against log Y_s on the Y-axis. Data for all chemical species from all materials was included on the plot. Following initial observations of the resulting chart, the dataset was considered to require a number of modifications, as follows:

- The large number of data points representing each tank-leaching test was causing a significant cluster of test results in the regions around the L/S ratios of these tests. To reduce the weighting of data in these areas each set of tank-leaching test data was reduced to a single data point with a value equal to the maximum normalised cumulative release observed for that test;
- The relatively low cumulative release (mg/kg) observed in the permeameter tests when compared to the other tests was considered to adversely affect the overall trend of the data. The lower release would reduce the average leached quantity observed and thus would not represent a conservative assessment of leachability. It was therefore decided that the permeameter test results would be excluded from the data set used when fitting the trend line.

The resulting dataset used to produce the trend line included:

- Rapid leaching characterisation test
    - Shifted cumulative release at L/S = 2
    - Shifted cumulative release at L/S = 10
- Tank-leaching test on loose materials (TT-L)
    - Shifted maximum observed cumulative release
- Tank-leaching test on compacted materials (TT-C)
    - Shifted maximum observed cumulative release
- Lysimeters
    - Shifted cumulative release for all data points

This dataset was plotted as log of L/S ratio on the x-axis against log Ys (where, Ys = shifted cumulative release) on the Y-axis. Regression analysis was carried out on the data using a computer package. The dataset and trend line are shown in Figure 8-8.

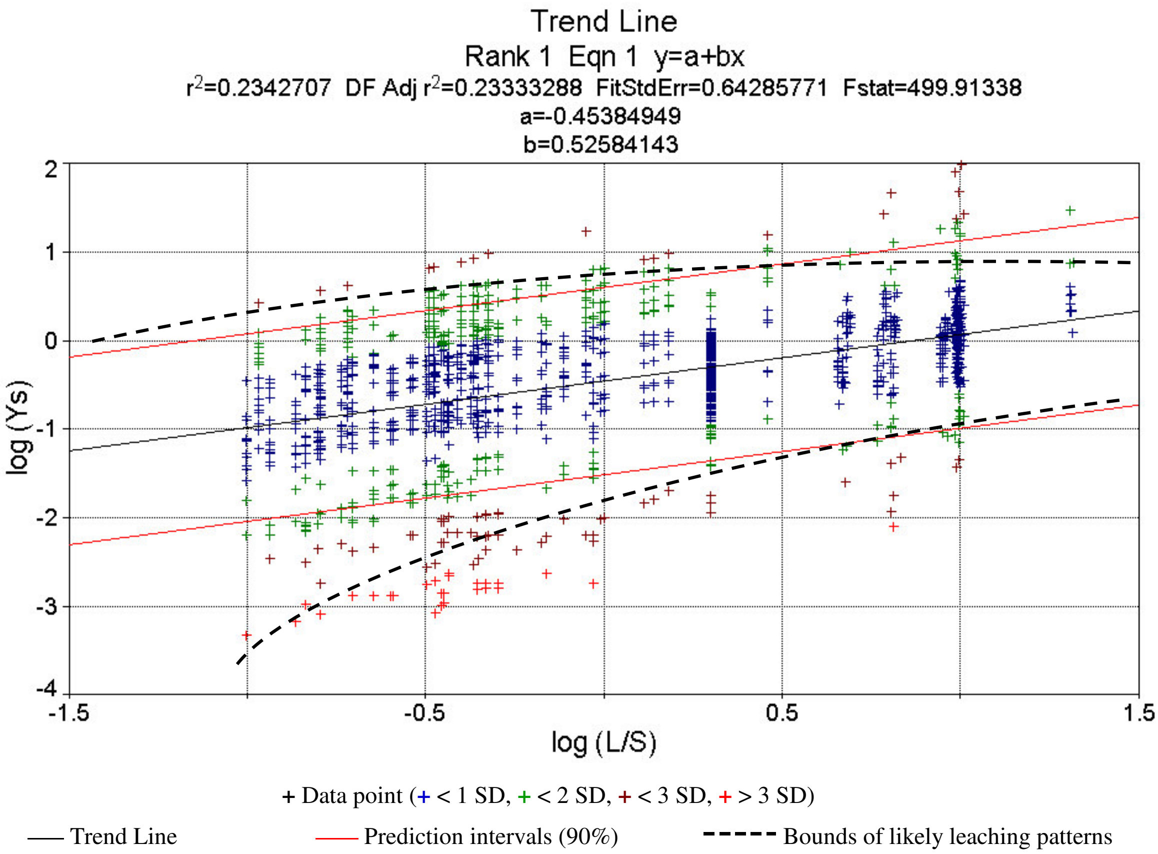


Figure 8-8: Regression analysis of shifted data

The resulting best fit curve is a straight line, of the form,

$$\log Y = \log c + b \log X$$

Equation 8-3

Where, for this data,

$\log c = a =$ Y_s axis intercept = -0.4548

b = gradient of the line = 0.5258

By substituting these values and rearranging, Equation 8-3 becomes:

$$Y_s = (0.3517) X^{0.5258}$$

Equation 8-4

Where,

Y_s = shifted value of cumulative release
X = L/S ratio

Figure 8-8 also shows the 90 percentile prediction lines indicating the uncertainty in predicted individual Y_s values given a known value at a particular L/S ratio. It can be seen that these 90 percentile limits coincide, approximately, with a one order of magnitude variation. Thus, the trend line defined by Equation 8-4 should be applied as a guide only.

Figure 8-8 also shows the approximate lines that bound the range of actual patterns of leaching observed in the laboratory and lysimeter test data (as discussed in Section 8.2). It can be seen that applying the trend line only provides an illustration of likely performance, and that some of the detailed information obtained from the laboratory and field tests is not fully taken into account when using this approach. It can also be seen that the magnitude of the variation from the trend line is greater at lower L/S ratios, with approximately a +1.3 orders of magnitude to -2.5 orders of magnitude variation from the trend line at a L/S ratio of 0.1. The implications of these errors for using the trend line to predict likely values for any L/S ratio, are most significant for those chemical species that have values greater than those indicated by the trend line. For those chemical species that have values that fall below the trendline, for which the magnitude of the likely error is potentially greater, the trend line would provide a conservative assessment of leaching performance, in terms of environmental risk.

It should also be noted that the data on which this trend is founded is largely based upon the more mobile chemical species such as the alkali and alkali earth metals and the group VII species. This is because these species were usually at concentrations above the limits of analytical detection in the leachates from the tests. Other less mobile species, such as the transition metals, were not always detected and are therefore not necessarily represented in the trend line. Such species are also more likely to be affected by other factors such as pH, co-ion effects etc.

8.3.4 Application of the Trend Line

The trend line described in Equation 8-3 and Equation 8-4 can be used for two specific purposes;

1. To estimate from a single laboratory test the potential cumulative release (mg/kg) at any other L/S ratio;
2. To estimate from a single laboratory test the potential concentrations (mg/l) in the leachate at any other L/S ratio.

Being able to obtain estimates of cumulative release (mg/kg) and concentrations (mg/l) in the leachate at any L/S ratio is potentially a very useful tool for the assessment of leaching from aggregate materials, as it helps to relate laboratory determined values with likely in-situ values. For example, if one can determine the flow of leachate through the road then one can determine concentrations (mg/l) and cumulative release (mg/kg) at a given time and also the flux ({concentrations (mg/l) x volume (litres)}/time) of chemical species leached. Predicted concentrations can also be useful in risk assessments to determine changes in concentrations, and thus risk, with time.

Given the availability of a single data point (X_n, Y_n) from, for example, a rapid leaching characterisation test at L/S=10 or the maximum release in a tank-leaching test, the following methods may be employed to predict the cumulative release (mg/kg) and concentrations (mg/l) in the leachate at any other L/S ratio.

From Equation 8-3 it follows that:

$$\log Y - \log Y_n = b(\log X - \log X_n)$$

hence,

$$\log Y = b \log X + (\log Y_n - b \log X_n)$$ **Equation 8-5**

Where,

Y = cumulative release (mg/kg) at X = L/S ratio of choice
Y_n = value of cumulative release (mg/kg) from single laboratory test
X_n = value of L/S ratio from single laboratory test

b = gradient of trend line (as shown in Figure 8-8, and from Equation 8-3 and Equation 8-4).

By comparison of Equation 8-5 and Equation 8-3, it can be seen that the term $(\log Y_n - b \log X_n)$ is a measure of the cumulative release (mg/kg) at L/S ratio of 1 (where log L/S = 0). When this term equals log c (= a) then the reference line is obtained. Otherwise an offset is induced which will be the Shift Factor, F_s.

By substituting,

b = gradient of trend line = 0.5258

Equation 8-5 becomes,

$$\log Y = 0.5258 \log X + \log Y_n - 0.5258 \log X_n$$ **Equation 8-6**

and

$$Y = 10^{(\log Y_n - 0.5258 \log X_n)} . X^{0.5258}$$ **Equation 8-7**

As an example, Figure 8-9 below shows the relationship between L/S ratio and cumulative release (mg/kg) for values of F_s or $(\log Y_n - 0.5258 X_n)$ of -0.5, -0.25, 0, 0.25 and 0.5.

Once again the reader should note that this calculation provides an approximation only of likely cumulative release (mg/kg) at given L/S ratios, and that in practice this value may vary by an order of magnitude and thus only provides a guide.

The relationship between the L/S ratio and the concentrations (mg/l) of chemical species in the leachate can be defined by differentiating Equation 8-7 with respect to the L/S ratio, X, which gives;

$$Y = (0.5258)10^{(\log Y_n - 0.5258 \log X_n)}.X^{(-0.4742)}$$ **Equation 8-8**

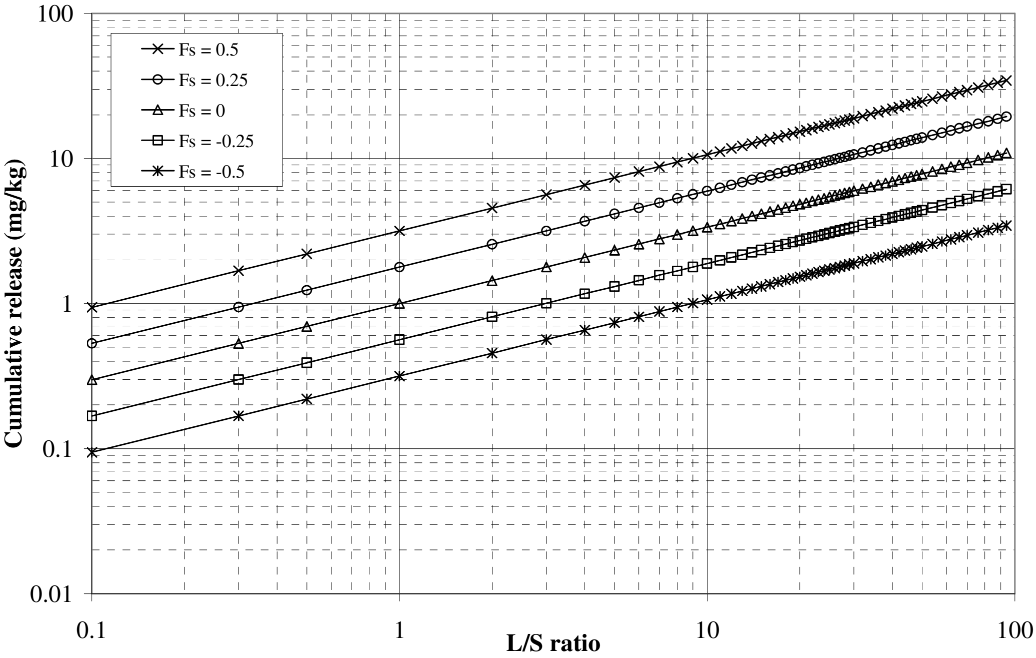


Figure 8-9: Examples of relationship between L/S ratio and cumulative release (mg/kg) obtained using the trend line

As an example, Figure 8-10 below shows the relationship between L/S ratio and species concentrations in the leachate (mg/l) for values of F$_s$ or $(\log Y_n - 0.5258 X_n)$ of -0.5, -0.25, 0, 0.25 and 0.5. F$_s$ can be determined by substituting the data point (X_n, Y_n) into $(\log Y_n - 0.5258 X_n)$.

The same limitations on use are true for estimating concentrations (mg/l) as were for estimating cumulative release (mg/kg), given above.

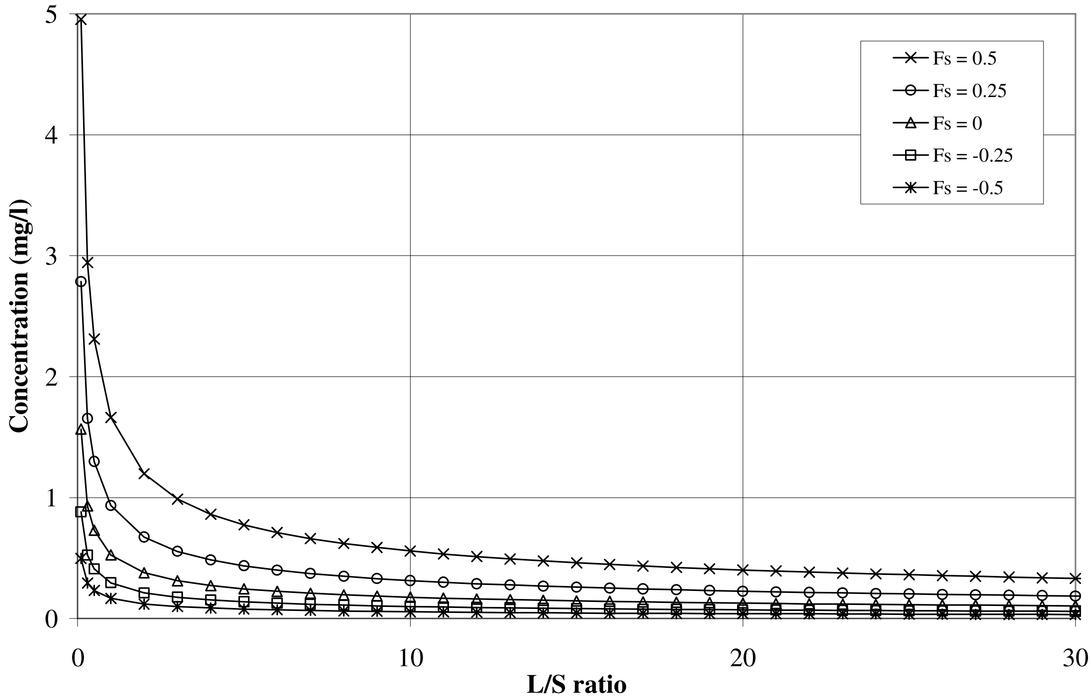


Figure 8-10: Examples of relationship between L/S ratio and species concentrations (mg/l) in the leachate obtained using the trend line

8.3.5 Implications for Assessment

Equation 8-8 and the illustration provided by Figure 8-10 indicate the relatively high concentrations of chemical species that are likely to be found in the leachates at low L/S ratios, which equate to the initial period of leaching of in-situ materials. These leachates, with associated high concentrations of chemical species are, however, only associated with small L/S ratios and thus, relatively small volumes (litres) of leachate (although these volumes would be dependant on the quantity of the material (m^3) used in the construction). It is therefore probable that, an assessment based on the likely concentrations at low L/S ratios would be overly conservative given the probable short duration and small leachate volumes at these elevated concentrations. Thus, a more realistic assessment of concentrations of chemical species in the leachate would be to consider the predicted concentrations at higher L/S ratios, of say 2, for predicting short term concentrations, and 10 for providing a conservative estimate of performance in the longer term, or by incorporating the declining source term defined by Equation 8-8 into an assessment of risk.

8.4 Summary

- The relationship between the laboratory tests and the large-scale lysimeters tests was presented and discussed for:
    - pH verses leached species concentration (mg/l);
    - L/S ratio verses leached species concentration (mg/l);
    - L/S ratio verses cumulative release (mg/kg) of leached species.

- A number of general patterns of leaching were identified. However, considerable scatter in the results was also reported in many cases.

- In general the concentrations (mg/l) of chemical species and the cumulative release (mg/kg) shown in the tank-leaching tests and rapid leaching characterisation test is largely illustrative of the performance of the lysimeter tests at a given L/S ratio.

- The concentrations (mg/l) of chemical species and the cumulative release (mg/kg) shown in the permeameter tests generally under predicts that shown in the other laboratory and lysimeter tests at a given L/S ratio.

- All data obtained from the laboratory and lysimeter tests was used to produce a trend line for cumulative release (mg/kg) against L/S ratio. The trend line was used to derive generic equations that can be used to predict cumulative release (mg/kg) and concentrations (mg/l) for any L/S ratio, given a single data point obtained from a laboratory test at a specific L/S ratio.

9 Application of Findings

9.1 Introduction

The selection of the materials to be used in any road construction project will be dependent on many factors with the principal drivers often being the cost and local availability of a material. Preferred materials may be primary or alternative aggregates and, in either case, it is necessary to demonstrate the suitability of the material for the desired engineering application. The technical suitability of a material should be proven for both structural and environmental performance criteria. To determine the adequacy, or otherwise, of any available material requires a reliable procedure for assessment.

Nunes (1997) and Atkinson et al (1999), amongst others, proposed routine methodologies for determining the suitability of the mechanical performance of both unbound and stabilised material from performance-related testing. The aim of this chapter is to provide a methodology that could be used as the basis for an assessment of environmental acceptability of any mechanically approved material. The methodology draws on the laboratory assessment techniques presented in this thesis and the large-scale trial validation. The assessment methodology presented could be applied to any conventional or alternative aggregate material or mixture.

9.2 Leaching Assessment Methodology

The methodology proposed for the assessment of leaching is presented schematically in Figure 9-1. The procedure is divided into four stages, and assumes that the material or mixture being assessed possesses suitable mechanical performance properties for the proposed application.

- **Stage 1:** Characterise the leaching properties of the material or mixture;
- **Stage 2:** Determine whether these leaching properties are acceptable to the regulator;
- **Stage 3:** Propose or modify binder treatments to improve the leaching properties of the material;
- **Stage 4:** Route to material approval.

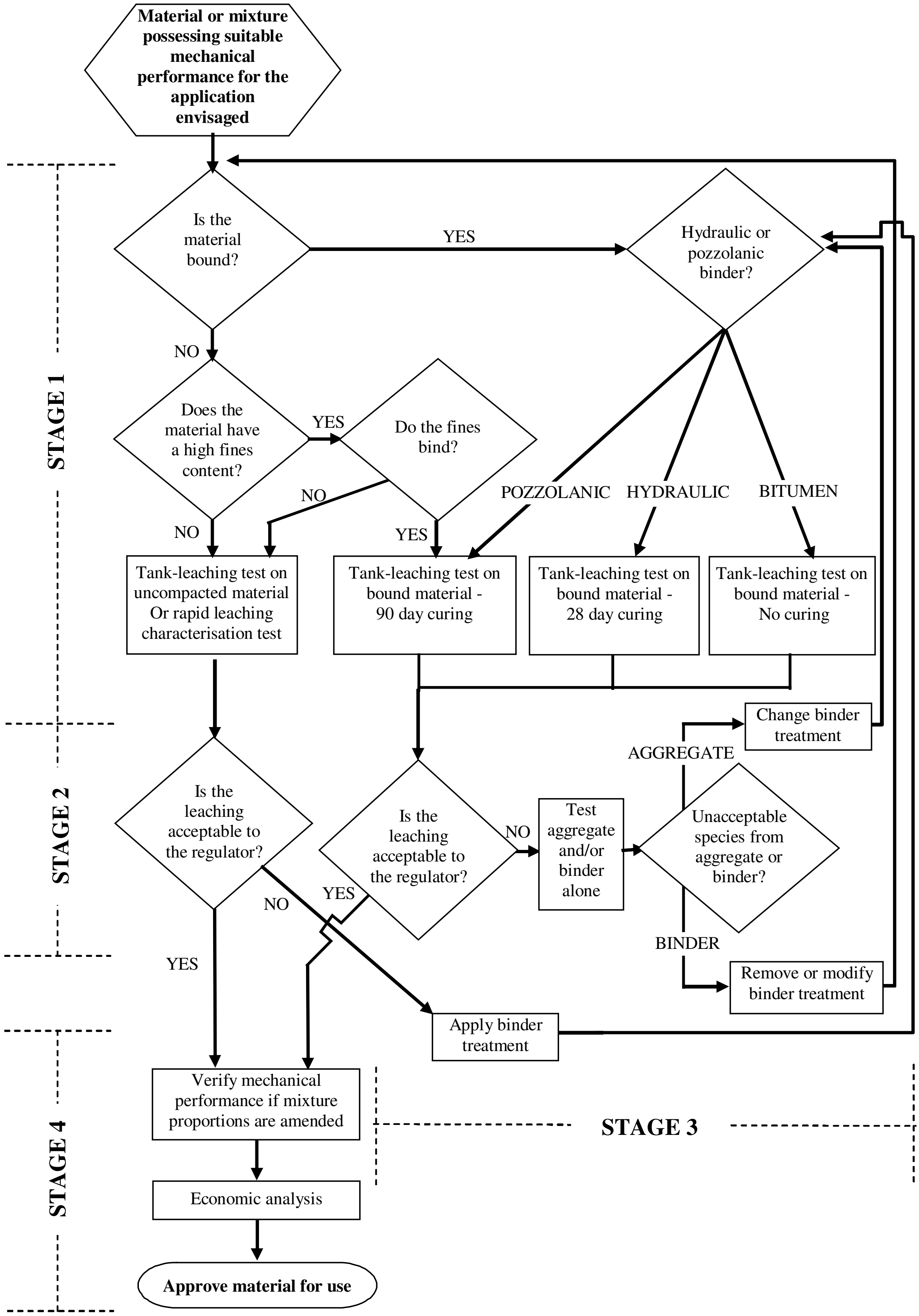


Figure 9-1: Proposed leaching assessment method (Leaching Assessment Chart 1)

STAGE 1

Stage 1 in the assessment method involves the characterisation of the leaching properties of the material or mixture. The leaching test proposed for use is dependant on the material or mixture being tested:

- *Unbound material* – perform either a tank-leaching test on non-compacted material or a rapid leaching characterisation test. If using the tank-leaching test, analysis of the leachate is recommended at time intervals of 6 hours, 24 hours and 4 days. It should not normally be necessary to continue the test beyond 4 days.
- *Bound materials* – perform a tank-leaching test on bound material. The recommended time intervals for analysis of the leachate are 6 hours, 24 hours and 16 days. It should not normally be necessary to continue the test beyond 16 days. The recommended duration for curing the test samples is dependant upon the type of binder, as follows:
    - Bitumen binder – no curing of the test sample is required;
    - Hydraulic binder – a curing period of 28 days is recommended for the test samples. A shorter curing period may be used although the full effect of the binder may not be observed.
    - Pozzolanic binder – a curing period of 90 days is recommended. Again a shorter curing period may be used although the full effect of the binder may not be observed.

STAGE 2

Stage 2 in the assessment method involves using the output from the leaching tests performed in Stage 1 to determine whether the leaching properties of the material are acceptable for the proposed application. The methods used for determining the acceptability of leaching performance are not discussed in detail in this thesis. However, a number of possible approaches are described further in Section 9.4. Any materials deemed to possess an unacceptable performance should then be subjected to Stage 3 of the leaching assessment method. Materials with acceptable leaching performance may continue to Stage 4 of the assessment.

STAGE 3

Stage 3 of the assessment procedure is only required for materials that are not considered to be acceptable to the regulator as determined from the leaching tests carried out in Stage 1 and the assessment carried out in Stage 2. In Stage 3 binder treatments or modifications to existing binder treatments are selected to improve the leaching performance of the unacceptable material. It may be necessary to carry out further testing on the sole aggregate and/or binder in order to determine the source of unacceptable species in a bound mixture. The selection of binder treatments is discussed further in Section 9.3. The material with a new or modified binder treatment is then reassessed using Stage 1 and 2 of the assessment procedure. If the binder treatment results in a mixture with acceptable leaching performance then the material can proceed to Stage 4 of the assessment procedure. If the material is still unacceptable further binder treatments can be selected and assessed through Stage 1 and 2.

STAGE 4

In Stage 4 of the leaching assessment the material with acceptable leaching properties undergoes final approval. The steps to approve the material are likely to include:

- Verification that the mechanical performance of any materials with modified binder treatments are acceptable;
- An analysis of the economics of using the particular material and binder treatment, where proposed;
- Final approval of the material for use.

9.3 Selection of Binder Treatment

Table 9-1 provides a summary of the effect of different binders on leaching. This table is produced from a comparison of the results of all the laboratory leaching tests and lysimeter trials on unbound and bound materials and draws on the similar tables presented previously in Chapters 6 and 7.

Table 9-1: Effect of binder on leaching (Leaching Assessment Chart 2)

Chemical species	**BINDER TREATMENT**					
	Cement	**Bitumen**	**Cement kiln dust + cement**	**Pulverized-fuel ash + lime**	**Granulated blastfurnace slag + lime**	**Flue-gas desulphur gypsum + lime**
pH	+	= / -	+	+	+	=
Conductivity	+++ / +	-- / ---	++++ / +++	++++	++ / +	+ / =
Ca	++++ / ++	++ / --	++++ / ++	++++ / +++	+ / -	-
Mg	- / -----	+++ / --	- ?	- ?	-	-?
Na	+++ / =	= / ----	+++	+++++	++ / -	- / --
K	++++ / -	= / ----	++++ / +++	+++	+ / =	=
HCO_3	+++ / +	- / ---	+++ / ++	++ / + ?	+++	++ / +
Cl	= / --	- / ---	++++	+++	++ / -	--
SO_4	+ / --	- / ---	+++ / ++	++	--	-- / ---
NO3	- / --	+++ / =	+ / =	X	+++	X
Br	-- / ---	- / ---	+ ?	+ ?	+	--
NO_2	+++ / +	- / --	+ ?	+ ?	+++ / +	= / -
F	= / --	++ / --	= / -	=	- / ----	- / --
TOC	+++ / -	++ / ----	+ ?	+ ?	++ / =	=?
TIC	+ / ---	- / ---	= / -	+ / =	++ / +	=
Total P	-	- / --	+ / =	X	--	=
Total S	++ / --	++ / ---	+++ / ++	++	-	-- / ---
NH_4	= / --	= / ---	=	+ / =	+ / =	-?
Si	+ / -	- / ---	++ / =	++ / +	--- / ----	-
Ba	++++ / ++	+ / =	+++	+ ?	++	--
Sr	+++++ / ++	= / --	+++	+ ?	++ / +	--
Mn	+ / ---	= / ---	= / -	- ?	--- / ----	X
Total Fe	+ / ---	+ / ---	+ ?	X	X	-?
Al	++++ / +	= / ----	+++ / ++	++++	- / ---	- / --
Co	X	X	X	X	X	X
Ni	X	X	X	X	-- / ---	X
Cu	-	+ / ----	X	X	-	X
Zn	++ / -	= / -----	=	=	--	- / --?
Cr	X	X	X	+ ?	X	-?
Mo	X	- / ----	X	+ ?	--	---
Cd	X	X	X	X	X	X
Pb	X	X	X	+ ?	X	X
V	X	X	X	+ ?	- ?	--
Li	++++ / +++	= / --	+++	+ ?	+ / -	- / --
B	-	= / --	X	+ ?	-- / ---	- / --?
As	X	X	-	+	X	-- / ---
Se	X	X	+	++	X	--

Key:

Symbol	+++++	++++	+++	++	+	=	-	--	---	----	-----
Cumulative release (mg/kg) from: bound material / unbound material	>1000	1000 to 100	100 to 10	10 to 2	2 to 1.25	1.25 to 0.8	0.8 to 0.5	0.5 to 0.1	0.1 to 0.01	0.01 to 0.001	<0.001

X	Effect of binder cannot be defined except that it does not cause a large increase in leaching
- ?	Binding results in a possible decrease in leaching
+ ?	Binding results in a possible increase in leaching
/	Denotes range of values
=	Denotes no change

Table 9-1 describes the likely effect on leaching of the addition of different types of binder to material mixtures. Table 9-1 can be used in two ways to assist in the selection of appropriate binder treatments to control the leaching of particular chemical species.

1. To propose a binder treatment to reduce the leaching of particular chemical species that are at unacceptable levels;
2. To identify whether the chemical species leaching at unacceptable levels have originated from the binder, and thus whether that binder should be removed or modified to reduce leaching to acceptable levels (only binders included in Figure 9-1 can be used in this way. Other binder treatments will require further testing).

It should be noted that the data presented in Table 9-1 is based on the materials and mixtures tested in this research. The effect of the binder on the leaching of other materials may not always be the same as those used in the production of this table. The leaching properties of any new mixtures should always be confirmed via testing and may require individual testing of the sole aggregate and/or binder as well as the mixture, as proposed in Figure 9-1.

9.4 Acceptability of Leaching Performance

The identification of appropriate methods for assessing the acceptability of leaching performance is considered to be beyond the scope of the work presented in this thesis.

However, in practice the results of a laboratory leaching test alone is unlikely to be sufficient to satisfy a regulatory authority of the acceptability or not of a given material or mixture and it is likely that some form of assessment of the result will be required.

In the UK, methodologies for assessing risks to the environment are commonly used for scenarios such as contaminated land or waste disposal facilities. The risk assessments generally take a tiered approach with the complexity of the assessment increasing with the magnitude of the risks and the probability of their occurrence. The risk assessment usually uses a ‘source – pathway – receptor’ model that is based on the assumption that a risk to a receptor is present where there is a source of contamination and a pathway between the source of contamination and the receptor. For an aggregate in the sub-base layer of a road an example of the model may be [Hooper et al., 2004]:

- Source: Leaching of chemical species from the aggregate material (as addressed in this thesis) and characterised through simple tests and the use of Equation 8-7 and Equation 8-8;
- Pathway: Water movement through the sub-base into:
    - an adjacent drainage ditch that flows into a public surface water drainage system;
    - the soil pore water beneath the road;
    - the ground below, through the ground into an aquifer which is use to supply drinking water;
    - highway drainage where it combines with surface run-off water from the road and is then treated before discharge into a local stream;
- Receptor: Depending on the site, may include:
    - The public surface water drainage system;
    - The soil pore water;
    - The aquifer;
    - The local river.

9.5 Summary

- This chapter proposes a methodology for assessing the leaching of aggregate materials and mixtures for use in road construction.

- A number of leaching tests are proposed for use and the choice of test is dependant on the properties of the material to be tested. The proposed tests are relatively simple to perform and could be carried out within a similar timescale to any mechanical performance tests that may be required

- A matrix is presented to assist in the selection of appropriate binder treatments to control the leaching of particular chemical species. Applications of the matrix are:
    - To propose a binder treatment to reduce the leaching of particular chemical species that are at unacceptable levels;

- To identify whether the chemical species leaching at unacceptable levels have originated from the binder, and thus whether that binder should be removed or modified to reduce leaching to acceptable levels.

10 Conclusions and Recommendations for Future Research

10.1 Introduction

The research presented in this thesis has been concerned with the determination and assessment of the leaching of chemical species from conventional and alternative aggregate materials and mixtures when utilised in road construction or bulk fill applications.

This chapter provides a summary of the research carried out and the main conclusions presented in this thesis. The conclusions have been grouped into the following sections: Laboratory Testing; Effect of Binding on Leaching; Large-scale Simulations; and Assessment and Implications.

10.2 Conclusions

10.2.1 Laboratory Testing

A total of 14 aggregates and 6 binders were chosen for investigation in this thesis. The selected materials included both conventional and alternative aggregates and binders to enable the leaching properties shown by widely accepted materials to be compared to those from secondary sources.

A series of laboratory tests were used to characterise and assess the leaching properties of the aggregates and binders. The tests were designed to determine the effects of material particle size, compaction and the hydraulic regime of the in-situ leaching environment on the leaching properties of the materials. Test apparatus were designed, fabricated and adjusted to enable particle size reduction and leaching vessel agitation and, more generally, to enable the tests to be performed.

The analysis of the chemical composition of the solid fraction of each material revealed that the concentrations of each chemical species within the solid matrix varied over several orders of magnitude between the different materials. The major proportion of each of the materials was silicon or calcium, with iron, sulphur and aluminium also making significant contributions. A wide range of trace elements were also detected.

A rapid leaching characterisation test was used to obtain a rapid estimate of the leachability of the different aggregates and binders. In general, the alkali and alkali earth metals were the most mobile species, with the exception of magnesium and sometimes the barium and strontium that exhibited lower mobility. The transition metals generally exhibited lower mobility, except for molybdenum and, in some cases, zinc and chromium.

A tank-leaching test methodology was adapted from existing standards to enable conventional and alternative aggregate materials to be tested at realistic gradings in both non-compacted and compacted forms.

The tank-leaching tests showed a general trend of an increase in leached species over time, with uncompacted materials generally reaching equilibrium conditions after 2 to 4 days and compacted materials over the longer period of 4 to 16 days.

From the tank-leaching tests on materials at realistic gradings it was concluded that the rate of leaching was affected by both material particle size and sample agitation, with the rate of leaching being slower from large particles and without sample agitation.

From the tank-leaching tests on compacted materials it was shown that compaction reduces the rate of leaching from most materials. The effect of compaction was most pronounced for the materials with high fines content within a coarse aggregate that exhibit a relatively low compacted permeability compared to their uncompacted form.

Two permeameter test methodologies were utilised for the assessment of leaching by advection from coarse and fine graded aggregates. The equipment and methodologies were adapted to enable the tests to be performed and the leachate sampled in a way that was suitable for a leaching test.

The general trend shown by the data in the permeameter tests was a decrease in species concentrations with an increasing number of bed volumes passing, or L/S ratio.

The concentration of chemical species in the leachates and the cumulative release (mg/kg) at a given L/S ratio were generally lower in the permeameter tests than in the tank-leaching test on the same material. This was attributed to the relatively short contact time shown in the permeameters and therefore concluded that steady state conditions were not established.

10.2.2 Effect of Binding on Leaching

The effect of binding on the leaching properties of different aggregates and mixtures was determined using the laboratory leaching tests developed.

The rapid leaching characterisation test was used to compare the leaching properties of unhydrated hydraulic binders with those of the cured mortars of these binders. The results showed significant reductions in the leaching of many species from the cured mortars when compared to the unhydrated binder. It was concluded that any assessment of leachability from binders or mixtures should be carried out on the materials in the form in which they are to be utilised.

Tank-leaching and permeameter tests were used to assess diffusive and advective leaching from bound materials. The results showed that aggregates bound with the cement and lime based binders produced leachates with a high pH and increased conductivity and alkali and alkali earth metals. Materials bound with bitumen showed similar or lower pH and conductivity than that shown by the unbound material and a general reduction in the rate and quantity of leaching of many other chemical species.

In a number of cases the addition of a binder treatment resulted in increases in the rate and quantity of leaching of many chemical species. This was particularly apparent for aggregates that produced very low concentrations of leached species when tested solely. The detrimental as well as the positive effects of binding on leaching performance should be assessed to determine whether the mixture is acceptable.

The relationship between the results of the unbound and bound material tests was used to produce a matrix illustrating the effect of binding on the leaching of each chemical species from the mixtures tested. The matrix can be used to identify binder treatments that are likely to reduce the leaching of specific chemical species and also to determine whether a particular chemical species leached from a bound mixture originates from the aggregate or the binder.

10.2.3 Large-Scale Simulations

Large-scale lysimeter tests were designed and constructed to simulate the physical dimensions, material properties and environmental conditions of aggregate materials within a road construction or bulk fill. Nine materials and mixtures were examined.

The absolute concentrations of chemical species within the leachates from the lysimeter tests were generally higher than those observed in the laboratory tests. This is probably due to the lower L/S ratios in the lysimeters resulting in less dilution of the leached species.

The concentrations of chemical species in the leachates from the lysimeters generally reduced with increasing time and L/S ratio. It can thus be concluded that the source of chemical species from aggregate materials in road construction or bulk fill applications is likely to decline and that leaching will improve over time (i.e. lower concentrations).

The patterns of leaching shown by different chemical species were often similar for a particular material, although the absolute concentrations differed.

The effect of binding on leaching was determined for three mixtures by comparison to the observed leaching from the sole aggregate. In general, binding resulted in a decrease in the cumulative release (mg/kg) of many of the chemical species, but an increase in some.

The concentrations of chemical species in leachates often form the basis of an assessment of environmental acceptance. Thus the higher concentrations observed at lower L/S ratios in the lysimeters could have a significant impact on what is deemed to be acceptable data for use in assessments given the probable low L/S ratios in a road construction. The use of flux rate, which considers leachate volumes as well as concentrations (mg/l), is likely to be more appropriate than an assessment based purely on concentrations. Alternatively the use of a declining source term, such as that defined by the model presented in Chapter 8, may be used.

10.2.4 Assessment and Implications

The large number of laboratory and field scale leaching experiments conducted during this research generated a significant database of information on the leaching properties of many conventional and alternative aggregates, binders and mixtures. This data can be used by industry in the assessment of the environmental acceptability of these materials and can also be used to indicate how other similar materials may leach.

A direct comparison of leaching test results to common acceptability criteria revealed that, based on such an assessment, all materials, including conventional limestone and granite would be deemed unacceptable for use. Thus it is concluded that such an assessment system is inappropriate, and that some consideration of the in-situ environmental conditions is required to ensure that the assessment is not overly conservative.

The relationship between the laboratory tests and the large-scale lysimeters tests was presented and discussed for:

- pH verses leached species concentration (mg/l);
- L/S ratio verses leached species concentration (mg/l);
- L/S ratio verses cumulative release (mg/kg) of leached species.

In general the concentrations (mg/l) of chemical species and the cumulative release (mg/kg) shown in the tank-leaching tests and rapid leaching characterisation test is largely illustrative of the performance of the lysimeter tests at a given L/S ratio.

The concentrations (mg/l) of chemical species and the cumulative release (mg/kg) shown in the permeameter tests generally under predicts that shown in the other laboratory and lysimeter tests at a given L/S ratio. The lower concentrations (mg/l) of chemical species in the leachates from the permeameters tests show that this type of test is generally less suitable for a rapid assessment of leachability, since the result of the test is unconservative in terms of environmental assessment.

Data obtained from selected laboratory tests and the lysimeters was used to produce a trend line for cumulative release (mg/kg) against L/S ratio. The trend line was used to derive generic equations that can be used to predict cumulative release (mg/kg) and concentrations (mg/l) for any L/S ratio, given a single data point obtained from a simple laboratory test at a specific L/S ratio.

A methodology for assessing the leaching properties of aggregate materials and mixtures for use in road construction and bulk fill is presented. The methodology includes recommendations for the leaching tests to be used for different types of materials and mixtures.

A matrix is presented to assist in the selection of appropriate binder treatments to control the leaching of particular chemical species. Applications of the matrix are:

- To propose a binder treatment to reduce the leaching of particular chemical species that are at unacceptable levels;
- To identify whether the chemical species leaching at unacceptable levels have originated from the binder in the mixture, and thus whether that binder should be removed or modified to reduce leaching to acceptable levels.

10.3 Recommendations for Future Research

10.3.1 Laboratory Testing

- Investigate how the results of the laboratory leaching tests used in this research compare to the results of tests being developed as European Standards by CEN 154 and CEN 292 and any others.

- Determine whether the leaching of organic constituents from aggregates, binders and mixtures are at concentrations that may cause concerns to the regulator. Leaching tests specifically designed to retain and not cross-contaminate with organic species would be required.

- Carry out an assessment of the leaching properties of a wider range of primary aggregate materials to determine what is generally considered to be acceptable levels (concentrations (mg/l) or flux) of leaching. A comparison of the leaching from alternative materials with those of generally accepted materials could be used to produce a benchmark to define environmental acceptability for any alternative aggregate or mixture.

10.3.2 Effect of Binding on Leaching

- Develop a methodology for optimising binder treatments to control the leaching of particular chemical species. Mixtures containing different proportions, quantities and types of binders could be compared to determine the most efficient treatments for different types of materials and chemical species. A similar approach exists for the optimisation of mechanical performance.

- Investigate the effect of curing time on the performance of different binders at controlling leaching of particular chemical species. Again similar guidance is currently available for mechanical performance that indicates the curing time required to achieve certain proportions of optimum performance.

- Investigate the long-term performance of binders as a method for controlling leaching. The laboratory testing and lysimeters performed in this research were carried out over relatively short durations compared to the lifetime of a road construction and bulk fill. In many cases the observed test results indicated a slower rate of leaching initially, when compared to the sole material, but with increasing time or L/S ratio the rate of leaching generally began to increase and thus the long-term performance of the binder requires investigation.

10.3.3 Large-Scale Simulations

- To further examine the data obtained from this research to identify whether additional trends can be found to express the relationship between the leaching observed in the laboratory with that which is likely in the field. Trend lines for individual chemical species and for each material and mixture could be produced to enable a more accurate assessment of change in leached species with increasing L/S ratio or time.

10.3.4 Assessment and Implications

- Determine a methodology for assessing the migration of leached species (determined from leaching tests) into the wider environment for a road construction or bulk fill utilisation scenario. The assessment of risk would involve the consideration of the hydraulic conditions within the construction and the wider environment together with the natural processes of attenuation that occur in subsurface and water environments.

- Assess the environmental costs and benefits of using alternative materials as road construction materials when compared to the environmental issues associated with primary aggregate extraction such as noise, dust, depletion of finite resources and damage to visual amenity, landscape and wildlife.

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APPENDIX A:

Quality Standards

List I and List II substances – from the Groundwater Directive

List I of Families and Groups of Substances

1	Organohalogen compounds and substances which may form such compound in the aquatic environment.
2	Organophosphorus compounds.
3	Organotin compounds.
4	Substances which possess carcinogenic, mutagenic or teratogenic properties in or via the aquatic environment.
5	Mercury and its compounds.
6	Cadmium and its compounds.
7	Mineral oils and hydrocarbons.
8	Cyanides.

NOTE : List I contains the individual substances which belong to the families and groups of substances specified above, with the exception of those which are considered inappropriate to List I on the basis of a low risk toxicity, persistence or bioaccumulation. Such substances which with regard to toxicity, persistence and bioaccumulation are appropriate to List II are to be classed in List II.

List II of Families and Groups of Substances

<table>
<tr><td>1</td><td colspan="2">The following metalloids and metals and their compounds:</td></tr>
<tr><td rowspan="10"></td><td>1. Zinc</td><td>11. Tin</td></tr>
<tr><td>2. Copper</td><td>12. Barium</td></tr>
<tr><td>3. Nickel</td><td>13. Beryllium</td></tr>
<tr><td>4. Chrome</td><td>14. Boron</td></tr>
<tr><td>5. Lead</td><td>15. Uranium</td></tr>
<tr><td>6. Selenium</td><td>16. Vanadium</td></tr>
<tr><td>7. Arsenic</td><td>17. Cobalt</td></tr>
<tr><td>8. Antimony</td><td>18. Thallium</td></tr>
<tr><td>9. Molybdenum</td><td>19. Tellurium</td></tr>
<tr><td>10. Titanium</td><td>20. Silver</td></tr>
<tr><td>2</td><td colspan="2">Biocides and their derivatives not appearing in List I.</td></tr>
<tr><td>3</td><td colspan="2">Substances which have a deleterious effect on the taste and/or odour of groundwater and compounds liable to cause the formation of such substances in such water and to render it unfit for human consumption.</td></tr>
<tr><td>4</td><td colspan="2">Toxic or persistent organic compounds of silicon and substances which may cause the formation of such compounds in water, excluding those which are biologically harmless or are rapidly converted in water into harmless substances.</td></tr>
<tr><td>5</td><td colspan="2">Inorganic compounds of phosphorus and elemental phosphorus.</td></tr>
<tr><td>6</td><td colspan="2">Fluorides.</td></tr>
<tr><td>7</td><td colspan="2">Ammonia and nitrates.</td></tr>
</table>

Dutch Regulations on Soil Quality

Target and intervention values for micropollutants for a standard soil (10% organic material and 25% clay); mg/kg for soil/sediment, µg/l for groundwater, unless otherwise stated.

Substance	**Soil/sediment (mg/kg dry material)**		**Groundwater (µg/l)**	
	Target value	**Intervention value**	**Target value**	**Intervention value**
I metals				
Arsenic	29	55	10	60
Barium	200	625	50	625
Cadmium	0.8	12	0.4	6
Chromium	100	380	1	30
Cobalt	20	240	20	100
Copper	36	190	15	75
Mercury	0.3	10	0.05	0.3
Lead	85	530	15	75
Molybdenum	10	200	5	300
Nickel	35	210	15	75
Zinc	140	720	65	800
II Inorganic Compounds				
Cyanide – free	1	20	5	1500
Cyanide complex (pH <5)[1]	5	650	10	1500
Cyanide complex (pH≥5)	5	50	10	1500
Thiocyanates (total)		20		1500
(from earlier legislation[2])				
(Ammonia)				*(3000)*
(Fluoride)				*(4000)*
(Sulphur (total))				*(300)*
(Bromine (total))				*(2000)*
(Phosphate)				*(700)*
III Aromatic Compounds				
Benzene	0.05 (d)	1	0.2	30
Ethyl benzene	0.05 (d)	50	0.2	150
Phenol	0.05 (d)	40	0.2	2000
Cresoles (total)		5	(d)	200
Toluene	0.05 (d)	130	0.2	1000
Xylene	0.05 (d)	25	0.2	70
Catechol		20	(d)	1250
Resorcinol		10		600
Hydrochinon		10		800
IV Polycyclic Aromatic Hydrocarbons (PAHs)				
PAH (total of 10)[2, 11]	1	40	-	-
Naphthalene			0.1	70
Anthracene			0.02	5
Phenantrene			0.02	5
Fluoranthene			0.005	1
Benzo(a)anthracene			0.002	0.5
Chrysene			0.002	0.05
Benzo(a)pyrene			0.001	0.05
Benzo(ghi)perylene			0.001	0.05

Benzo(k)fluoranthene			0.0004	0.05
Indeno(1,2,3-cd)pyrene			0.0002	0.05
V Chlorinated Hydrocarbons				
1,2-dichloroethane	-	4	0.01 (d)	400
Dichloromethane	(d)	20	0.01 (d)	1000
Tetrachloromethane	0.001	1	0.01 (d)	10
Tetrachloroethane	0.01	4	0.01 (d)	40
Trichloromethane	0.001	10	0.01 (d)	400
Trichloroethane	0.001	60	0.01 (d)	500
Vinylchloride	-	0.1	-	0.7
Chlorobenzenes (total)[3, 11]	-	30	-	-
Monochlorobenzene	(d)	-	0.01 (d)	180
Dichlorobenzenes (total)	0.01	-	0.01 (d)	50
Trichlorobenzenes (total)	0.01	-	0.01 (d)	10
Tetrachlorobenzenes (total)	0.01	-	0.01 (d)	2.5
Pentachlorobenzene	0.0025	-	0.01 (d)	1
Hexachlorobenzene	0.0025	-	0.01 (d)	0.5
Chlorophenols (total)[3, 11]	-	10	-	-
Monochlorophenols (total)	0.0025	-	0.25	100
Dichlorophenols (total)	0.003	-	0.08	30
Trichlorophenols (total)	0.001	-	0.025	10
Tetrachlorophenols (total)	0.001	-	0.01	10
Pentachlorophenol	0.002	-	0.02	3
Chloronaphthalene	-	10	-	6
Polychlorobiphenyls (total of 7)[5]	0.02	1	0.01 (d)	0.01
VI Pesticides				
DDT / DDE / DDD [6]	0.0025	4	(d)	0.01
Drins[7]	-	4	-	0.1
Aldrin	0.0025	-	(d)	-
Dieldrin	0.0005	-	0.02 ng/l	-
Endrin	0.001	-	(d)	-
HCH compounds[8]	-	2	-	1
α-HCH	0.0025	-	(d)	-
β-HCH	0.001	-	(d)	-
γ-HCH	0.05 µg/kg	-	0.2 ng/l	-
Carbaryl	-	5	0.01 (d)	0.1
Carbofuran	-	2	0.01 (d)	0.1
maneb	-	35	(d)	0.1
atrazin	0.05 µg/kg	6	0.0075	150
VII Other Pollutants				
Cyclohexanon	0.1	270	0.5	15000
Phthalates (total)[9]	0.1	60	0.5	5
Mineral oil	50	5000	50	600
Pyridine	0.1	1	0.5	3
Styrene	0.1	100	0.5	300
Tetrahydrofuran	0.1	0.4	0.5	1
Tetrahydrothiophene	0.1	90	0.5	30

(d) = detection threshold

NOTE:

1. Acidity: pH (0.01M $CaCl_2$). In order to determine whether pH is greater than or equal to 5, or less than 5, the 90 percentile of the measured values is taken.
2. "PAH (total of 10)" here means the total of anthracene, benzo(a)anthracene, benzo(k)fluoroanthene, benzo(a)pyrene, chrysene, phenantrene, fluoroanthene, indeno(1,2,3-cd)pyrene, naphthalene and benzo(ghi)perylene.
3. "Chlorobenzenes (total)" here means the total of all chlorobenzenes (mono-, di-, tri-, tetra-, penta- and hexachlorobenzene).
4. "Chlorophenols (total)" here means the total of all chlorophenols (mono-, di-, tri-, tetra-, pentachlorophenol).
5. In the case of the intervention value, "polychlorobiphenyls (total)" means the total of PCBs 28, 52, 101, 118, 138, 153 and 180. For the target value it refers to the total excluding PCB 118.
6. "DDT / DDD / DDE" means the total of DDT, DDD and DDE.
7. "Drins" means the total of aldrin, dieldrin and endrin.
8. "HCH compounds" means the total of α-HCH, β-HCH, γ-HCH and δ-HCH.
9. "Phthalates (total) means the total of all phthalates.
10. "Mineral oil" means the sum of all the alkanes, both straight-chain and branched-chain. Where the contamination is due to mixtures (e.g. gasoline or domestic heating oil), then not only the alkane content but also the content of aromatic and/or polycyclic aromatic hydrocarbons must be determined. This aggregate parameter has been adopted for practical reasons. Further toxicological and chemical disaggregation is under study.
11. The values for total polycyclic aromatic hydrocarbons, total chlorophenols and total chlorobenzenes in soil/sediments apply to the total concentration of the compounds belonging to the relevant category. If the contamination is due to only one compound of a category, the value used is the intervention value for that compound, where there are two or more compounds the value for the total of these compounds applies, etc. For soil/sediment, effects are directly additive (i.e. 1mg of substance A has the same effect as 1mg of substance B) and can be checked/compared against an aggregate standard by summing the concentrations of the substances involved. For further information about this additivity see, for example, the Technical Committee for Soil Protection (1989)[1]. In the case of groundwater, effects are indirect, and are expressed as a fraction of the individual intervention values before being summed (i.e. 0.5 of the intervention value of substance A has the same effect as 0.5 of the intervention value of substance B). This means that an additional formula must be used to determine whether an intervention value is exceeded. The intervention value for a category of substances is exceeded if:

$$\sum \frac{conc_i}{I_i} \geq 1, \text{ waarbij}$$

Where,

$Conc_i$ = measured concentration of substance i in the category concerned

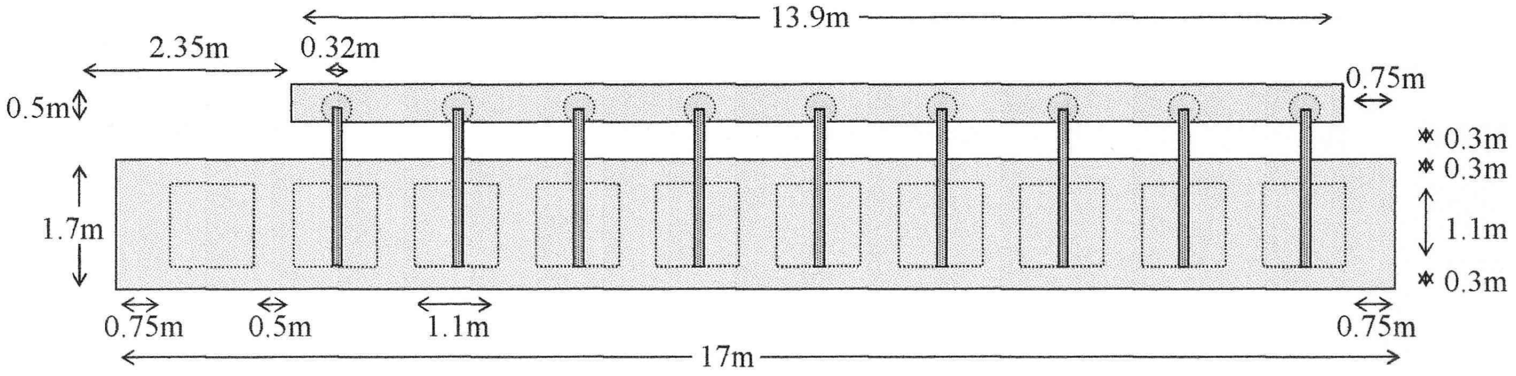
I_i = intervention value for substance i.

[[1] Technical Committee for Soil Protection (1989). "Advies beoordeling van bodemverontreiniging met polycyclische aromaten" (Advice regarding the assessment of soil contamination with polycyclic aromatics). TCB A89/03.]

[[2] from Guy, S. (1993). "Types of Pollutants". Groundwater Pollution: liabilities and Engineering Implications, East Midlands Geotechnical Group (EMGG), University of Nottingham, October 1993. Jones, R. H., Rogers, C. D. F. and Holden, J. M. W. (Eds.).]

APPENDIX B:

Lysimeter Drawings



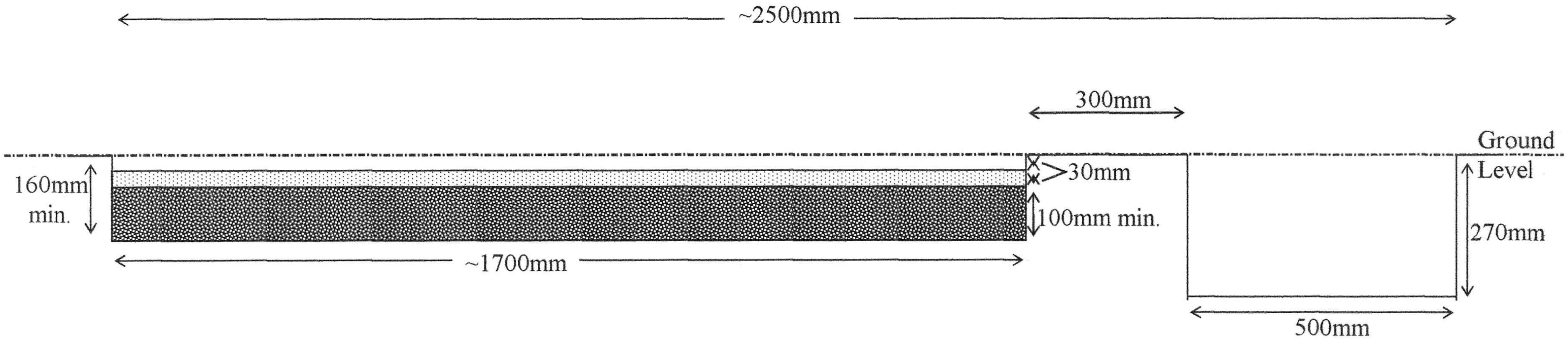
Plan view of area for excavation and location of individual lysimeters

▨ Area for excavation

⬚ Location of individual lysimeters

○ Leachate collection chamber

| Leachate collection pipe



End view of lysimeter site excavation and backfill

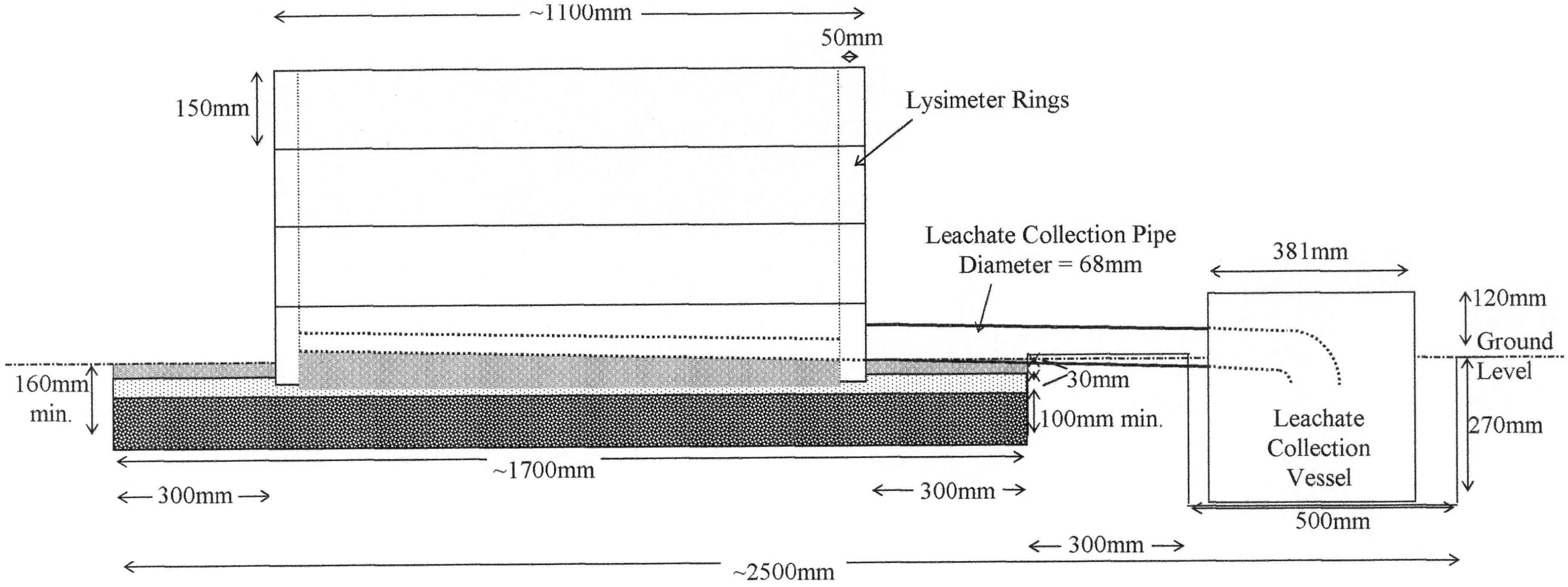
▩ Compacted granular fill to a minimum depth of 100mm

░ Compacted sand to a depth of 30mm

Volume of soil to excavate =
(17m x 1.7m x 0.16m) + (13.9m x 0.5m x 0.5m)
= 6.5 m^3

Volume of compacted granular fill =
17m x 1.7m x 0.1m = 2.9 m^3 (approx. = 7 tonnes)

Volume of compacted sand =
17m x 1.7m x 0.03m = 0.9 m^3 (approx. = 2 tonnes)



End view of site excavation and lysimeter construction

▩ Compacted granular fill to a minimum depth of 100mm

▨ Concrete infill

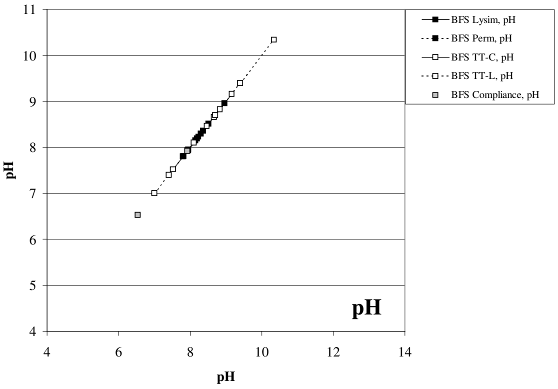
░ Compacted sand to a depth of 30mm

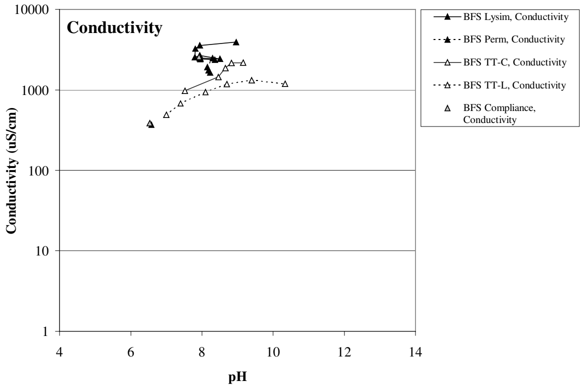
APPENDIX C:

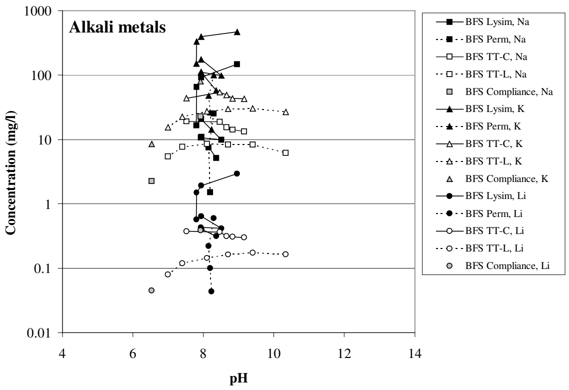
Graphs comparing laboratory leaching tests and lysimeters

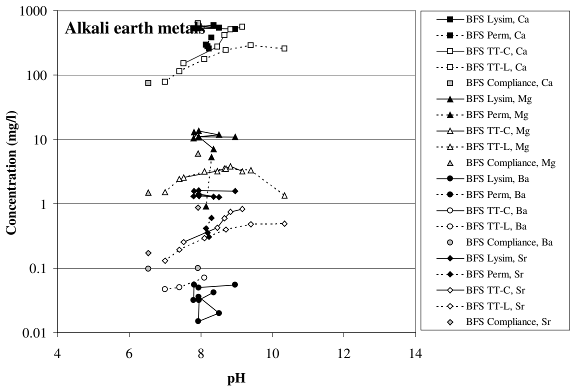
pH verses leached species concentration (mg/l)

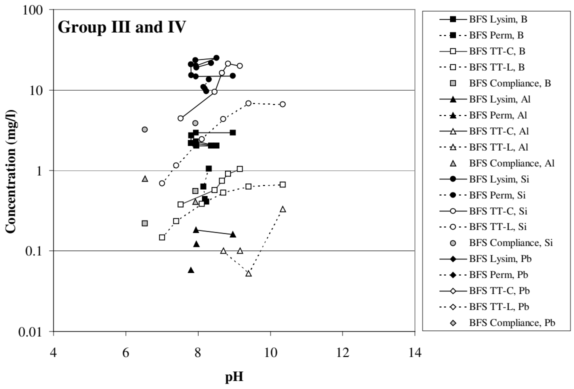
BLASTFURNACE SLAG
Concentration in the leachate (mg/l) against pH

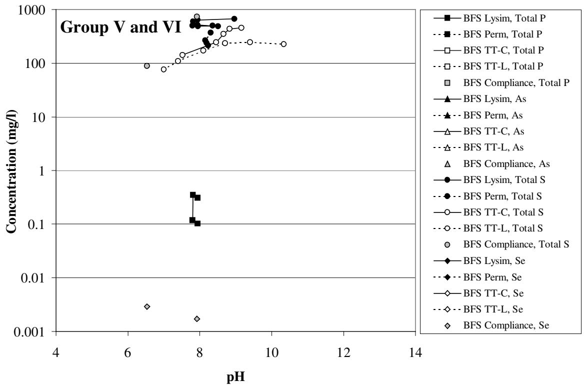


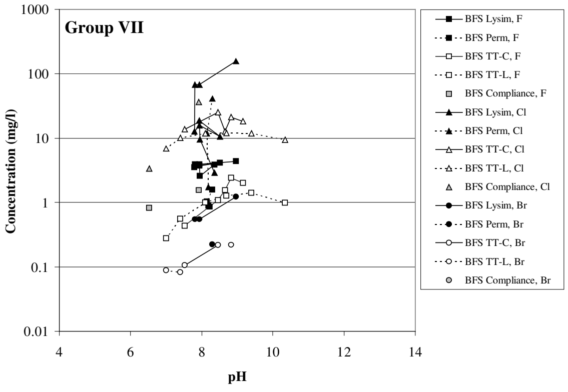


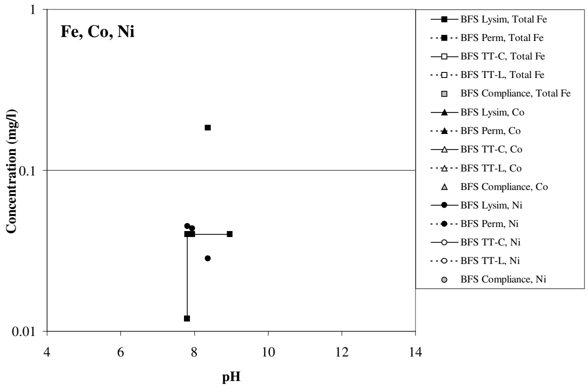




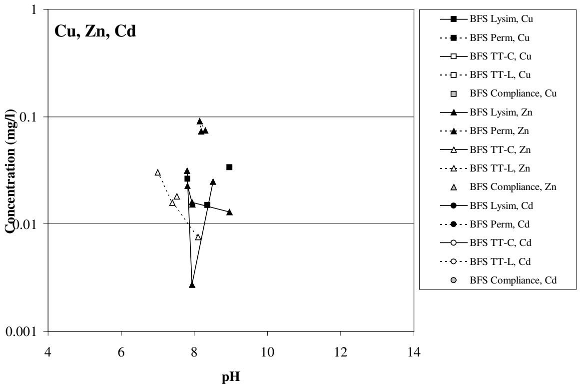


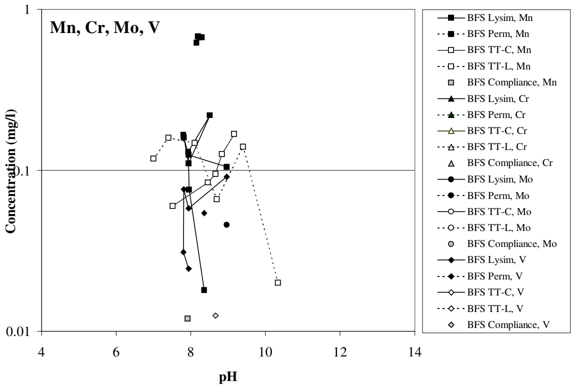


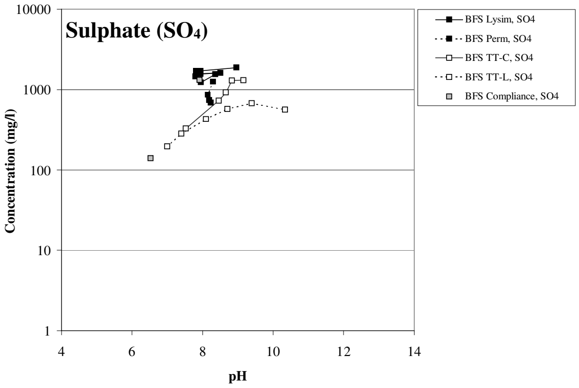


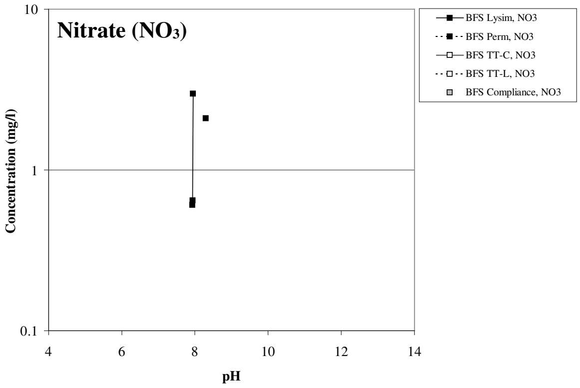


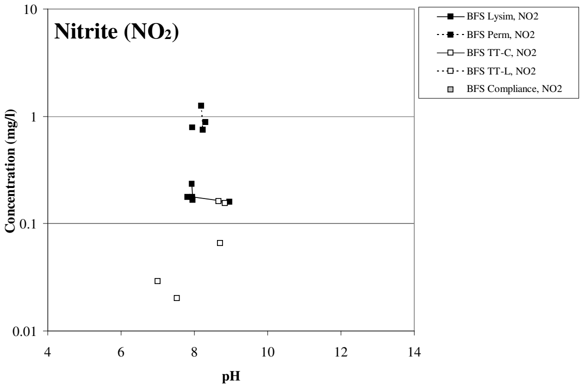
BLASTFURNACE SLAG
Concentration in the leachate (mg/l) against pH

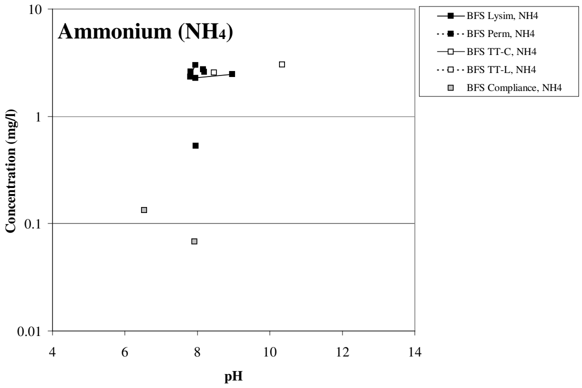




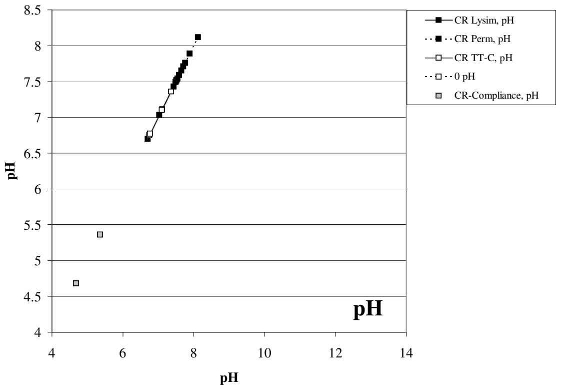


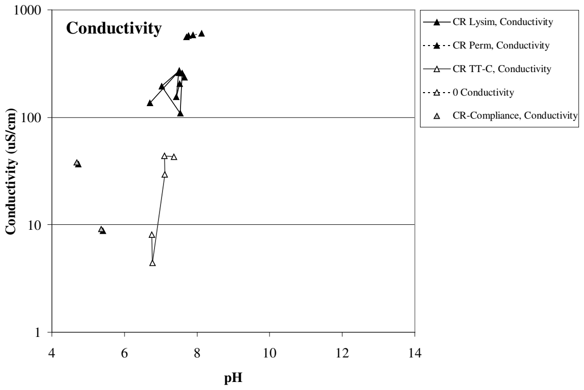


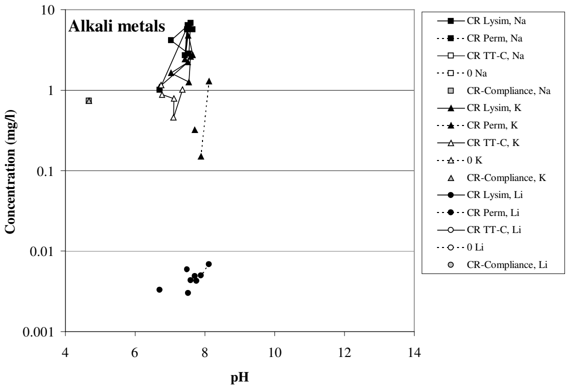


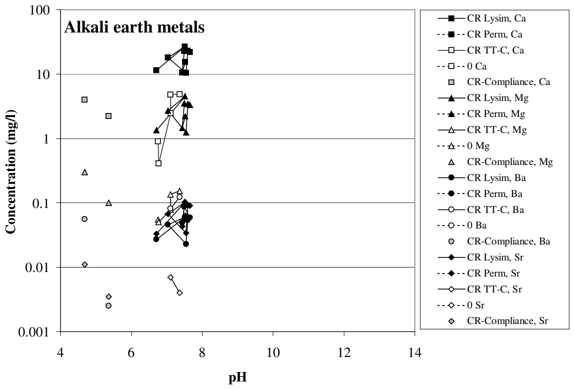


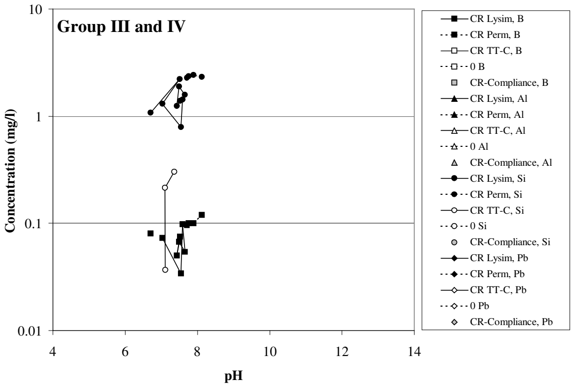
CRUMBED RUBBER (2-8mm)
Concentration in the leachate (mg/l) against pH

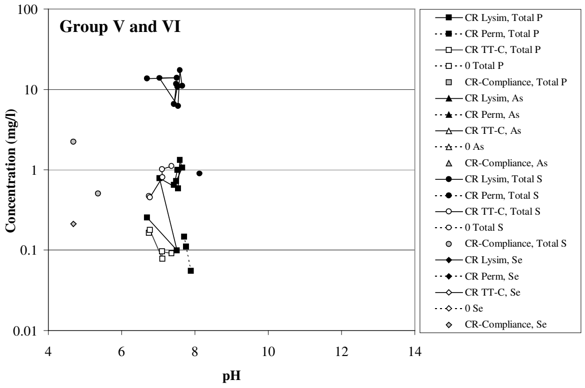


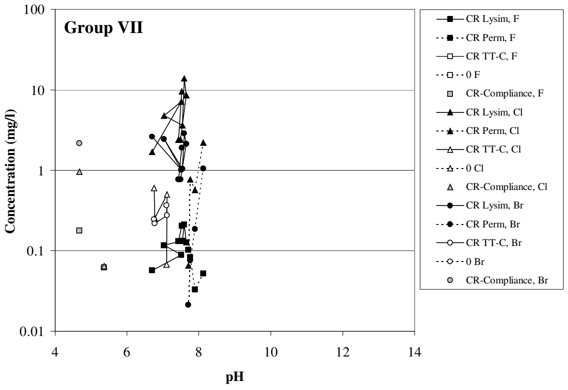


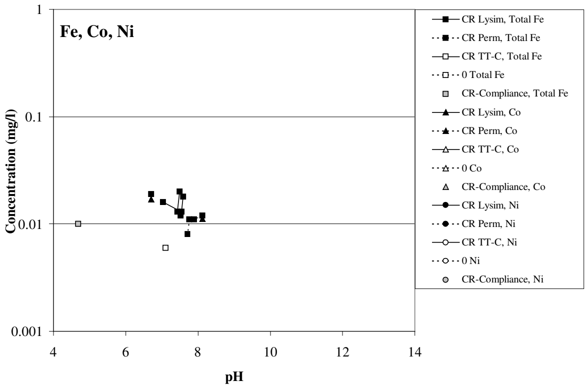




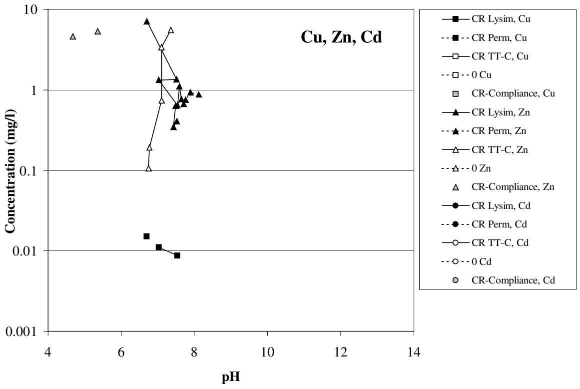


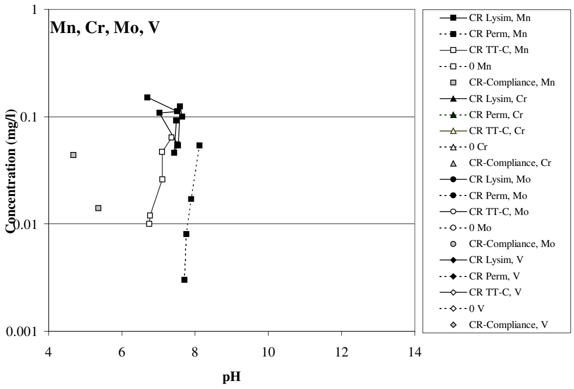


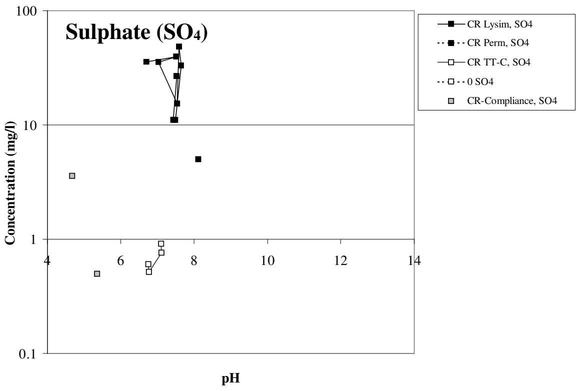


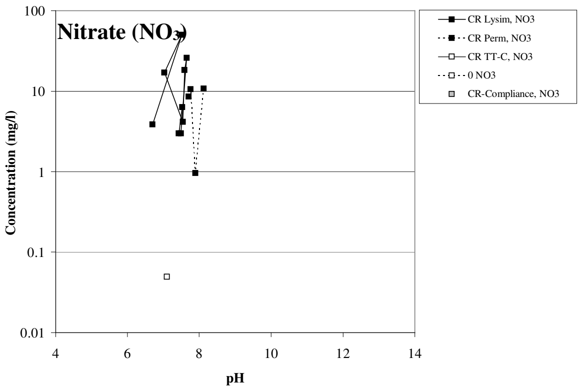


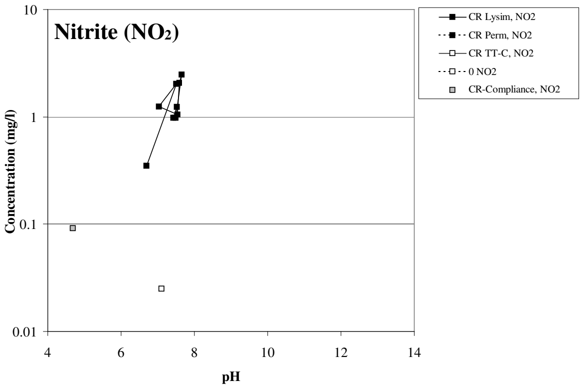
CRUMBED RUBBER (2-8mm)
Concentration in the leachate (mg/l) against pH

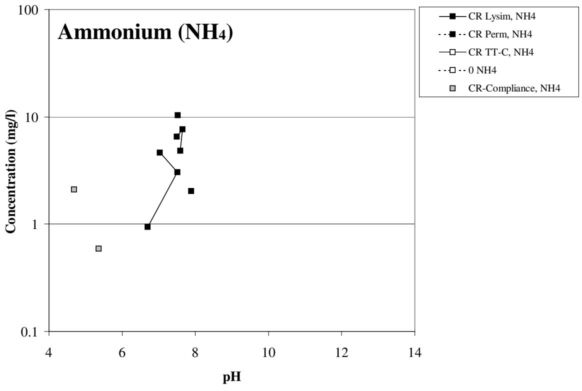




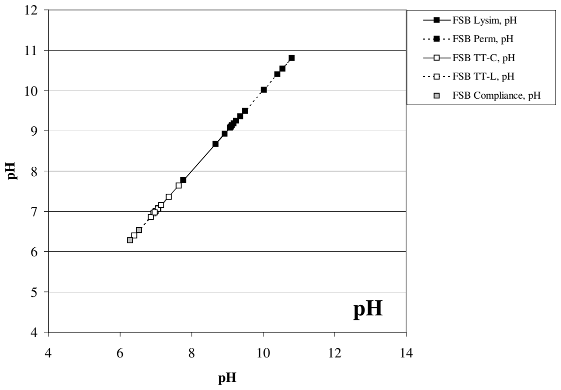


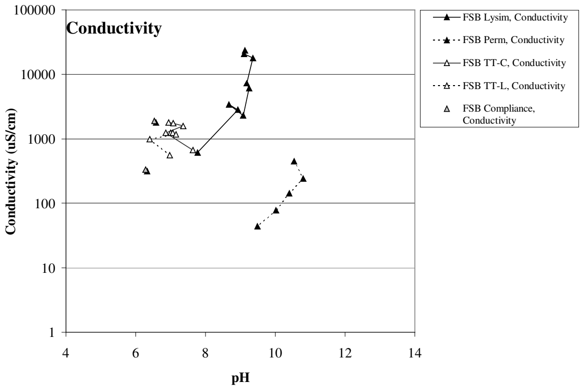


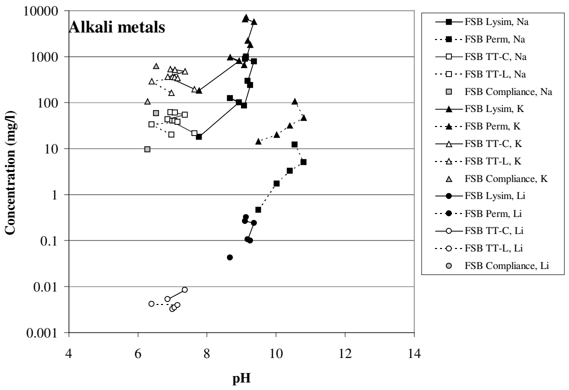


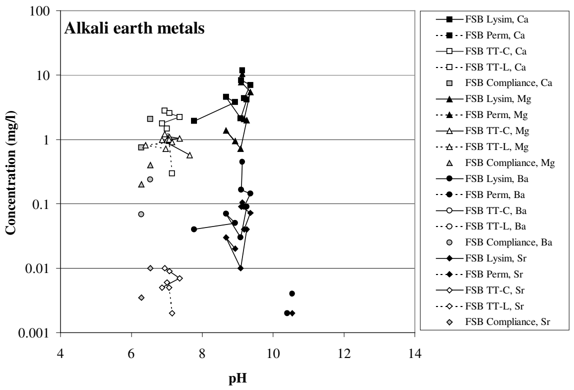


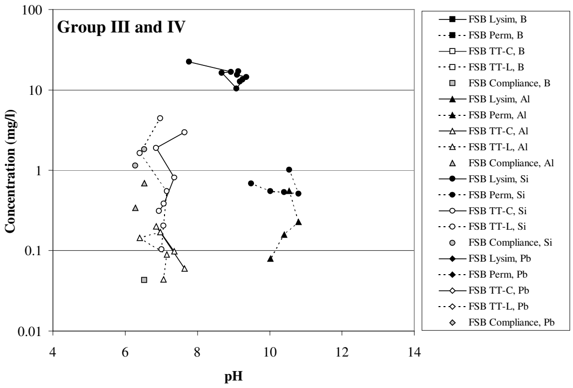
FOUNDRY SAND (PHENOLIC, BRASS)
Concentration in the leachate (mg/l) against pH

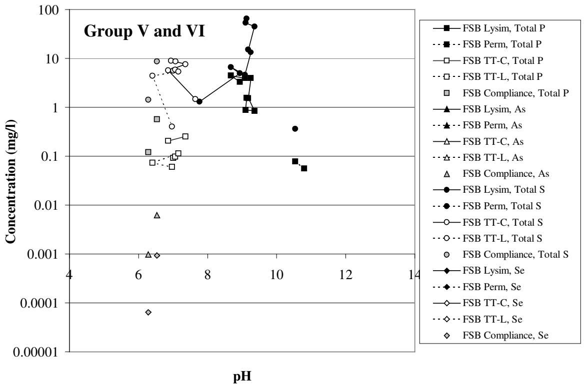


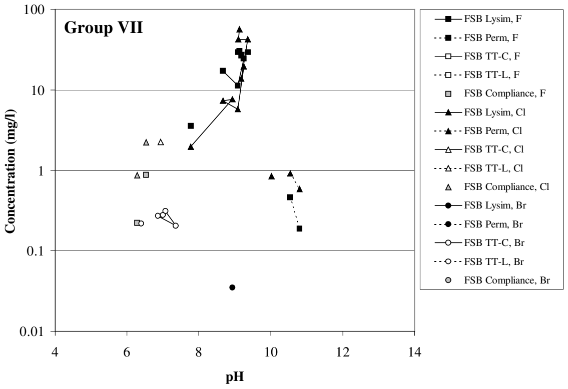


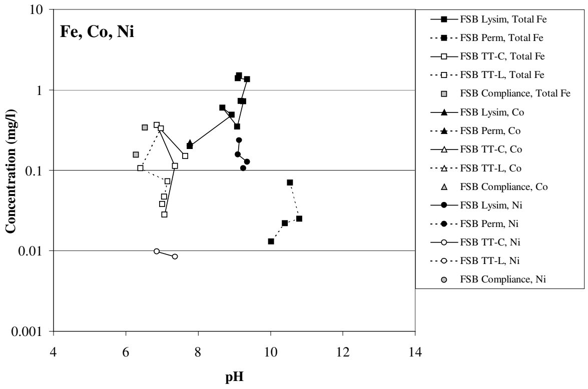




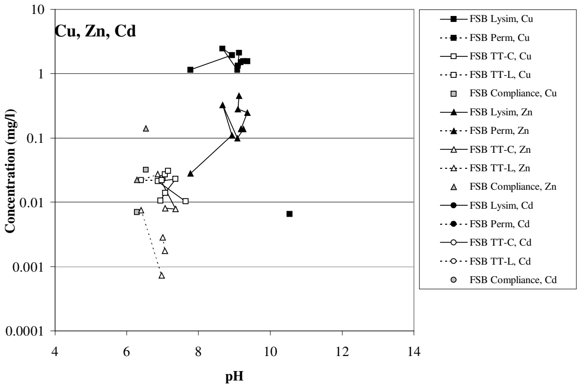


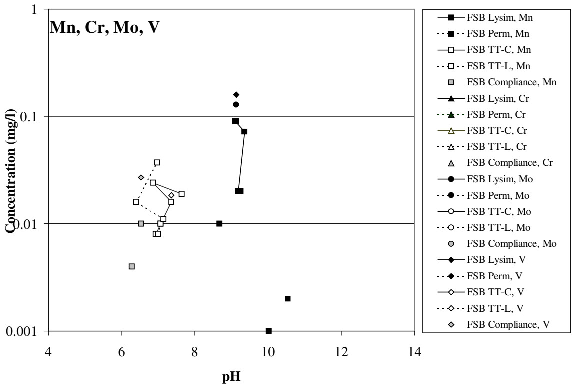


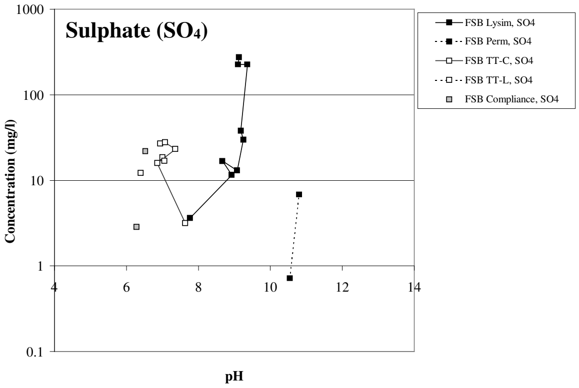


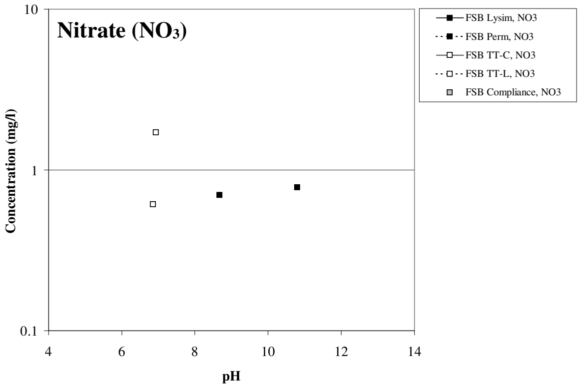


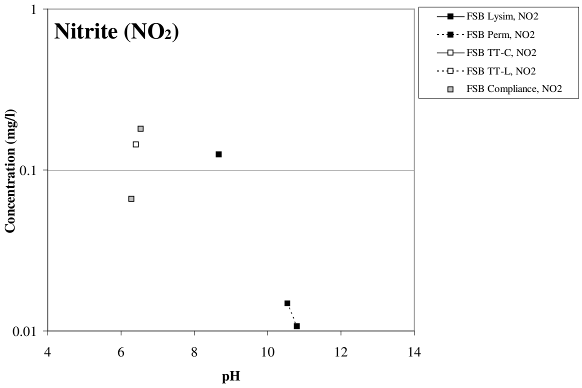
FOUNDRY SAND (PHENOLIC, BRASS)
Concentration in the leachate (mg/l) against pH

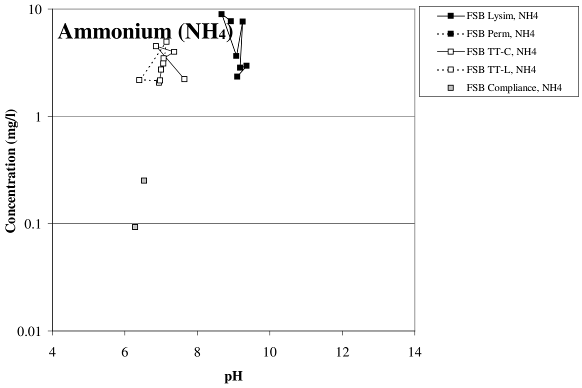




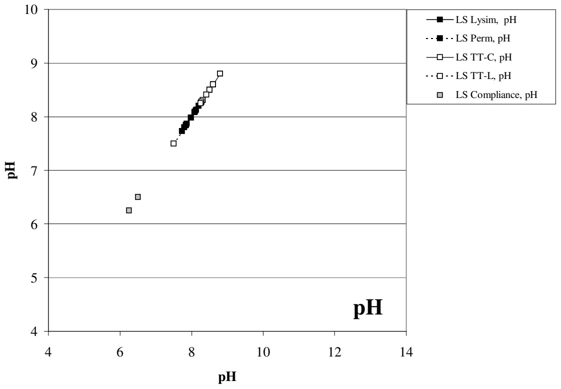


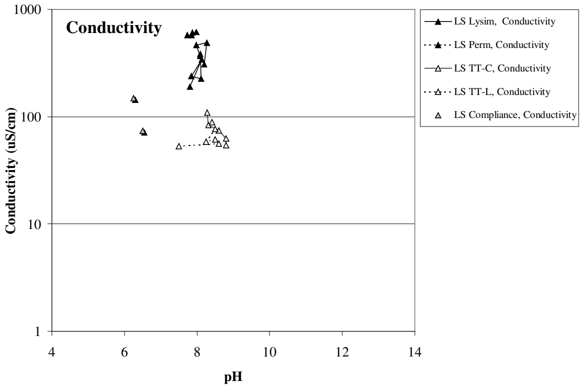


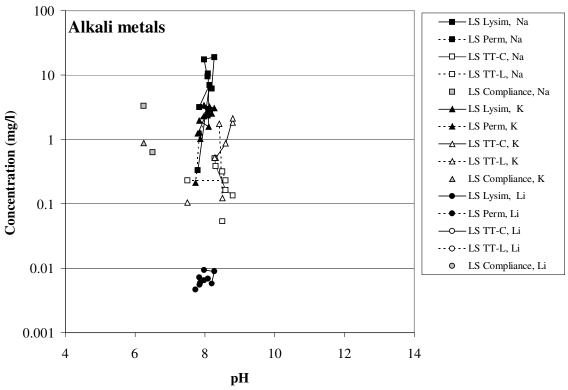


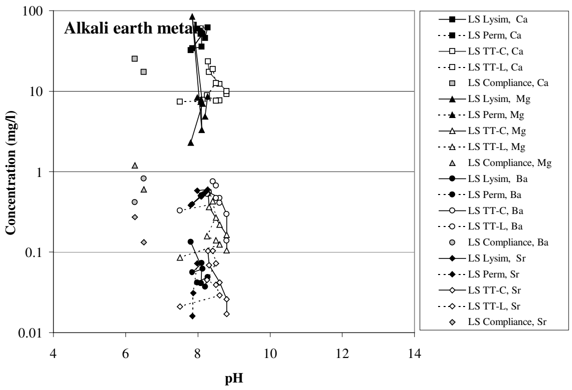


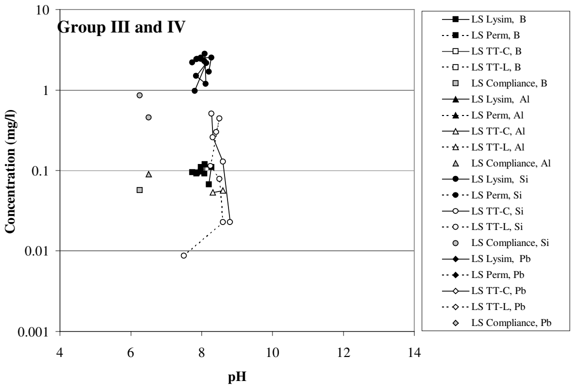
LIMESTONE
Concentration in the leachate (mg/l) against pH

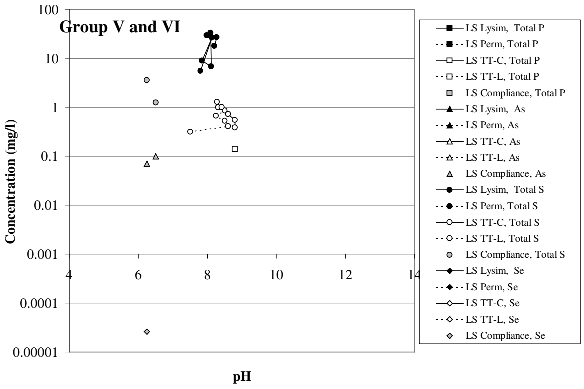


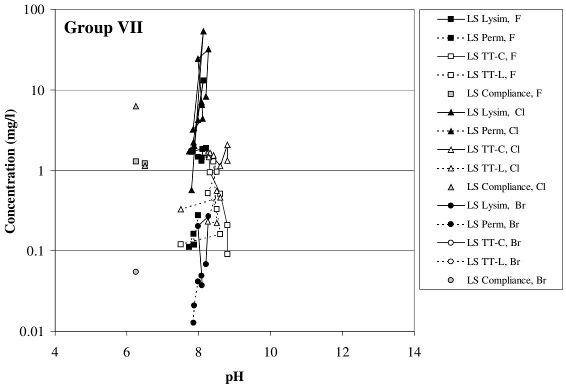


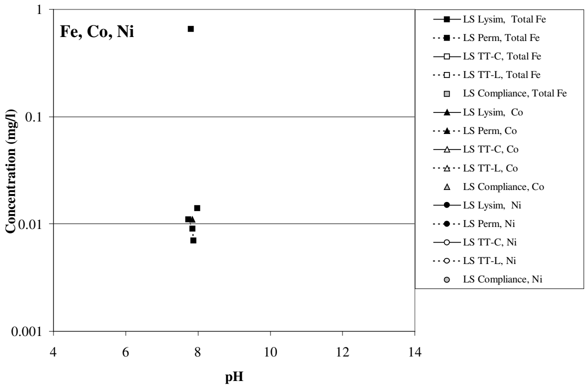




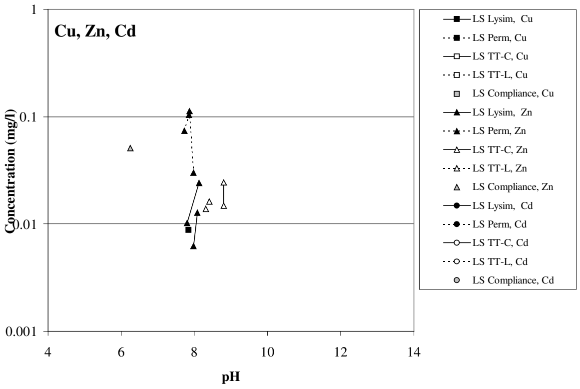


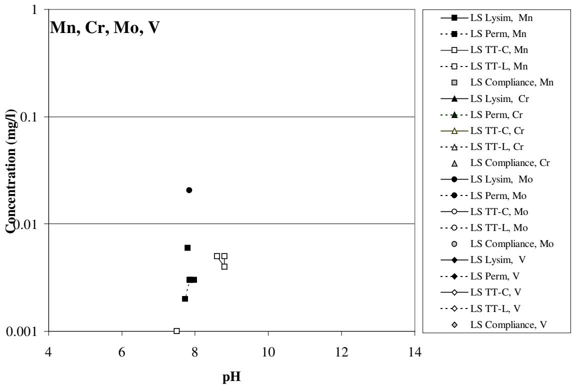


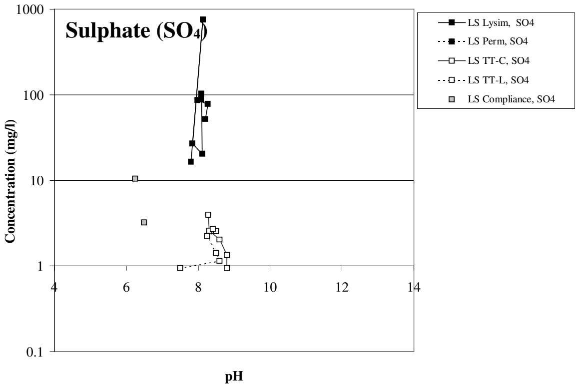


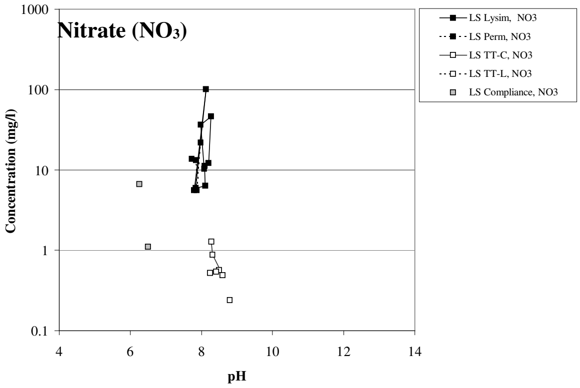


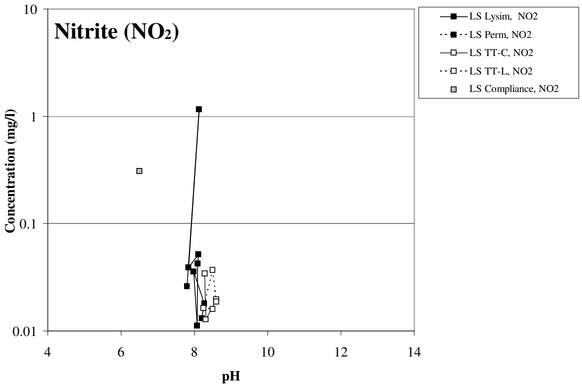
LIMESTONE
Concentration in the leachate (mg/l) against pH

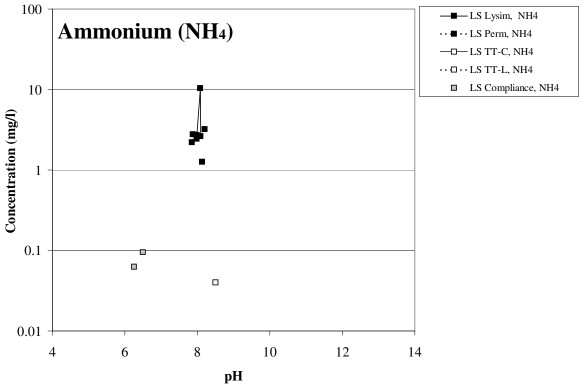




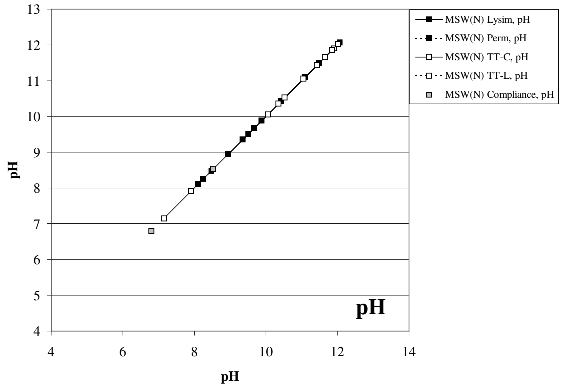


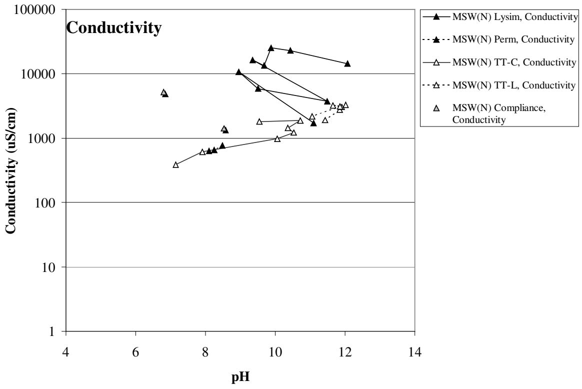


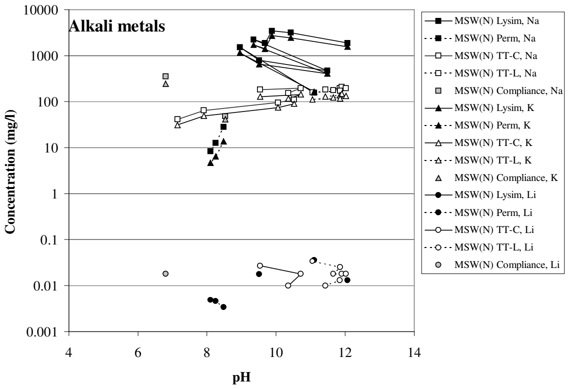


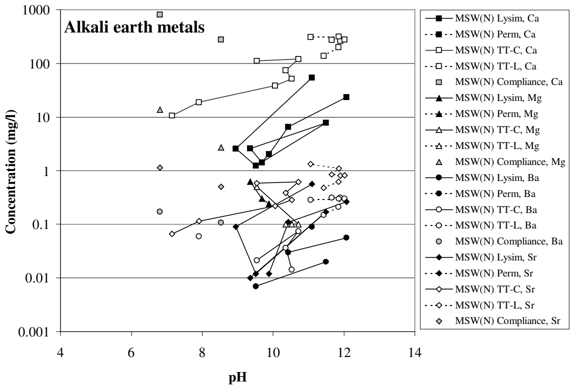


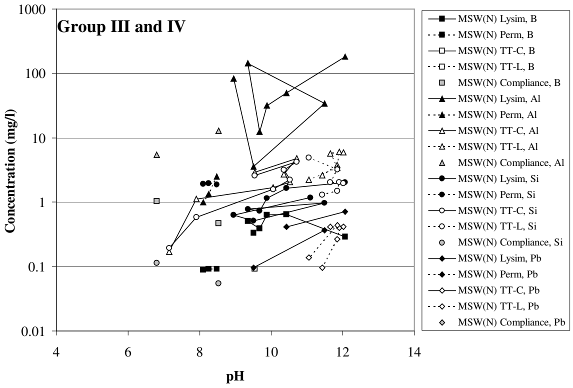
MSW INCINERATOR ASH
Concentration in the leachate (mg/l) against pH

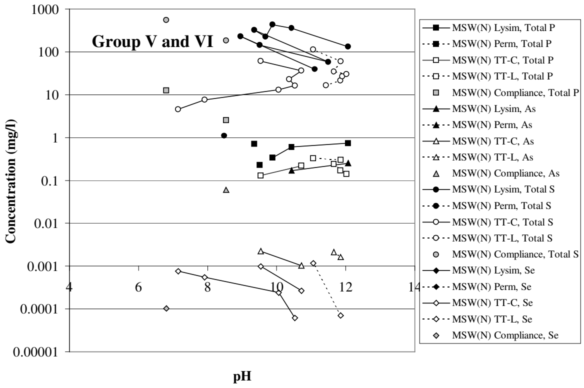


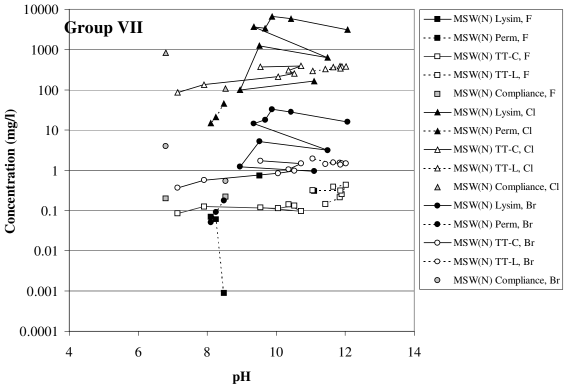


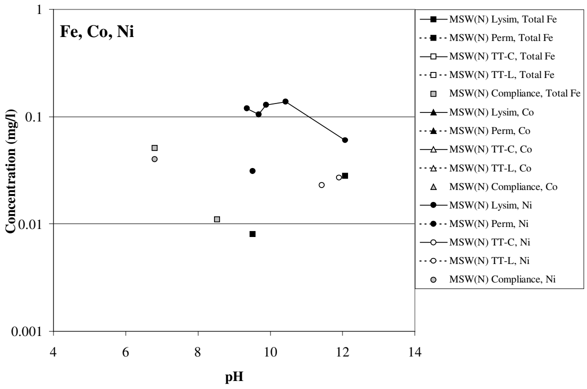




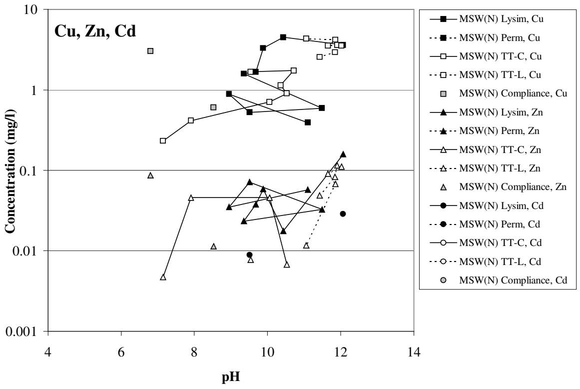


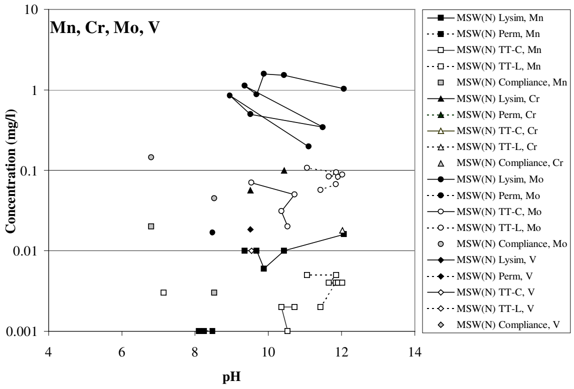


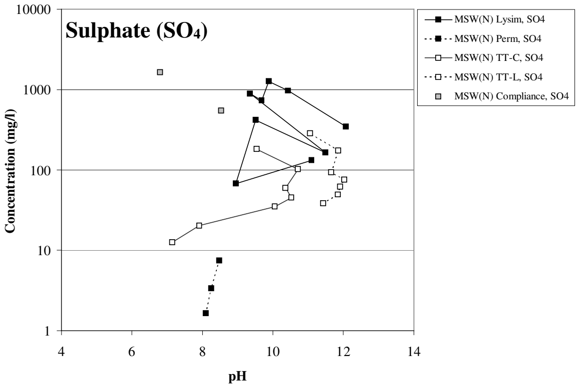


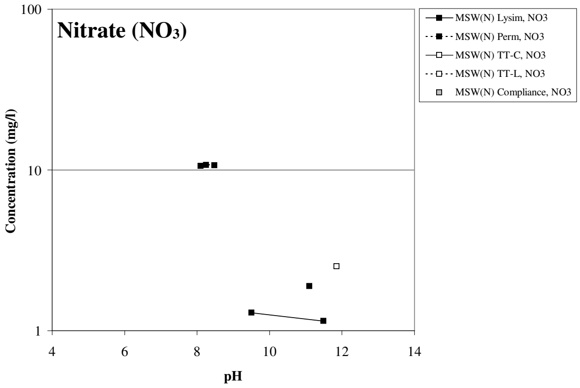


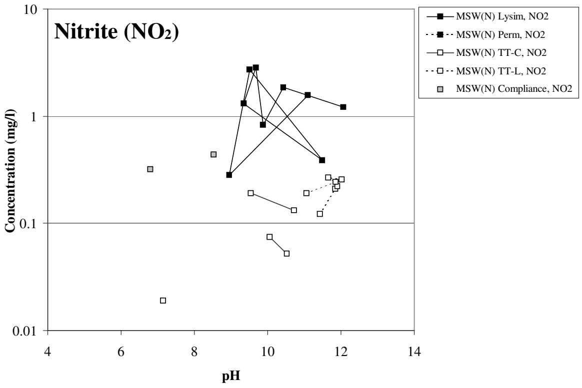
MSW INCINERATOR ASH
Concentration in the leachate (mg/l) against pH

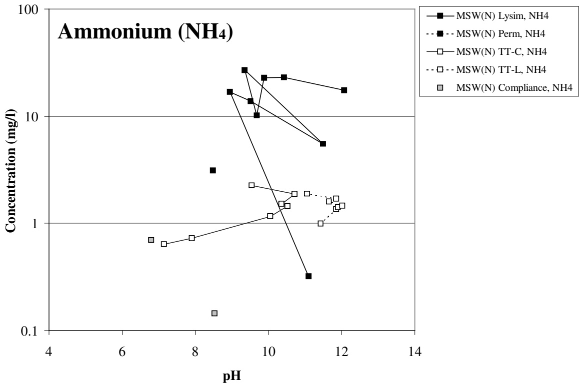




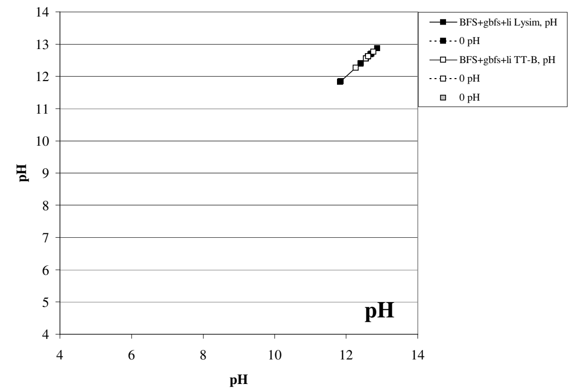


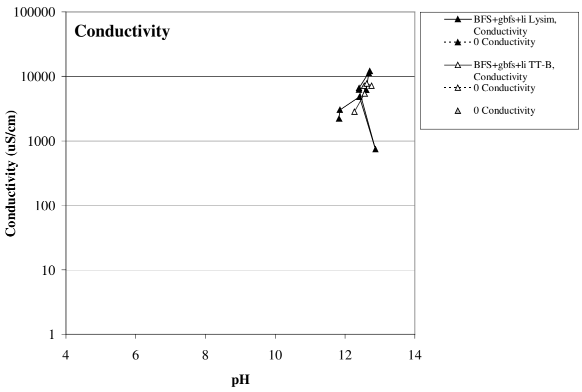


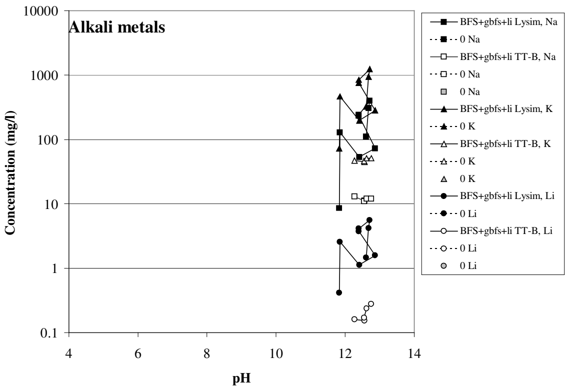


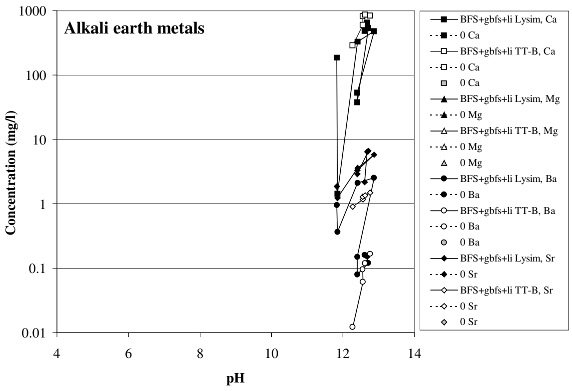


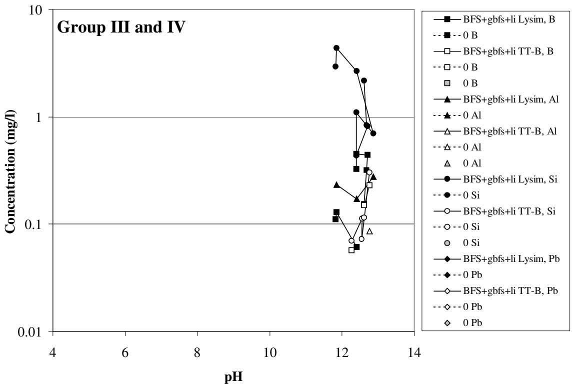
BLASTFURNACE SLAG + granulated blastfurnace slag + lime
Concentration in the leachate (mg/l) against pH

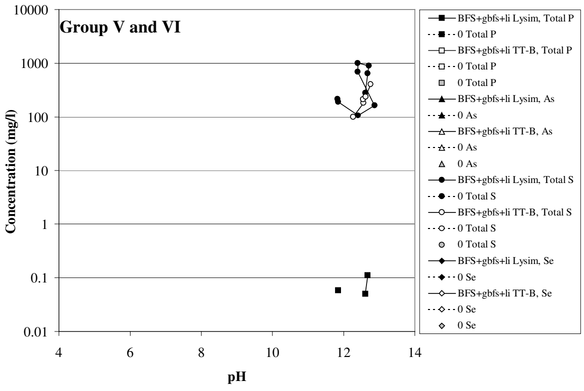


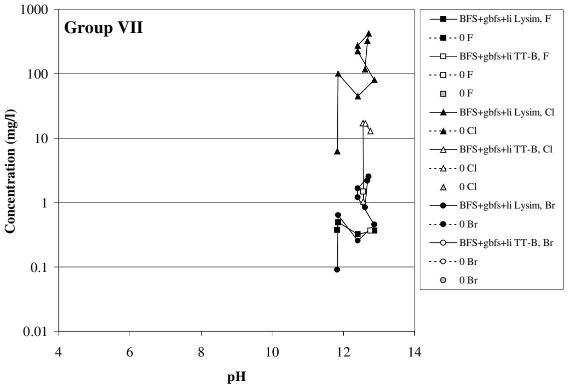


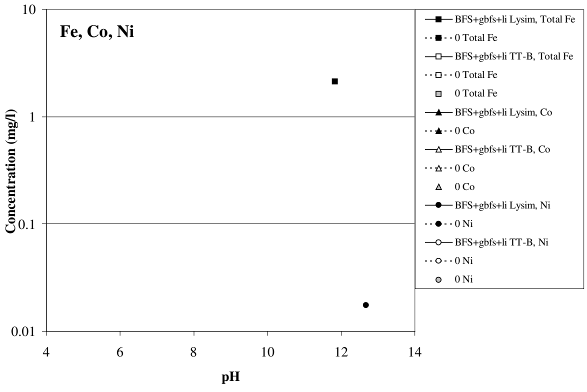




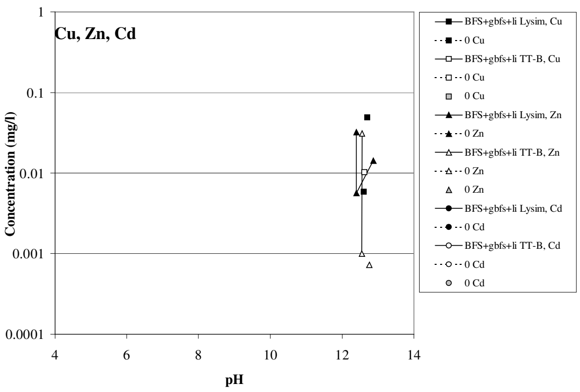


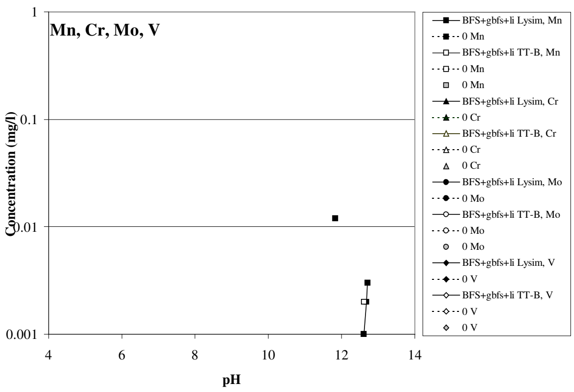


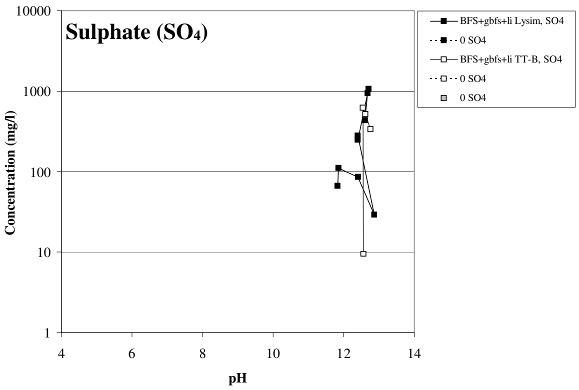


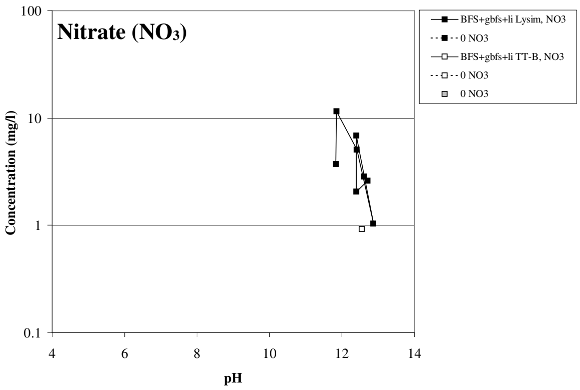


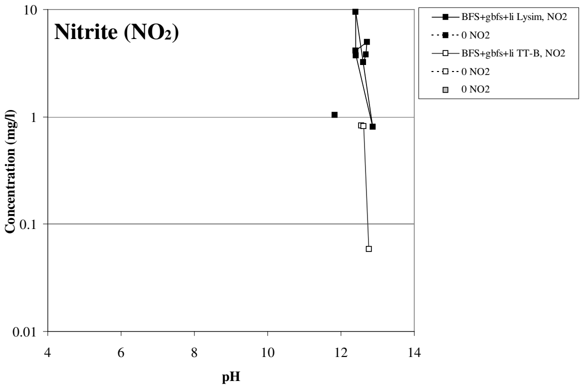
BLASTFURNACE SLAG + granulated blastfurnace slag + lime
Concentration in the leachate (mg/l) against pH

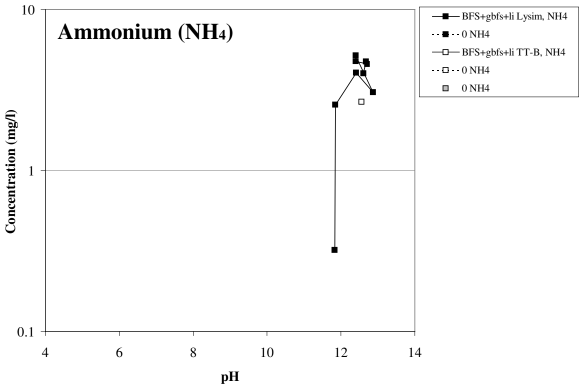




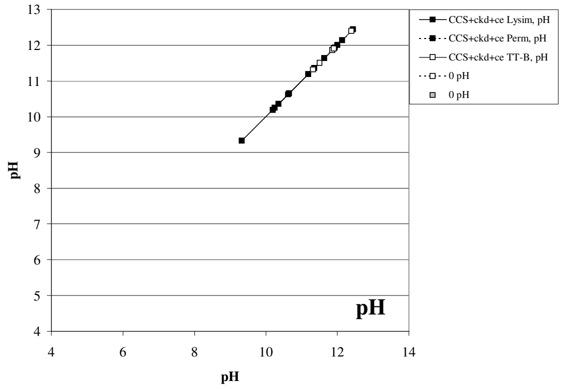


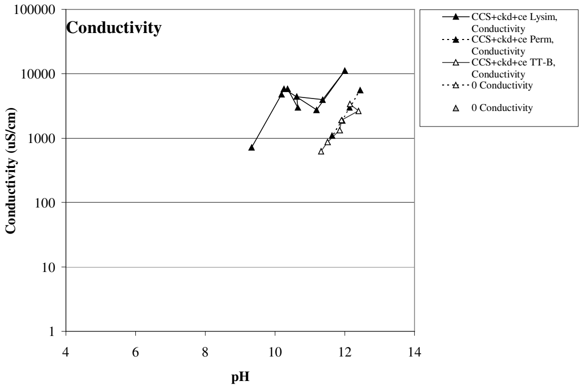


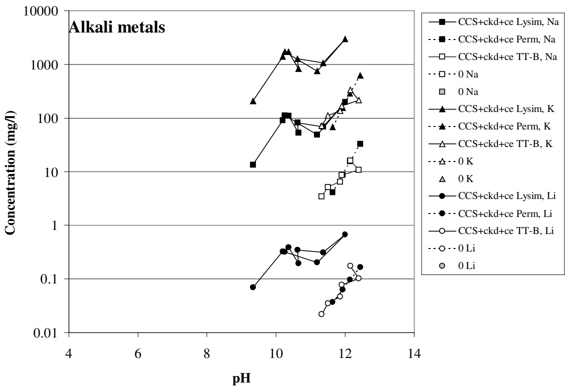


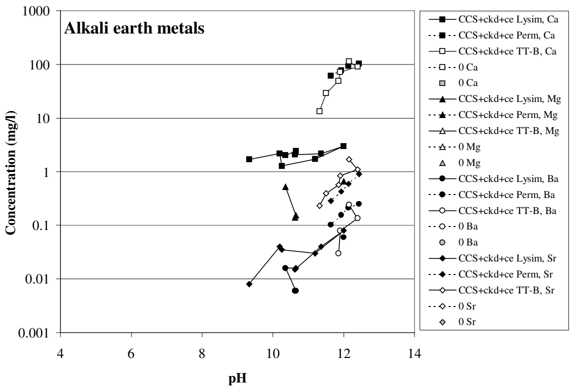


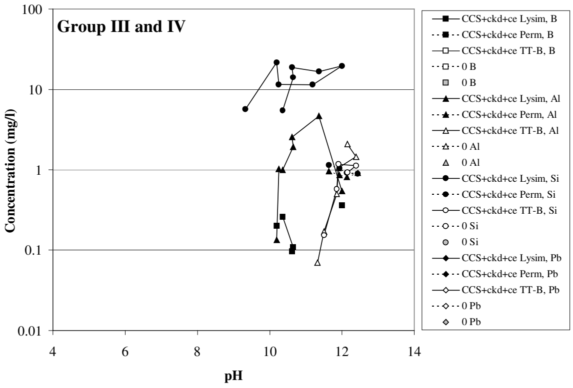
CHINA CLAY SAND + cement kiln dust + cement
Concentration in the leachate (mg/l) against pH

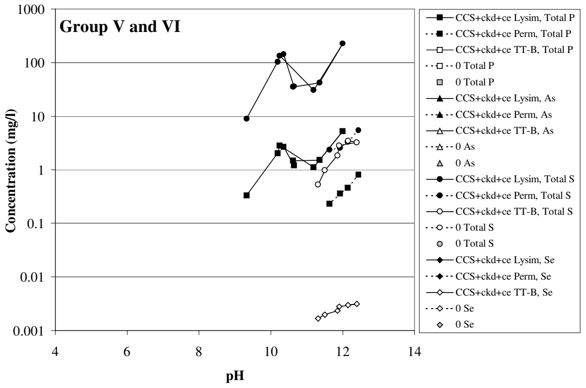


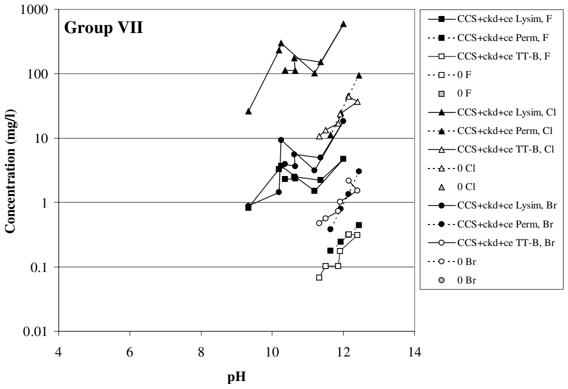


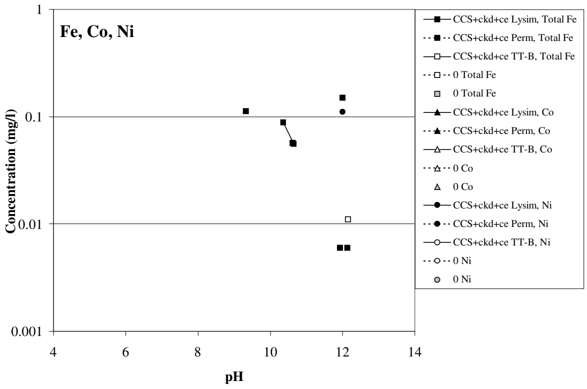




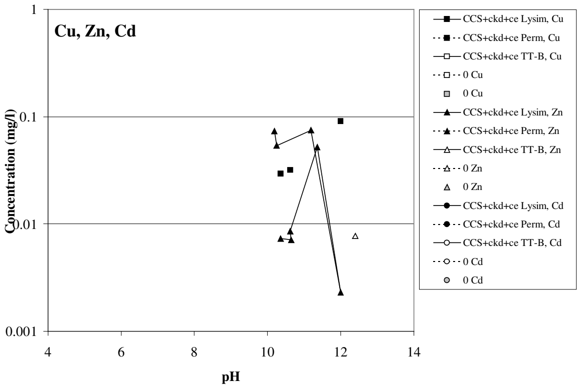


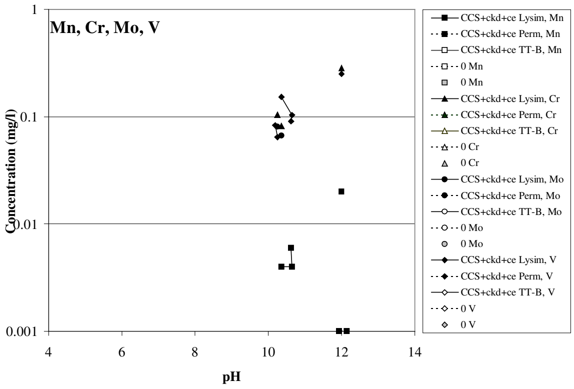


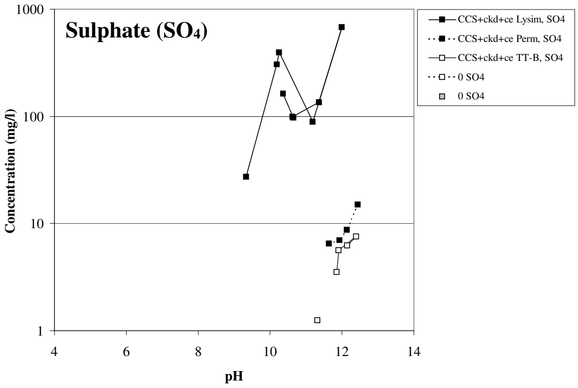


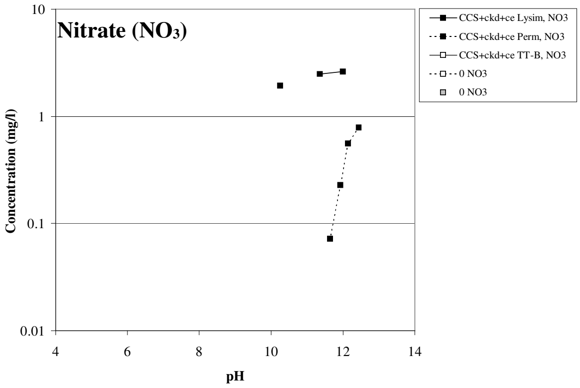


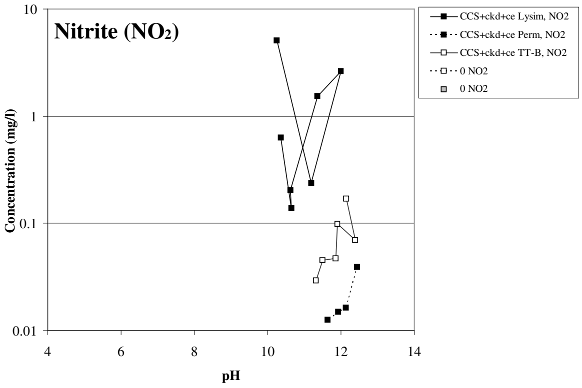
CHINA CLAY SAND + cement kiln dust + cement
Concentration in the leachate (mg/l) against pH

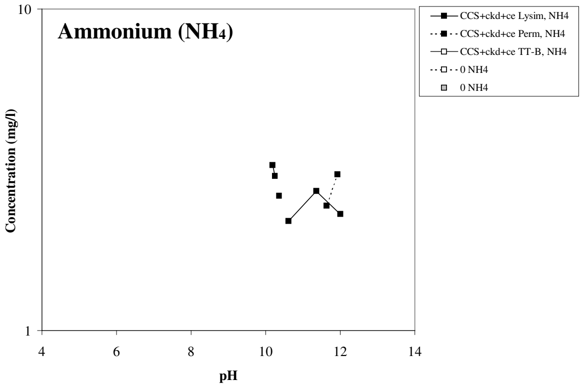




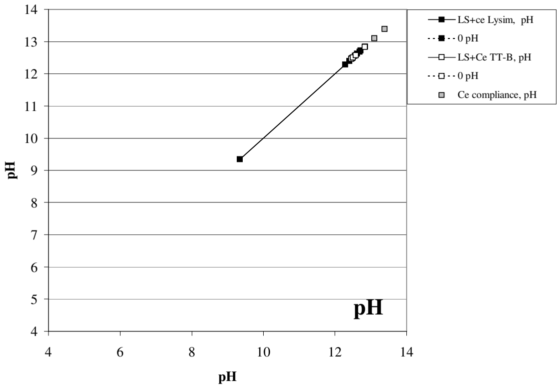


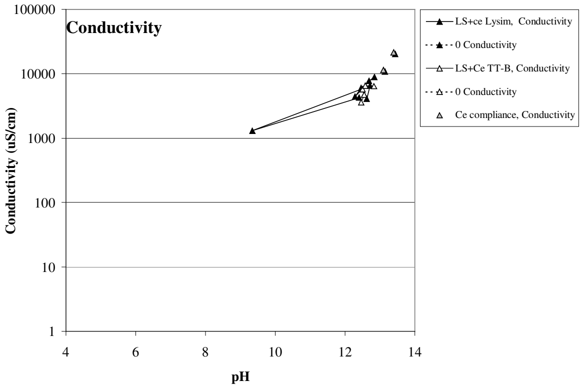


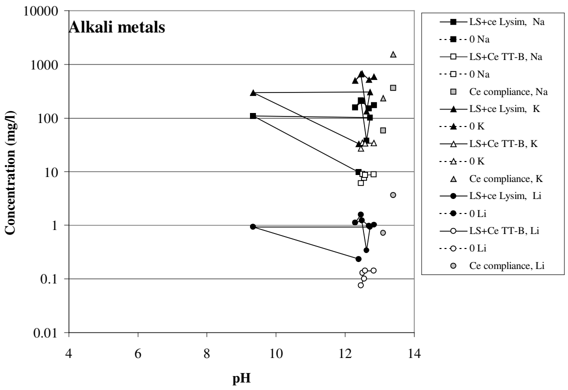


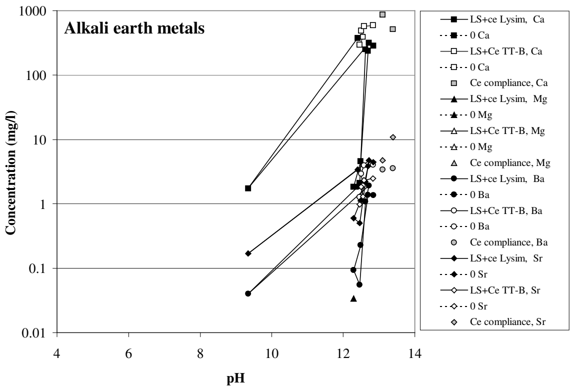


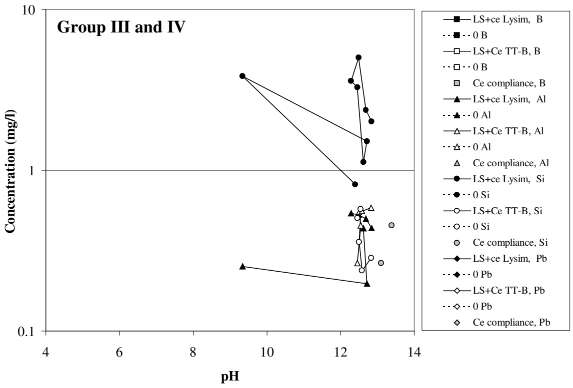
LIMESTONE + cement
Concentration in the leachate (mg/l) against pH

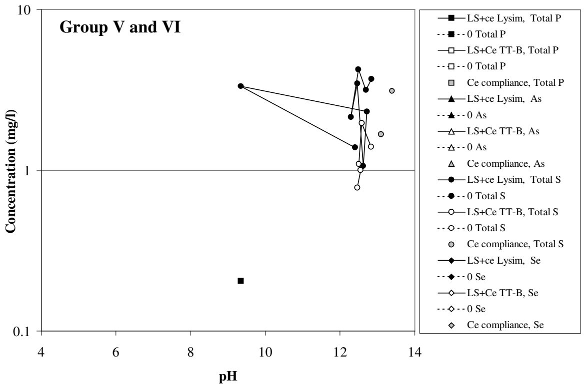


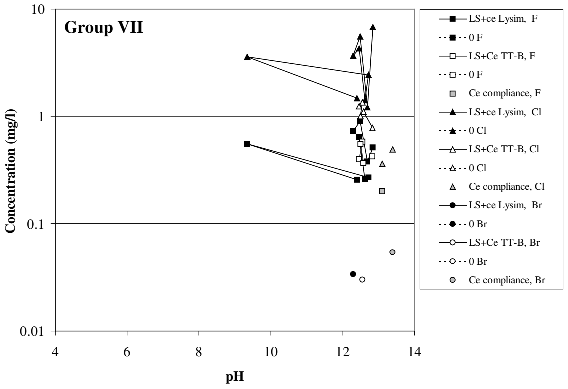


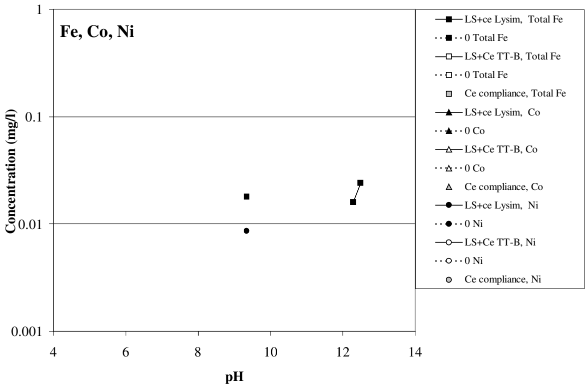




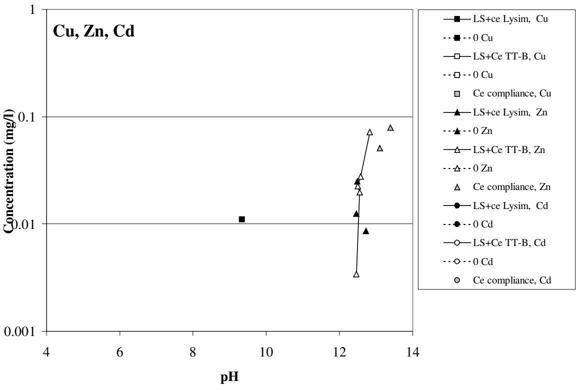


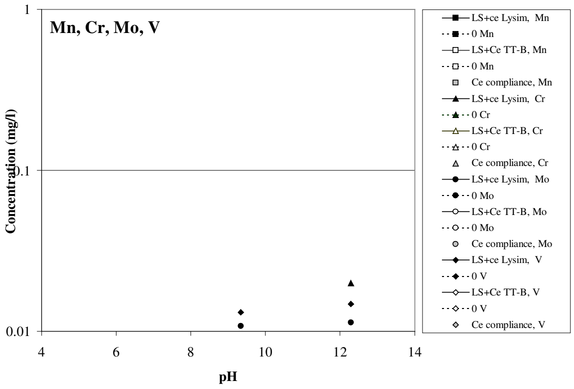


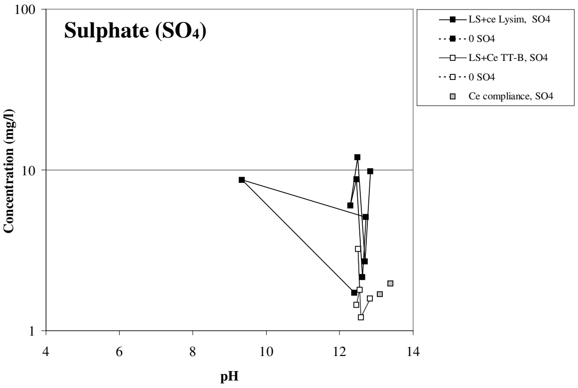


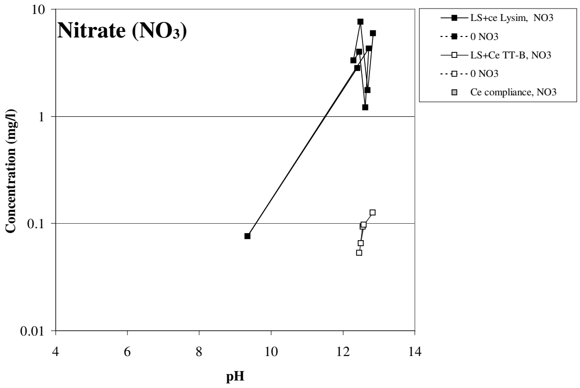


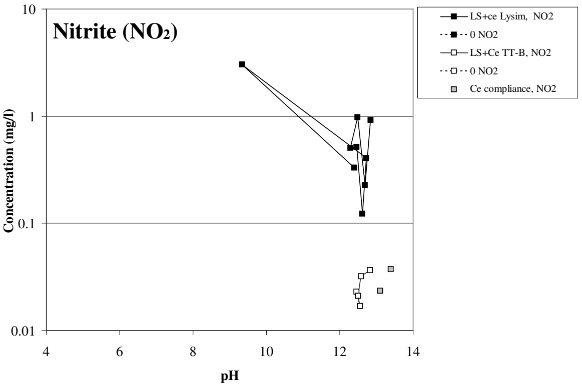
LIMESTONE + cement
Concentration in the leachate (mg/l) against pH

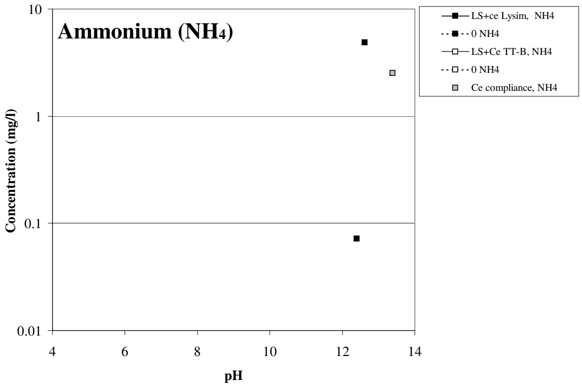




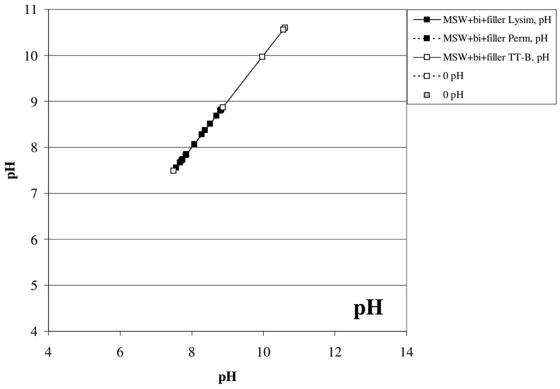


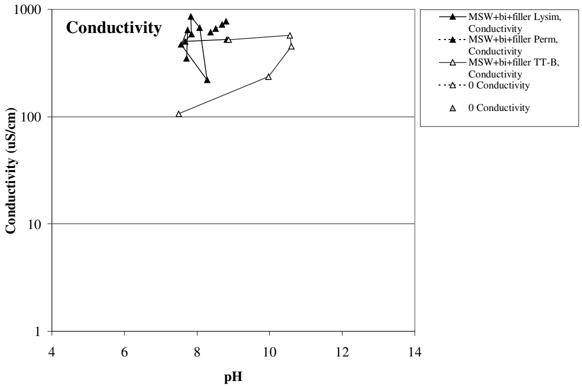


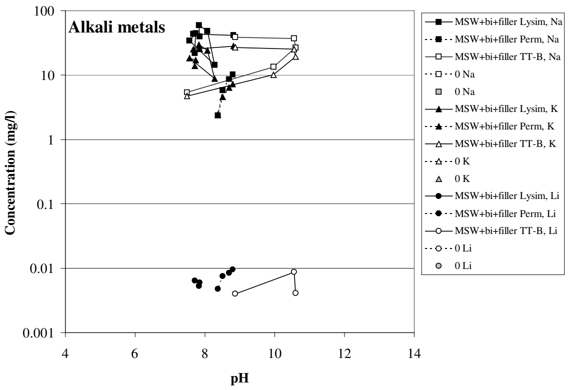


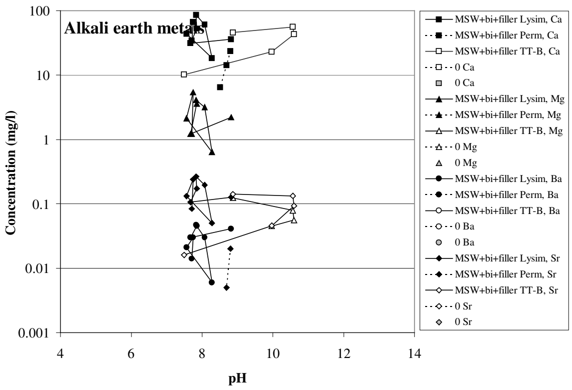


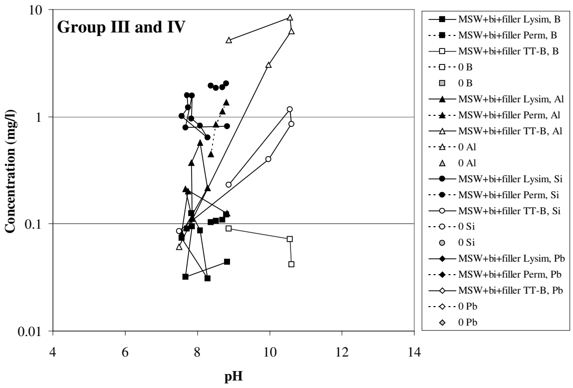
MSW INCINERATOR ASH +bitumen + filler
Concentration in the leachate (mg/l) against pH

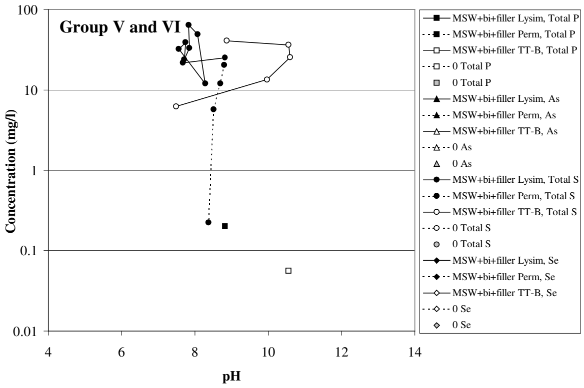


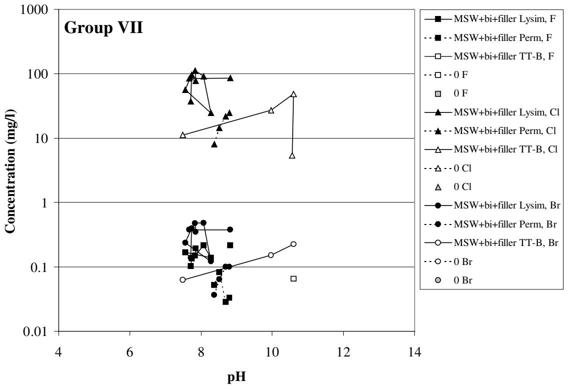


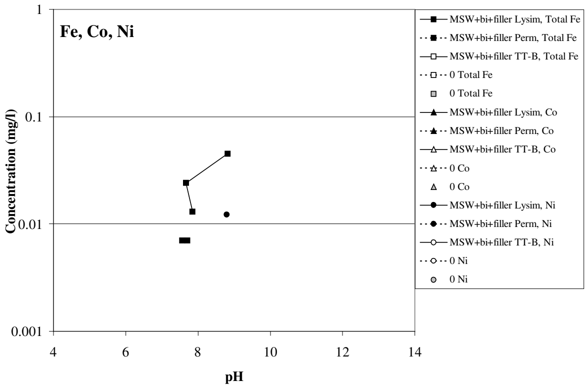




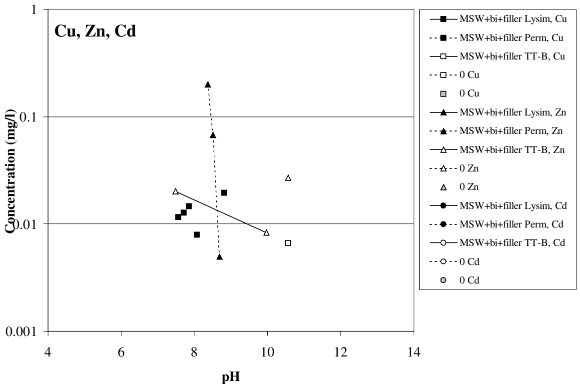


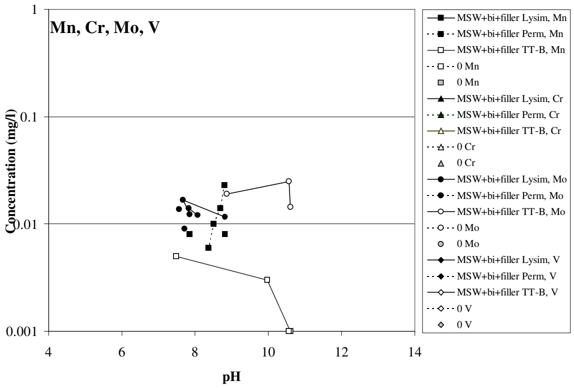


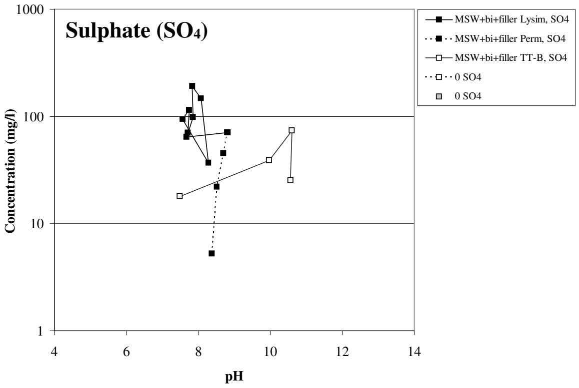


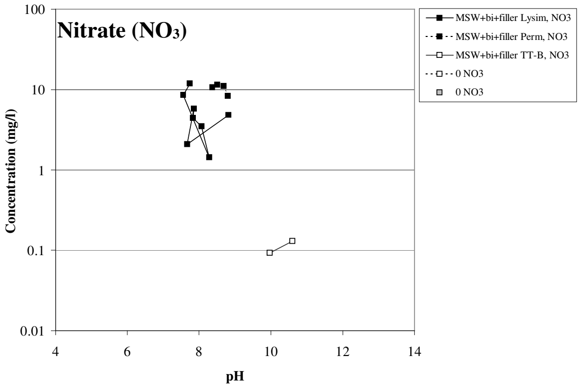


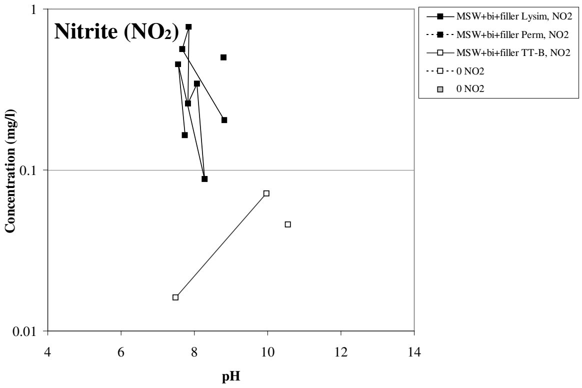
MSW INCINERATOR ASH +bitumen + filler
Concentration in the leachate (mg/l) against pH

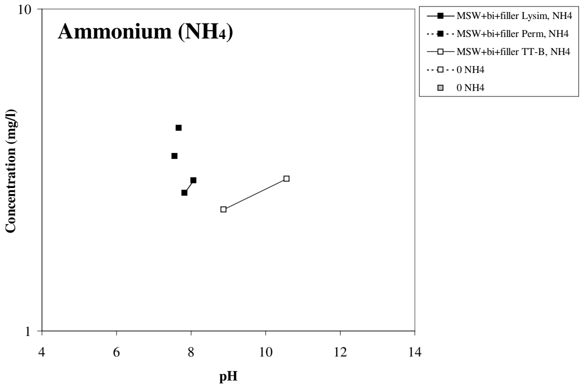










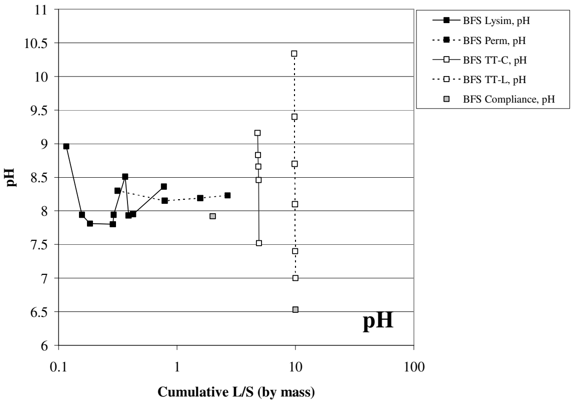


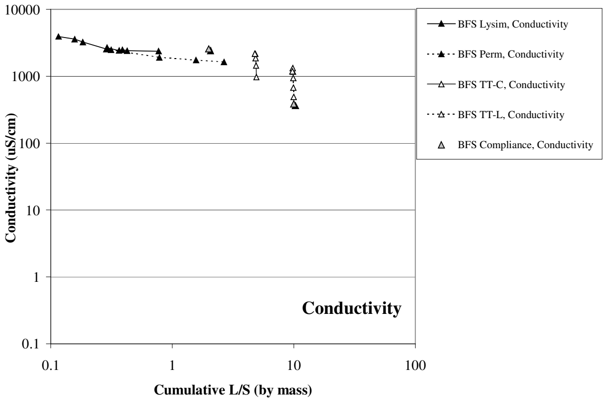
APPENDIX D:

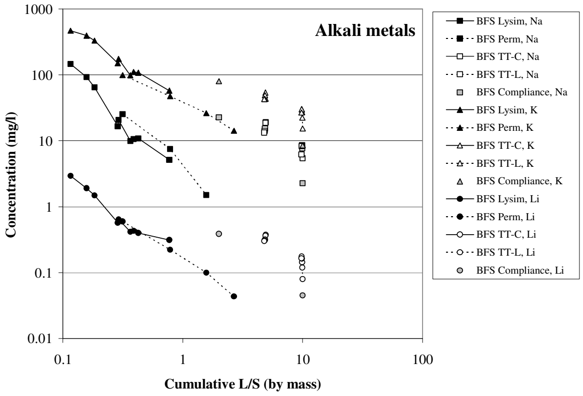
Graphs comparing laboratory leaching tests and lysimeters

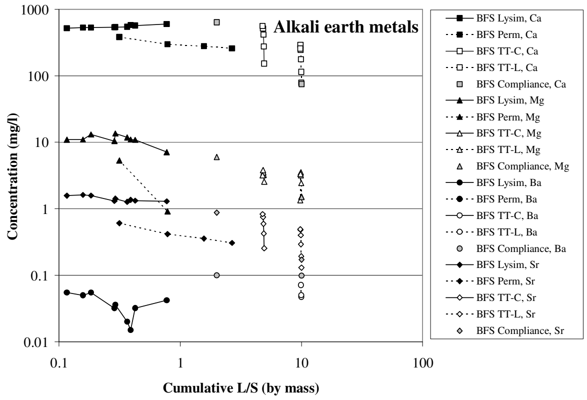
L/S ratio verses leached species concentration (mg/l)

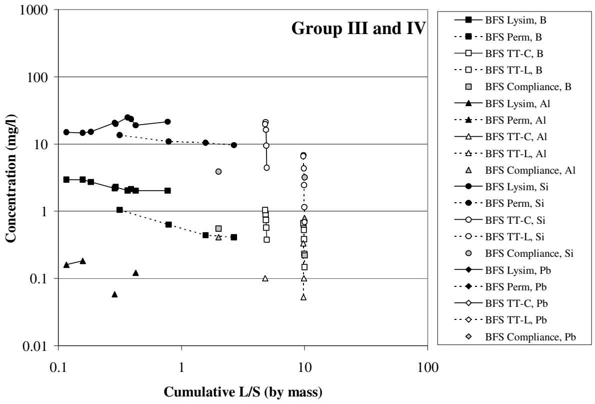
BLASTFURNACE SLAG
Concentrations in the leachate (mg/l) against cumulative L/S ratio

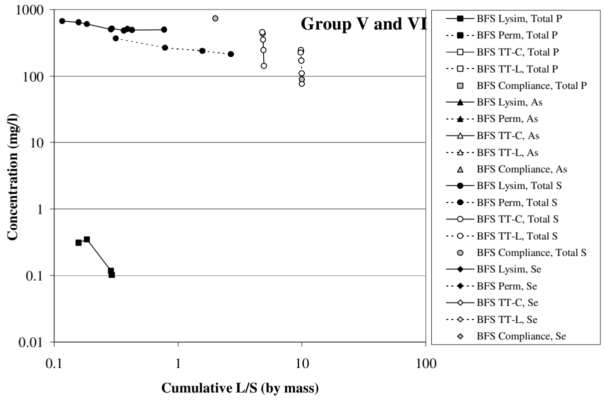


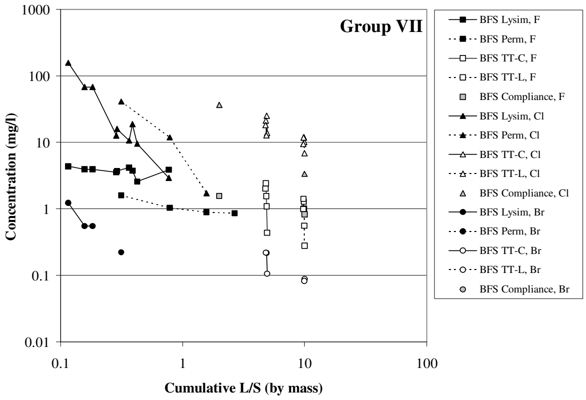


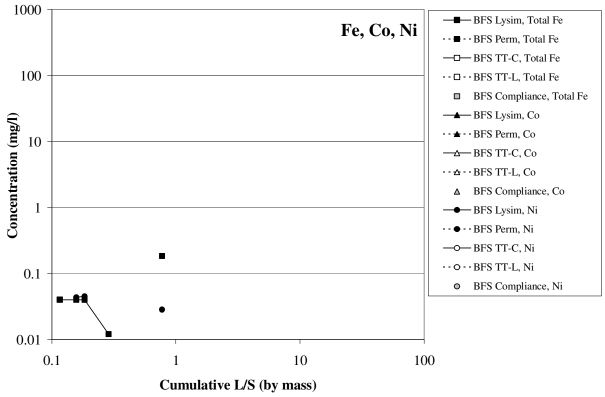




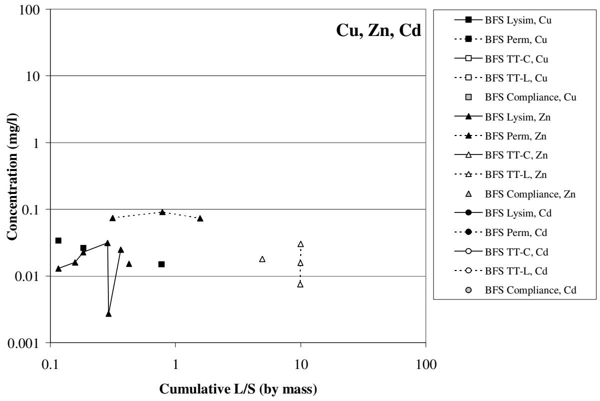


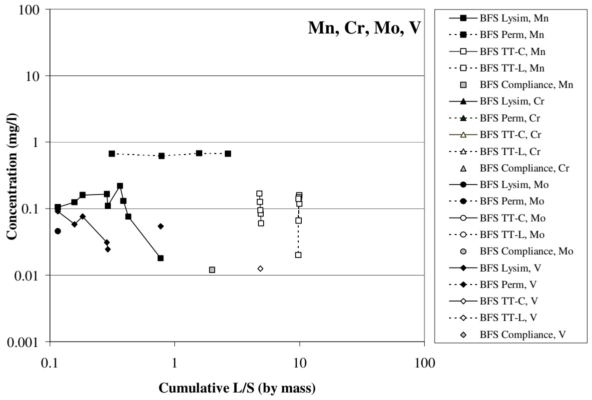


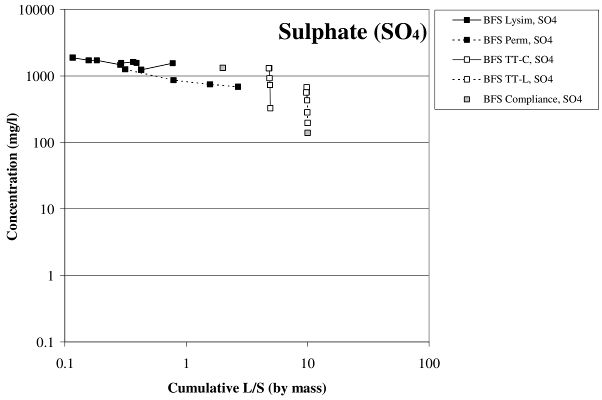


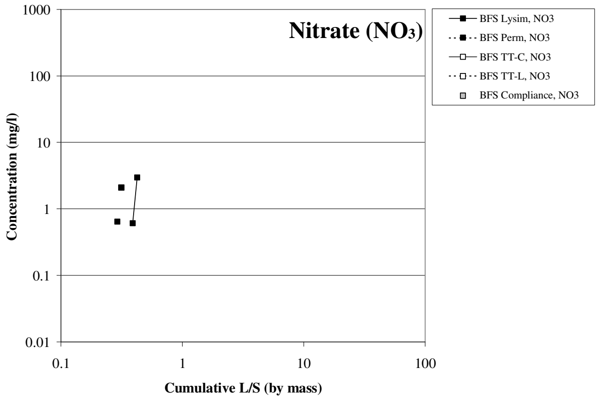


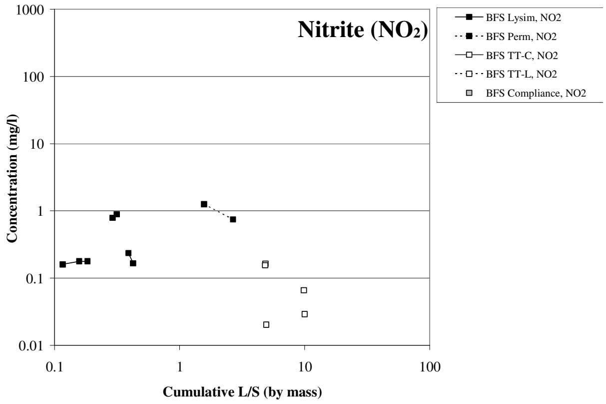
BLASTFURNACE SLAG
Concentrations in the leachate (mg/l) against cumulative L/S ratio

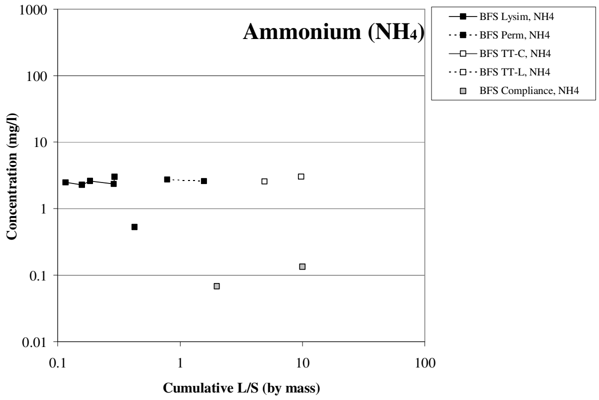


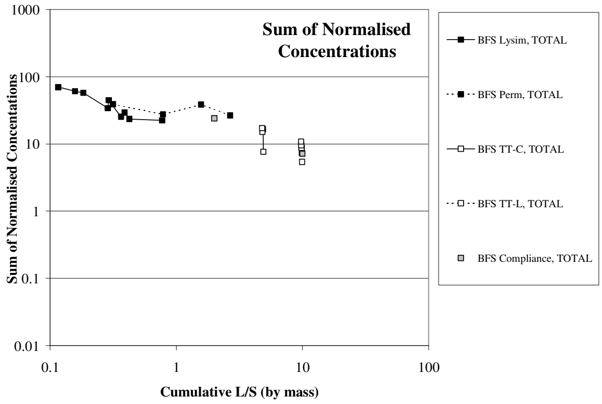


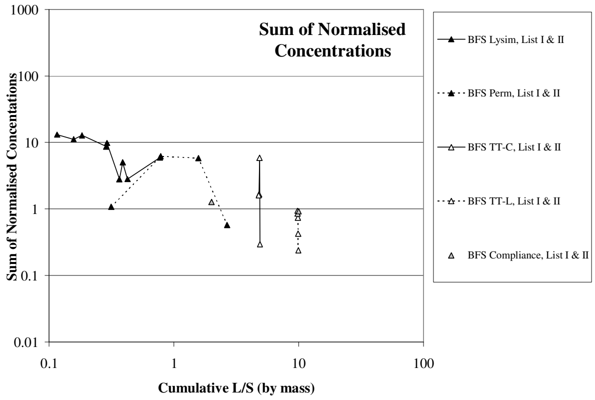




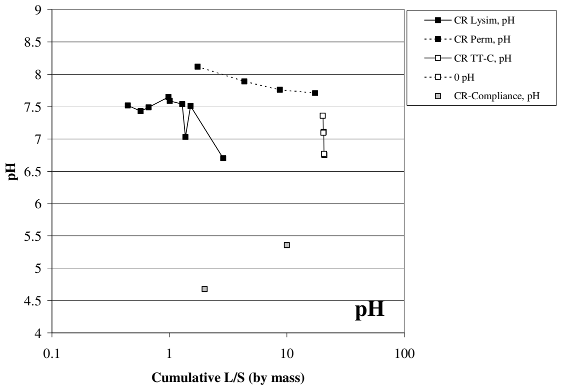


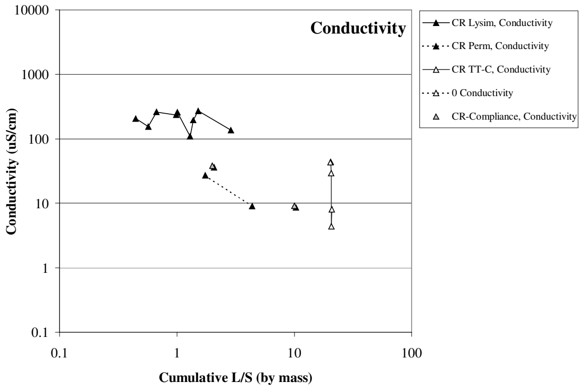


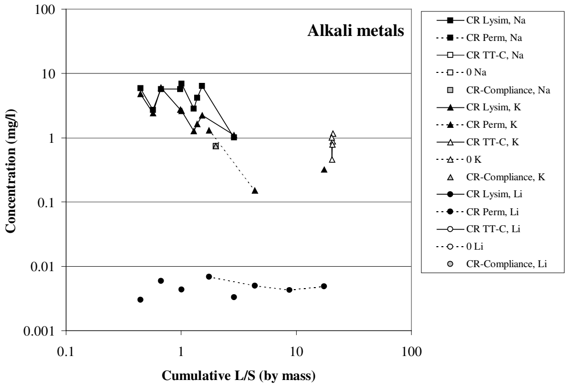


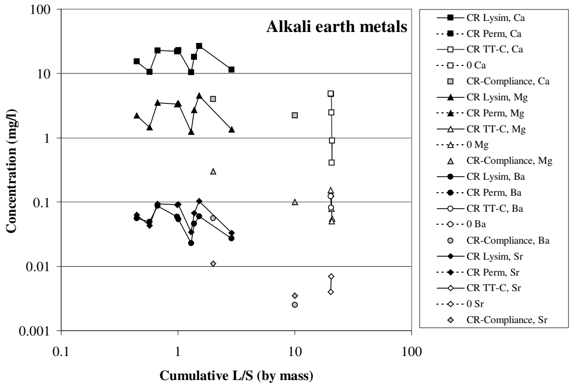


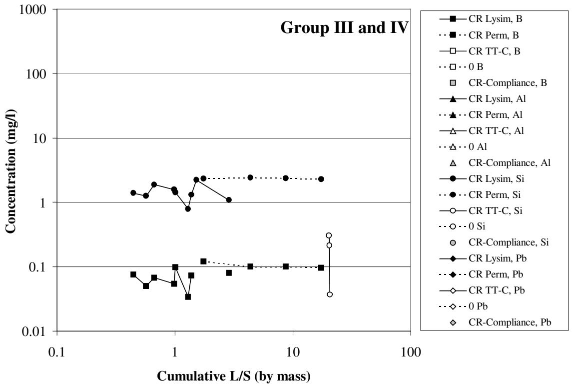
CRUMBED RUBBER (2-8mm)
Concentrations in the leachate (mg/l) against cumulative L/S ratio

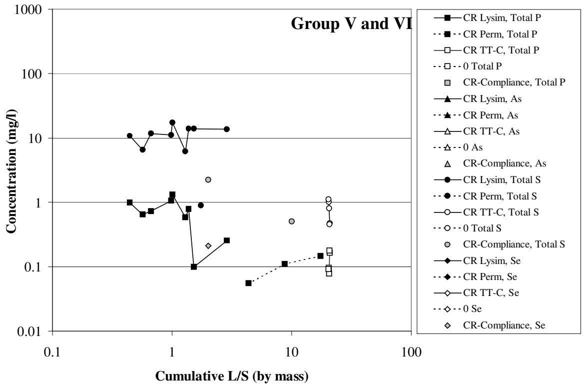


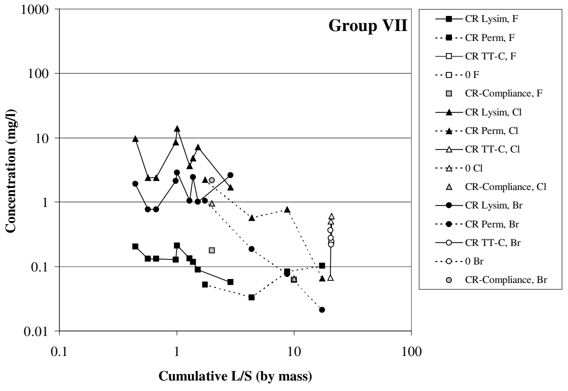


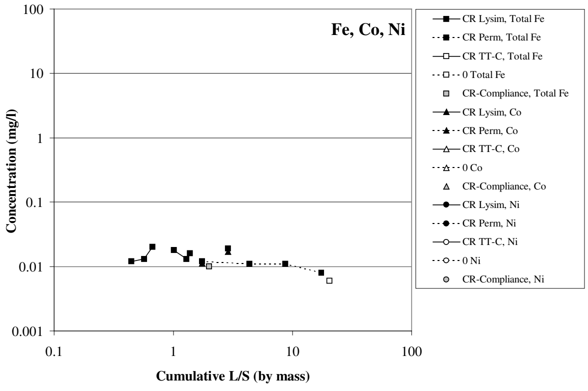




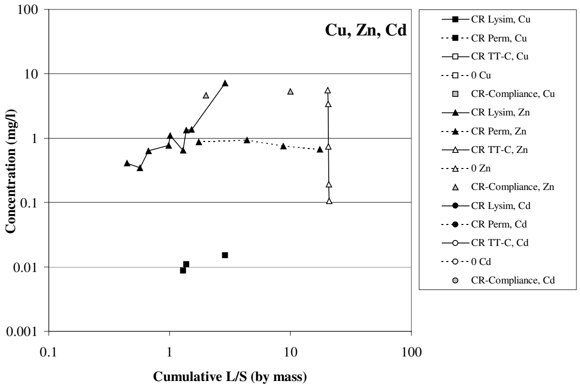


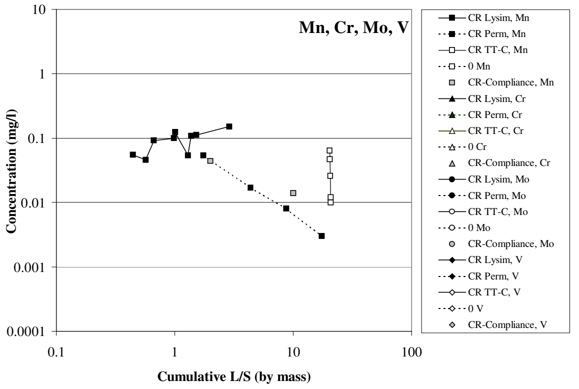


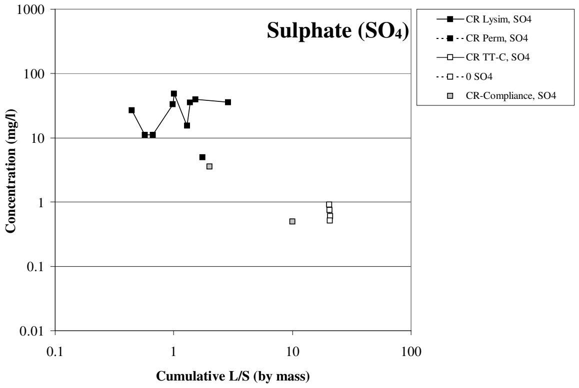


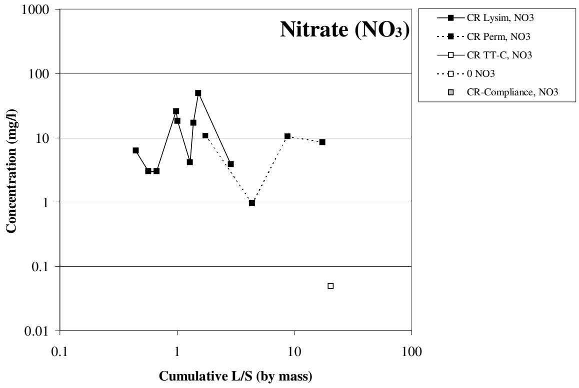


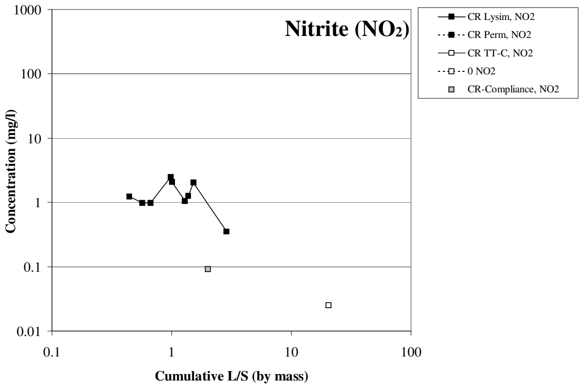
CRUMBED RUBBER (2-8mm)
Concentrations in the leachate (mg/l) against cumulative L/S ratio

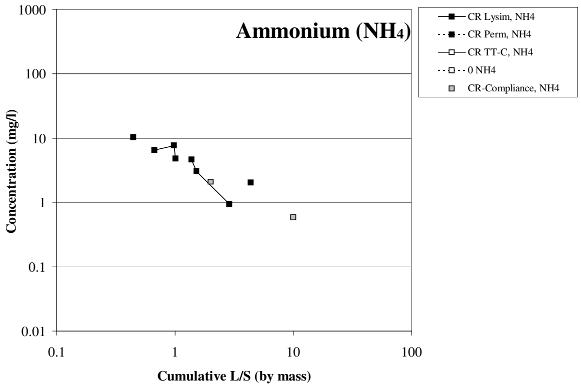


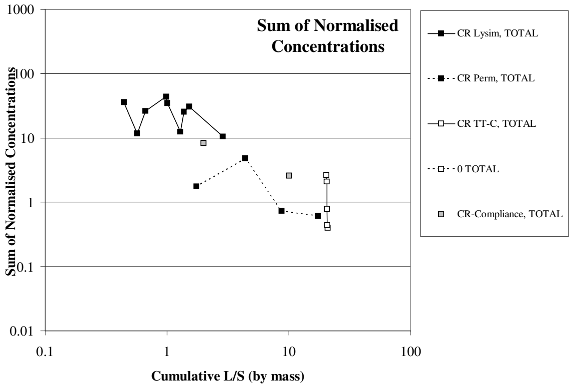


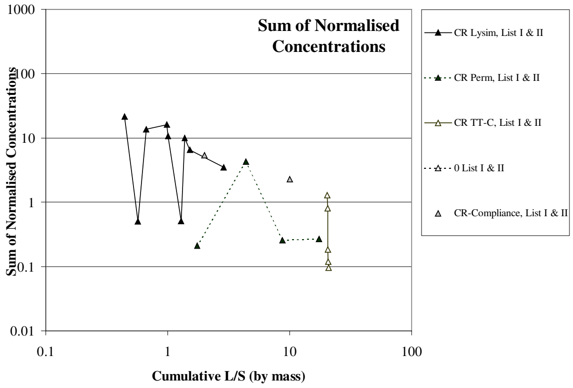




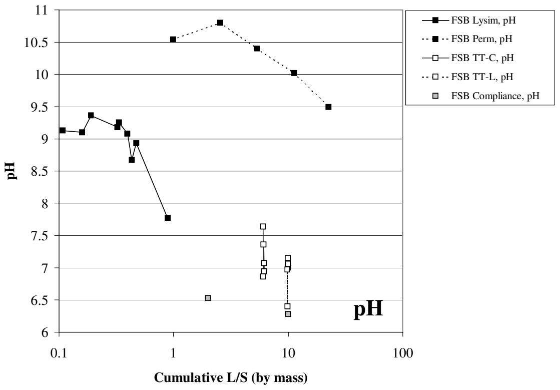


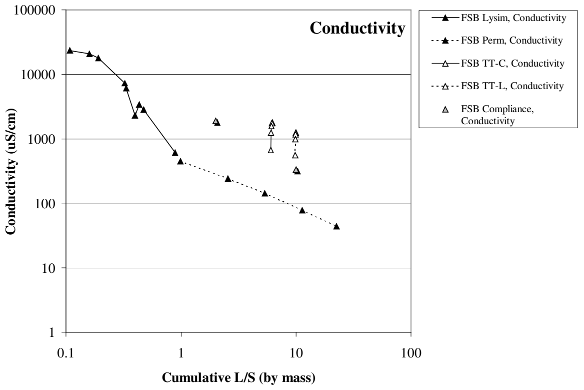


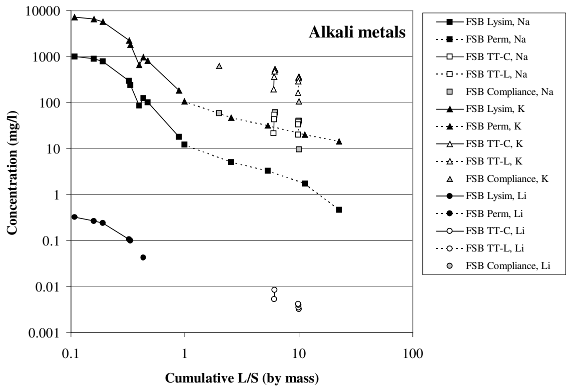


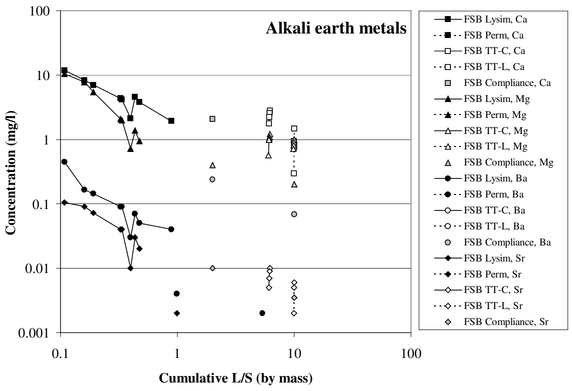


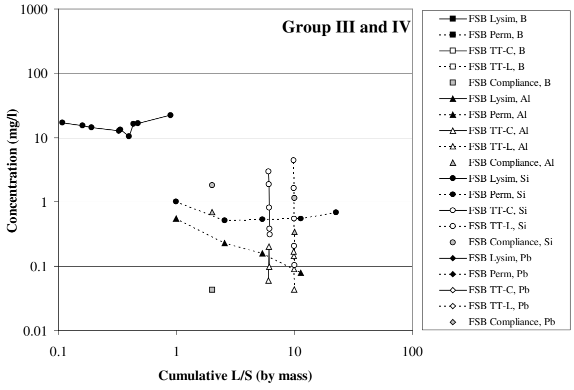
FOUNDRY SAND (PHENOLIC, BRASS)
Concentrations in the leachate (mg/l) against cumulative L/S ratio

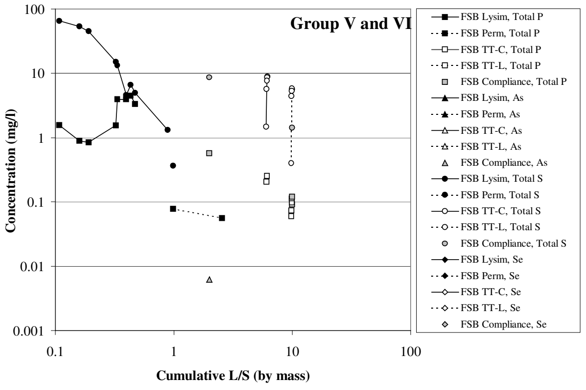


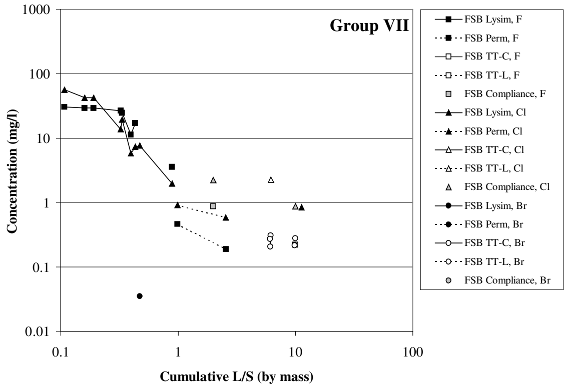


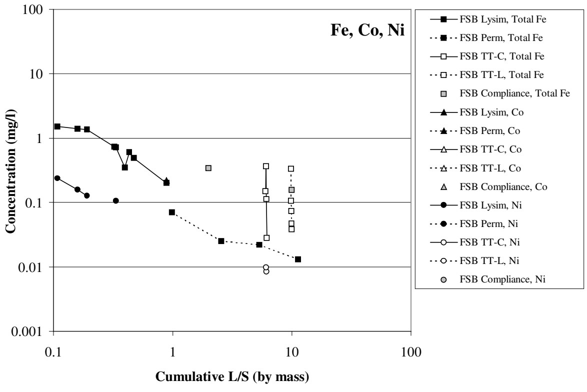




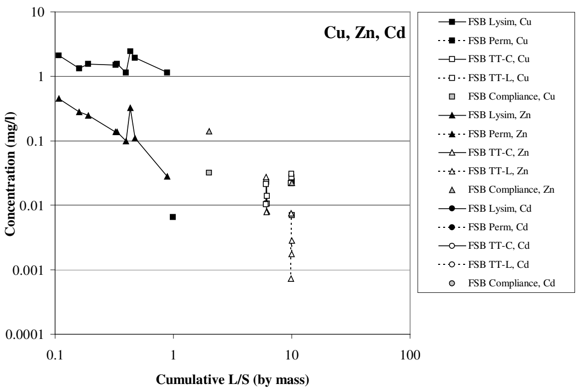


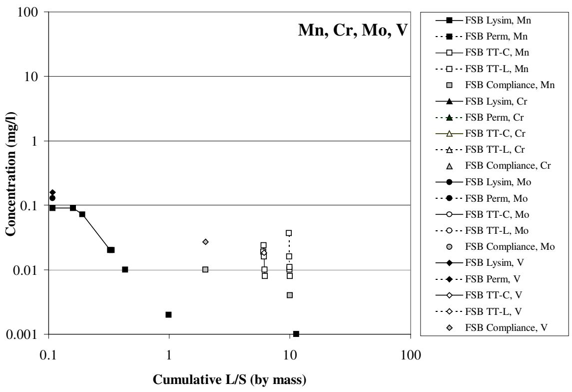


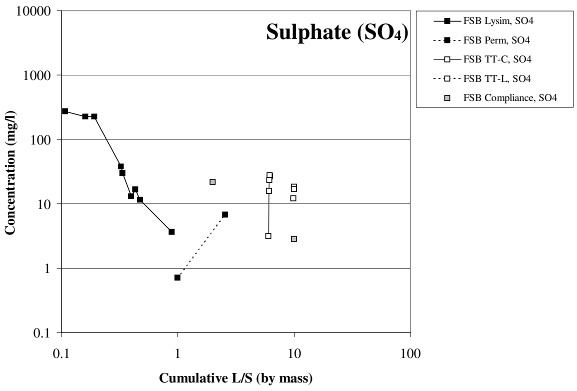


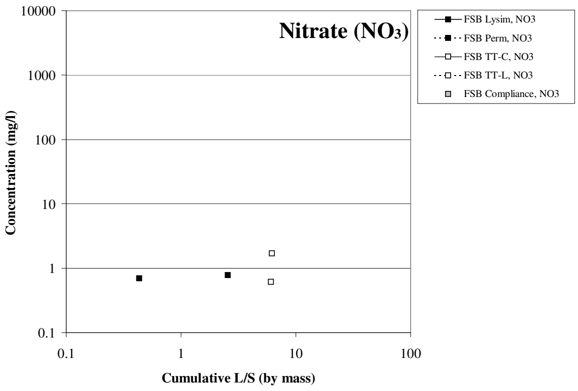


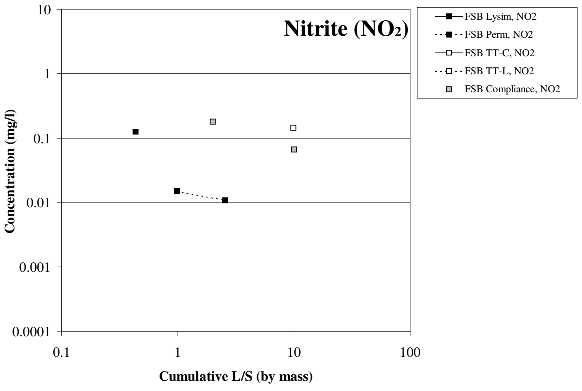
FOUNDRY SAND (PHENOLIC, BRASS)
Concentrations in the leachate (mg/l) against cumulative L/S ratio

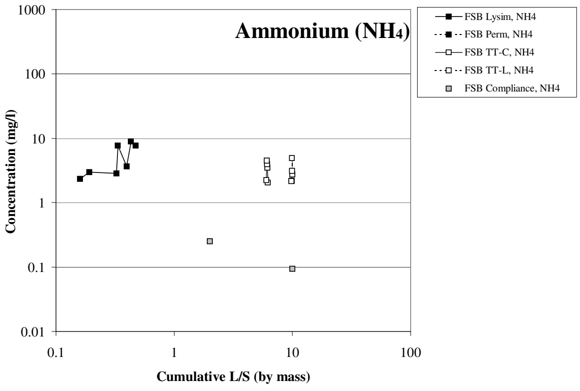


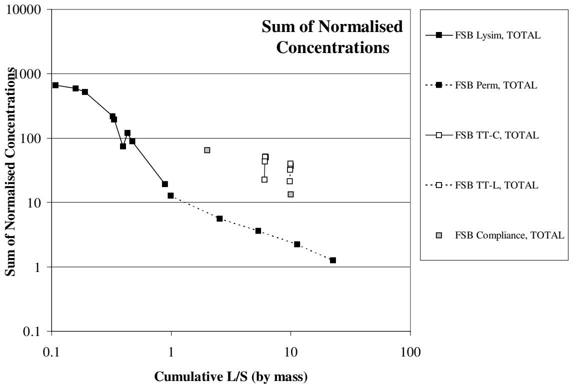


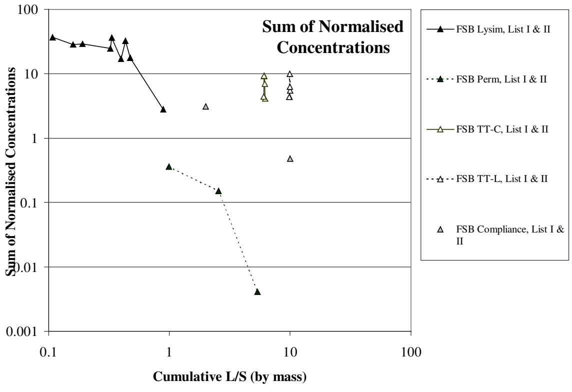




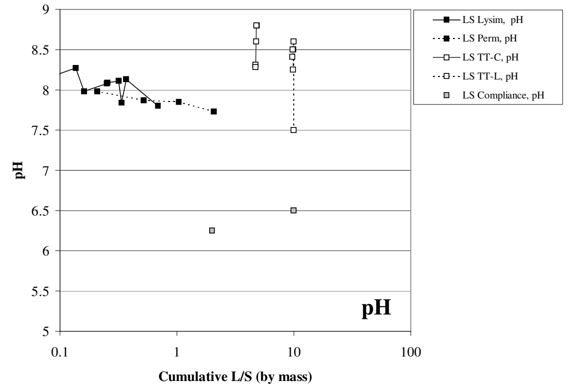


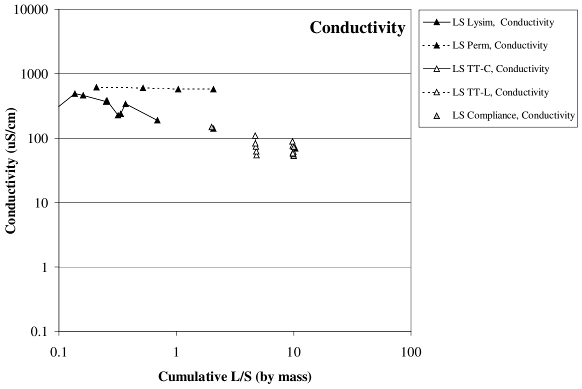


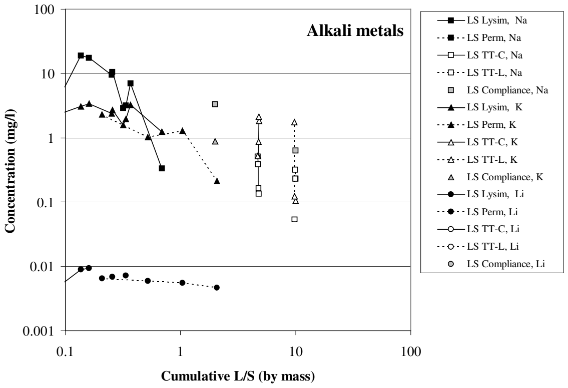


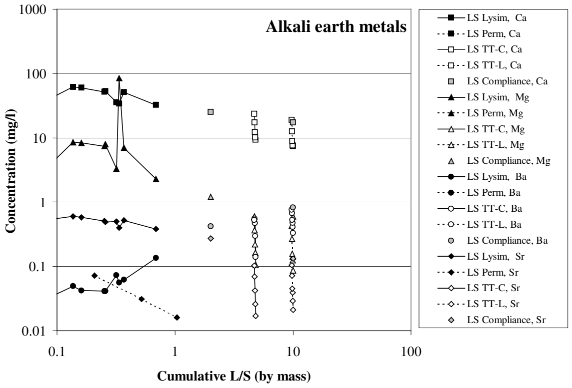


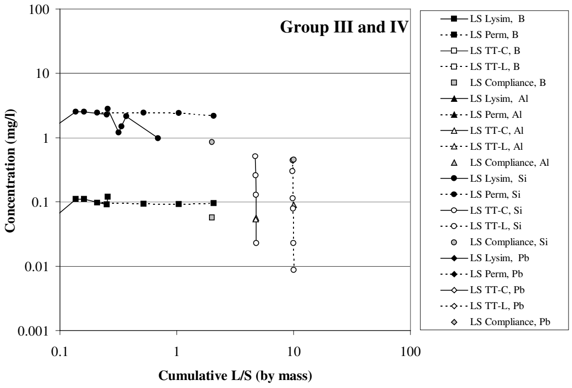
LIMESTONE
Concentrations in the leachate (mg/l) against cumulative L/S ratio

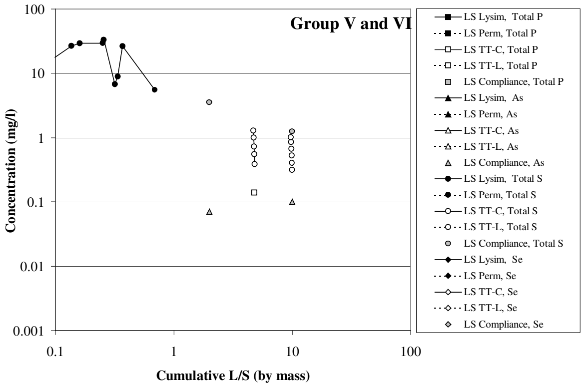


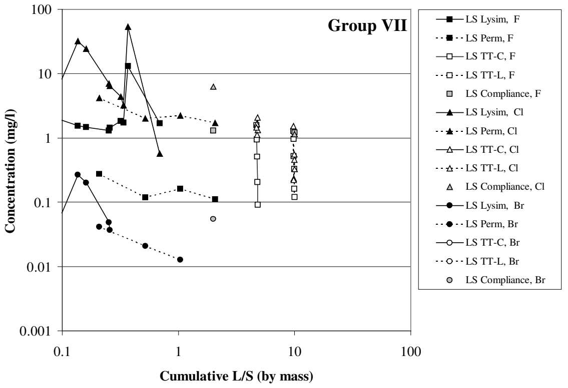


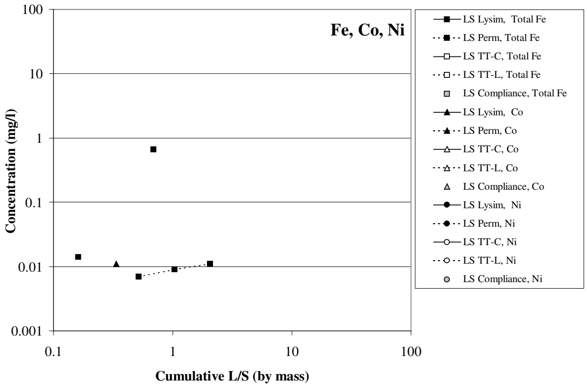




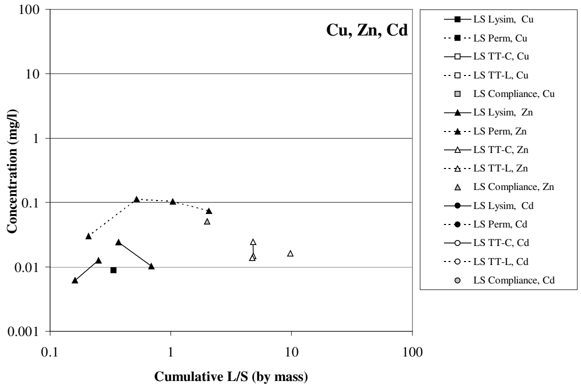


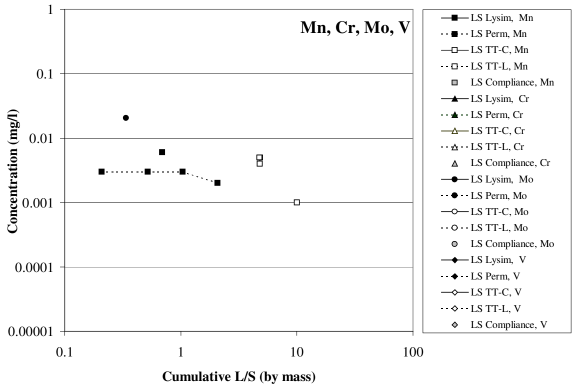


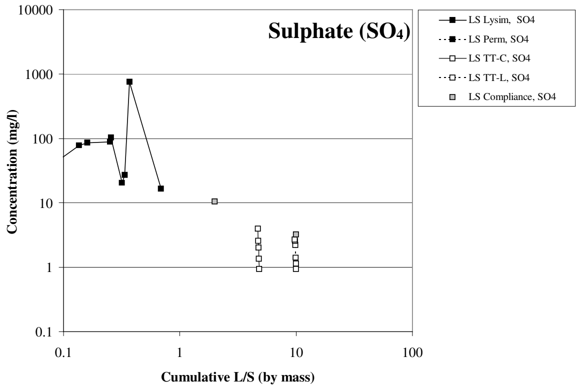


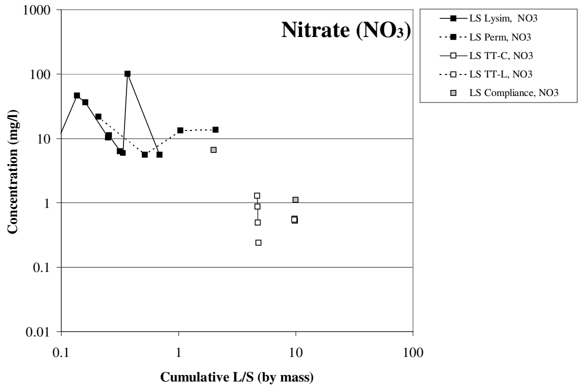


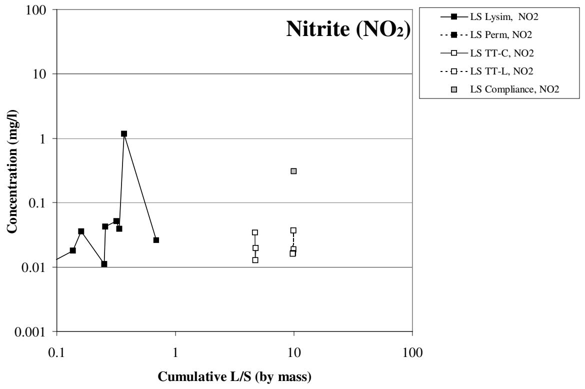
LIMESTONE
Concentrations in the leachate (mg/l) against cumulative L/S ratio

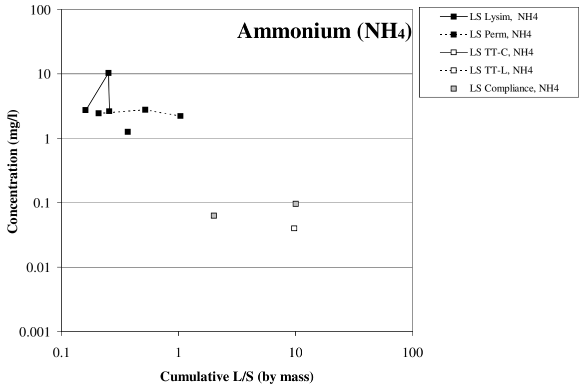


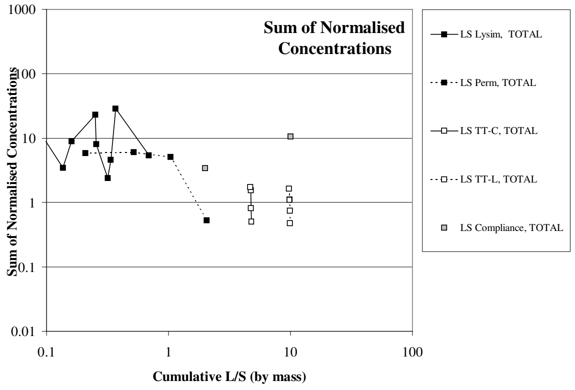


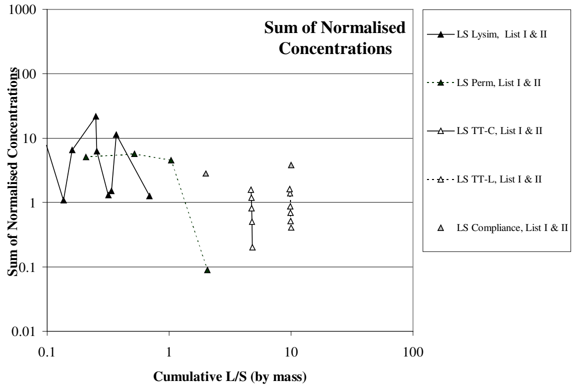




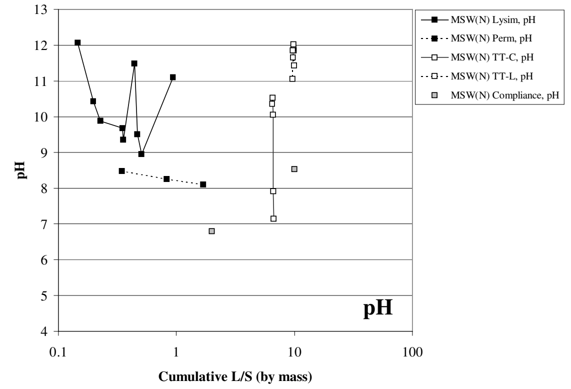


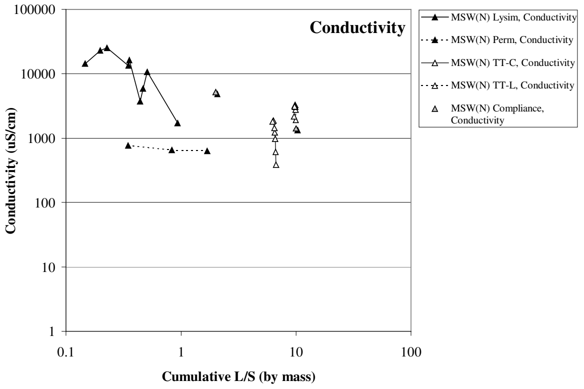


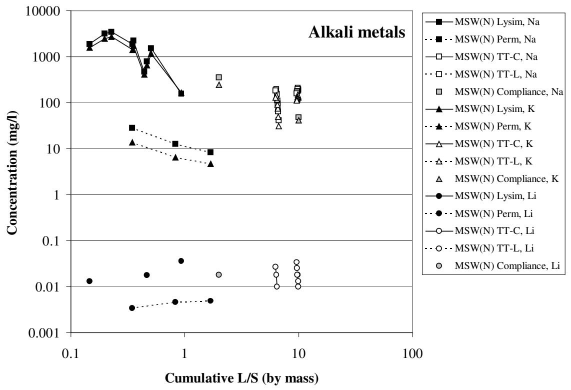


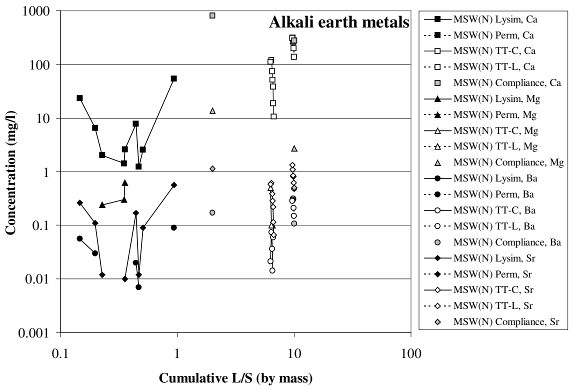


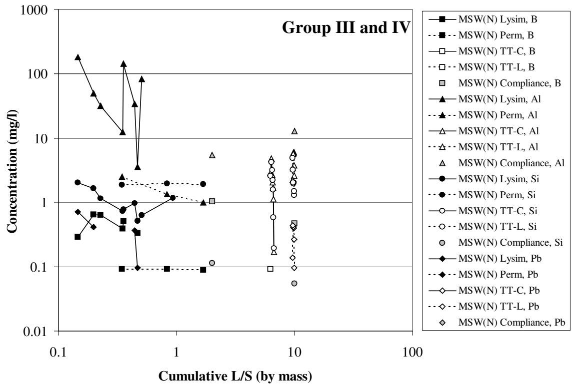
MSW INCINERATOR ASH
Concentrations in the leachate (mg/l) against cumulative L/S ratio

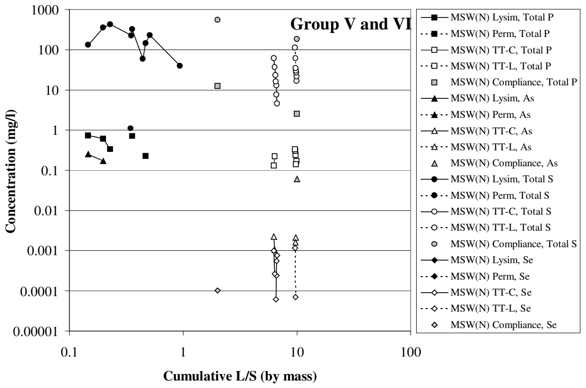


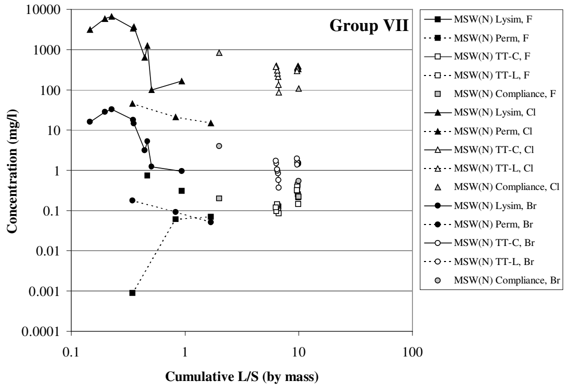


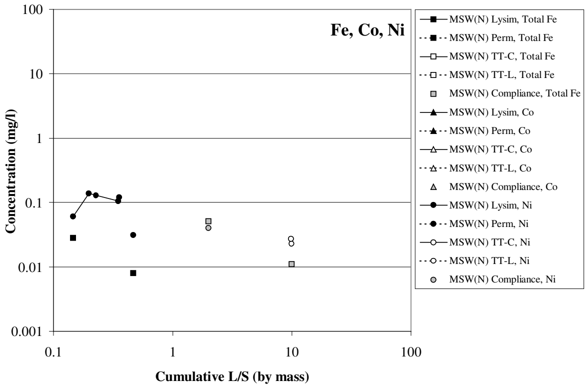




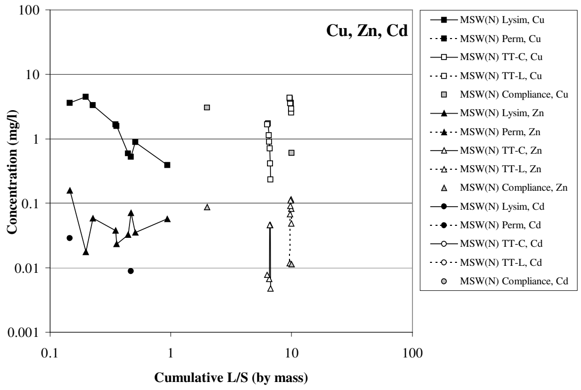


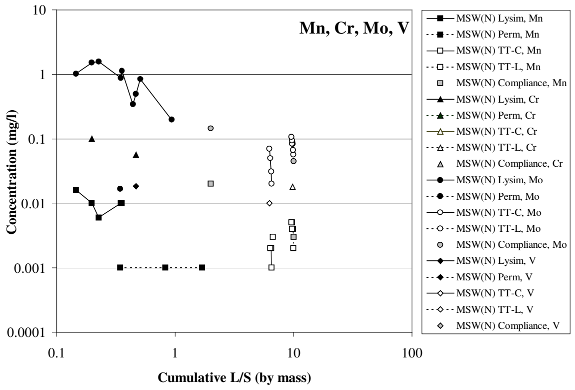


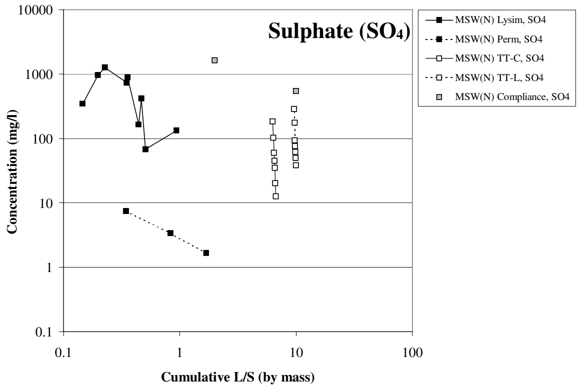


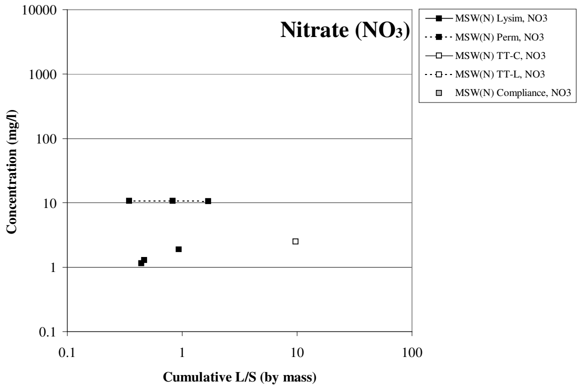


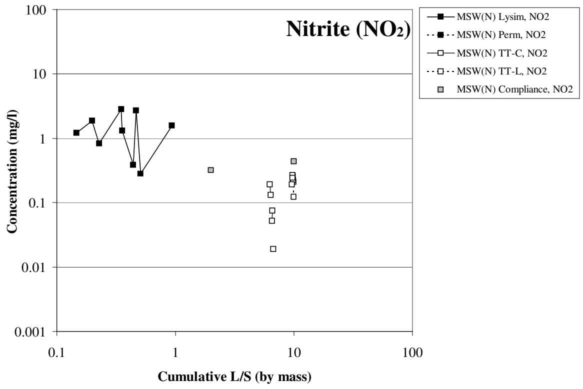
MSW INCINERATOR ASH
Concentrations in the leachate (mg/l) against cumulative L/S ratio

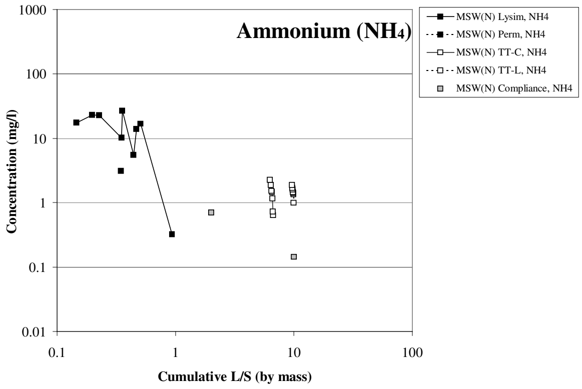


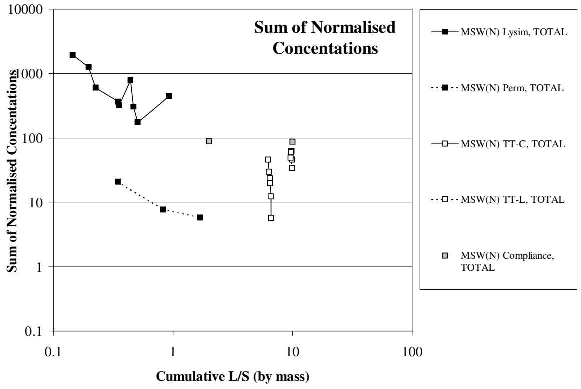


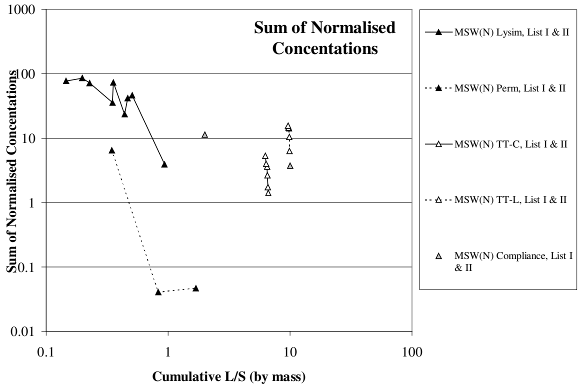




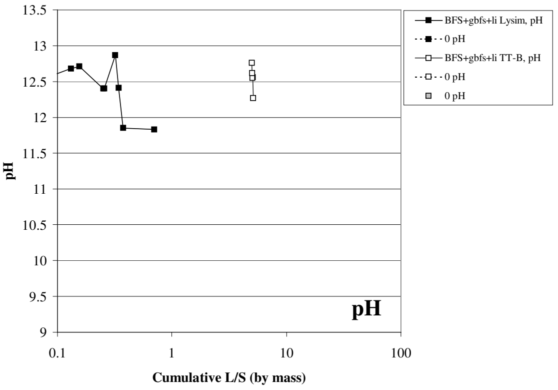


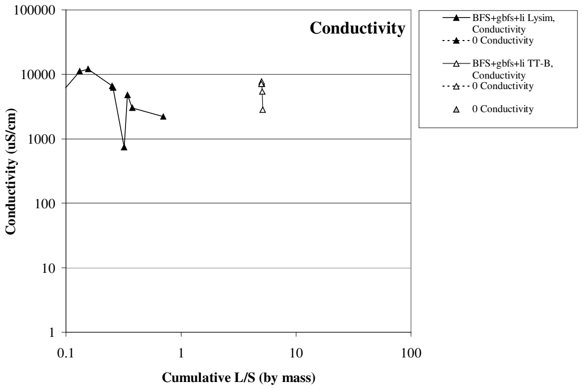


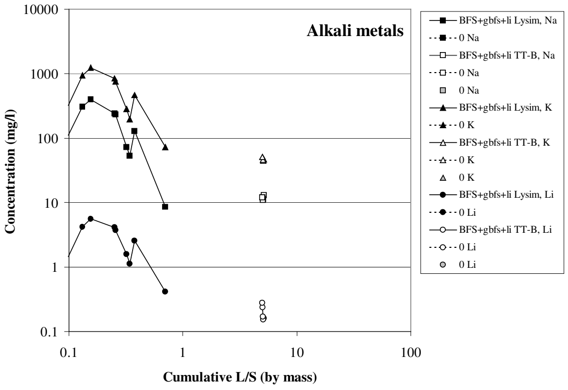


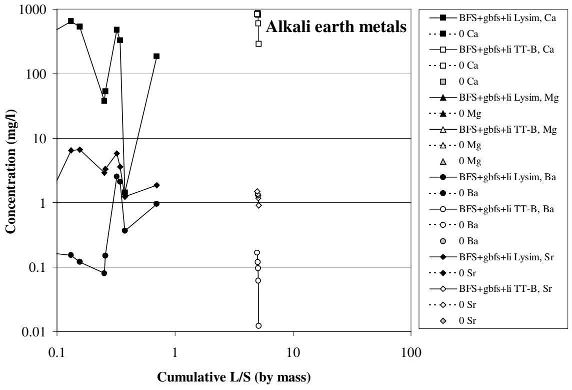


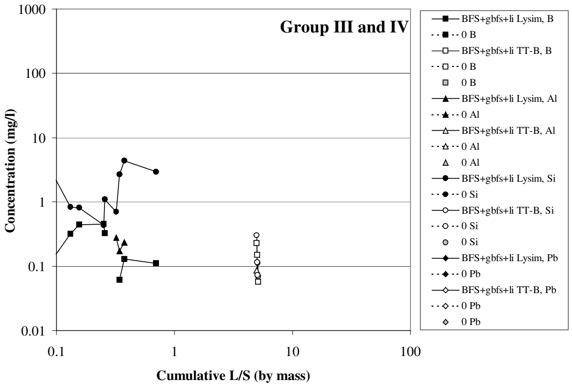
BLASTFURNACE SLAG + granulated blastfurnace slag + lime
Concentrations in the leachate (mg/l) against cumulative L/S ratio

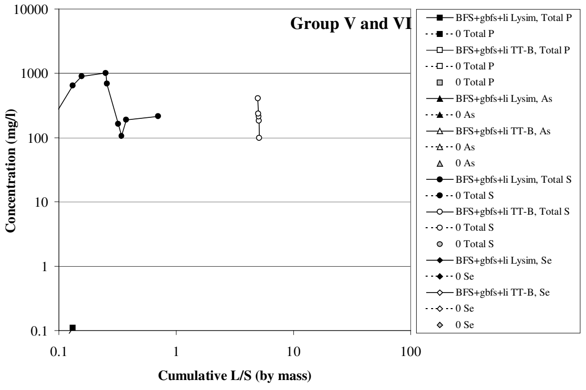


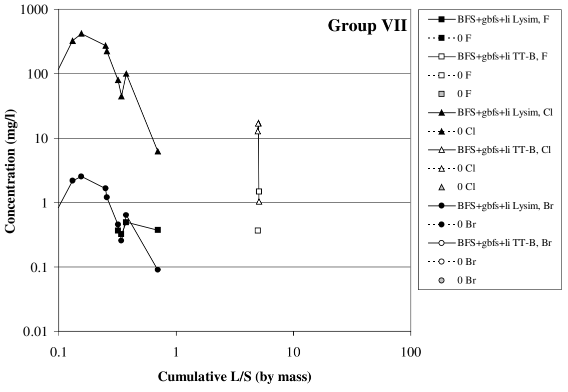


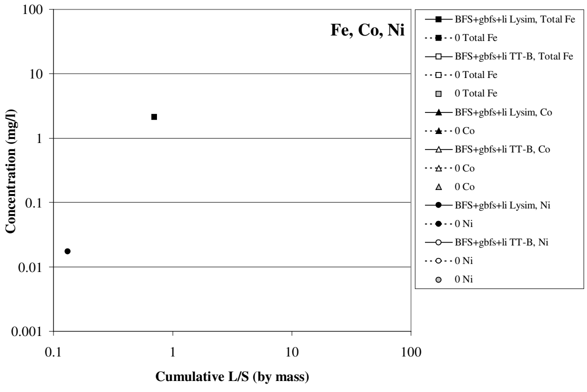




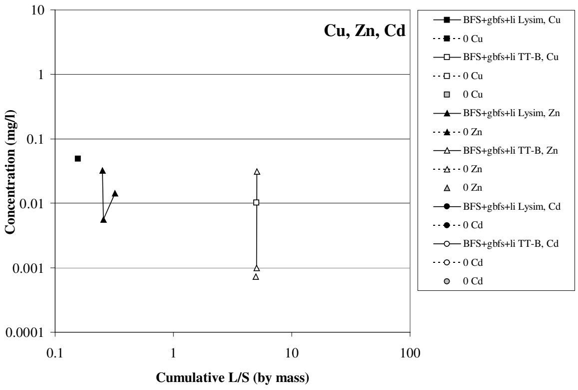


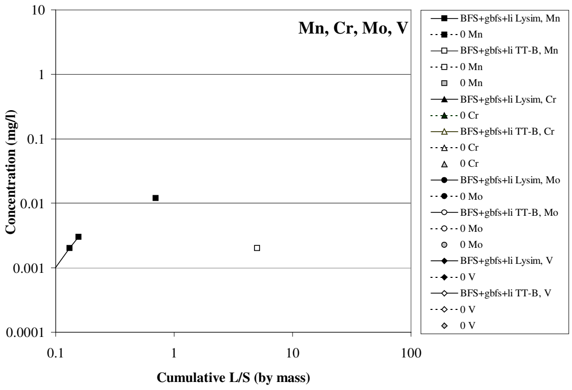


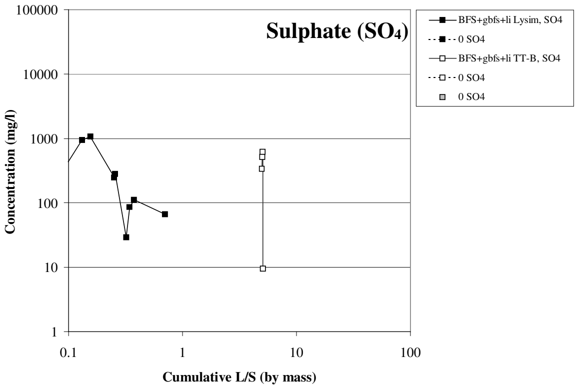


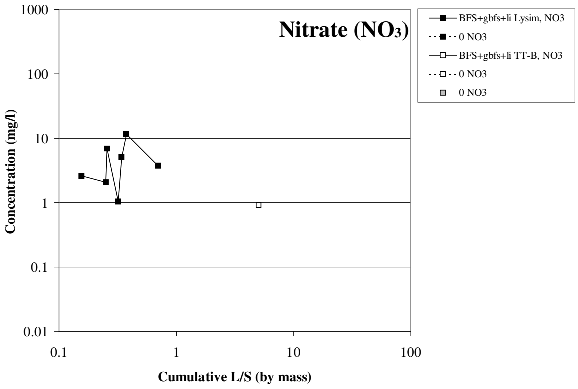


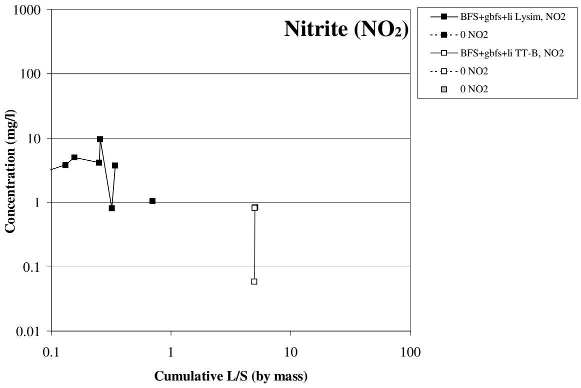
BLASTFURNACE SLAG + granulated blastfurnace slag + lime
Concentrations in the leachate (mg/l) against cumulative L/S ratio

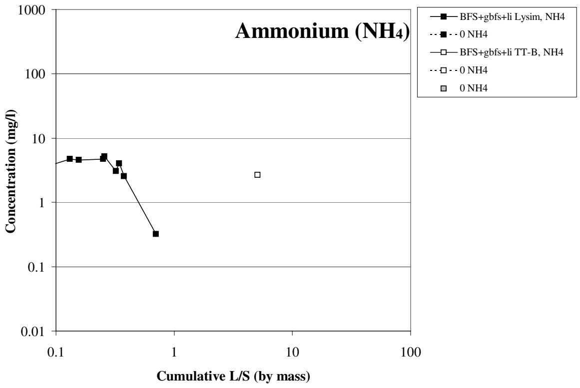


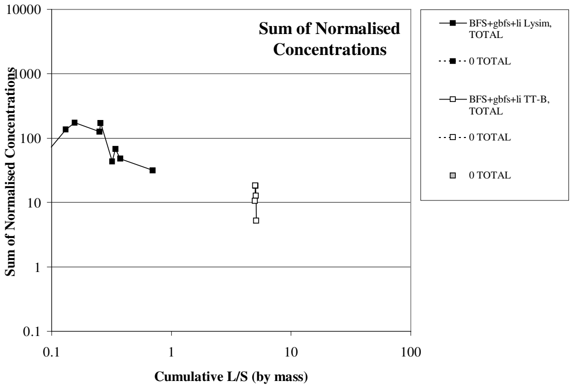


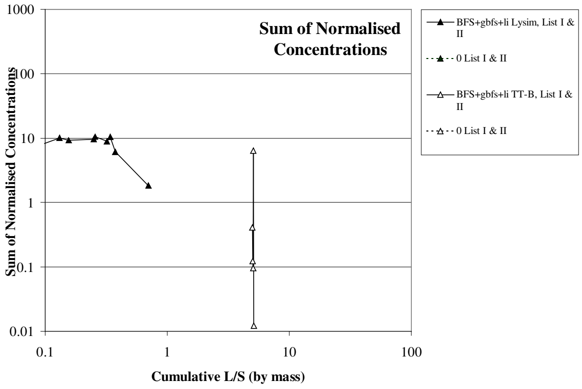




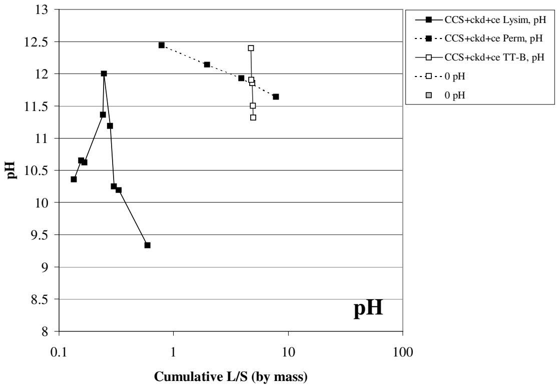


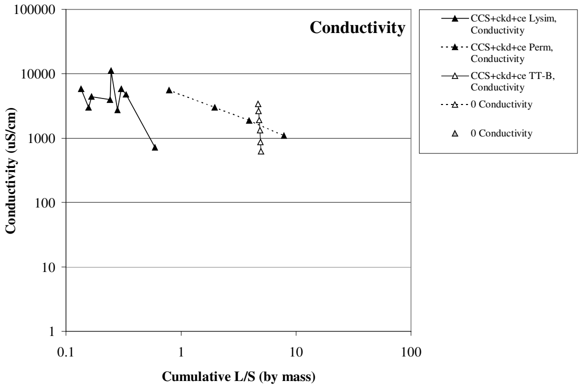


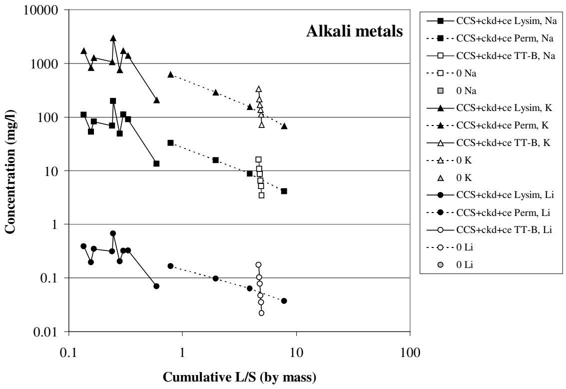


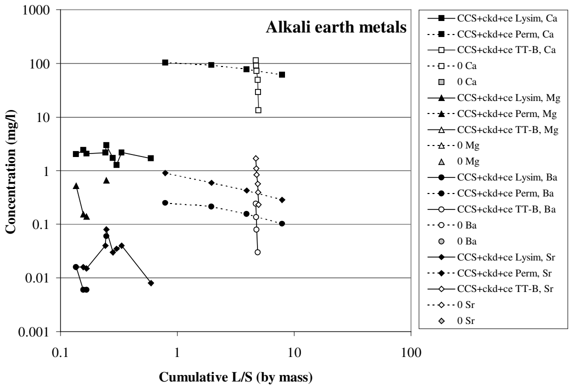


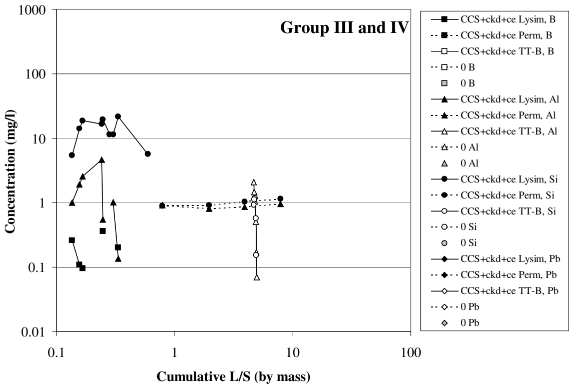
CHINA CLAY SAND + cement kiln dust + cement
Concentrations in the leachate (mg/l) against cumulative L/S ratio

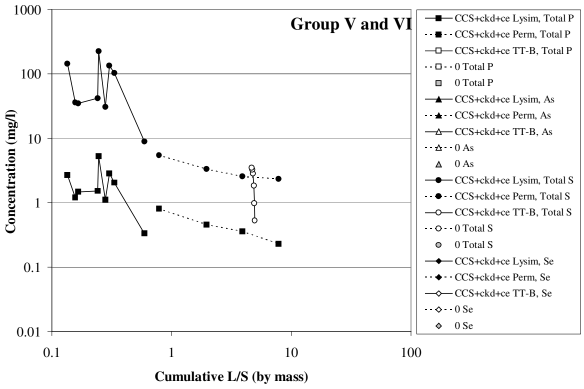


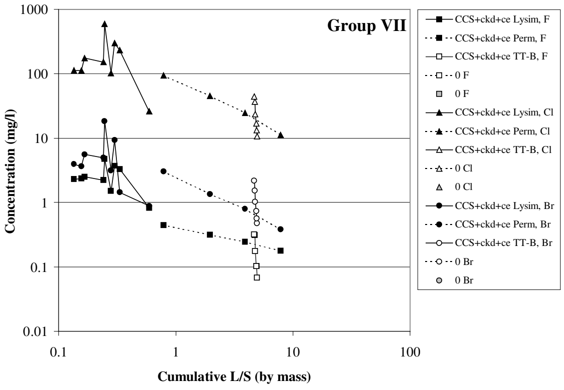


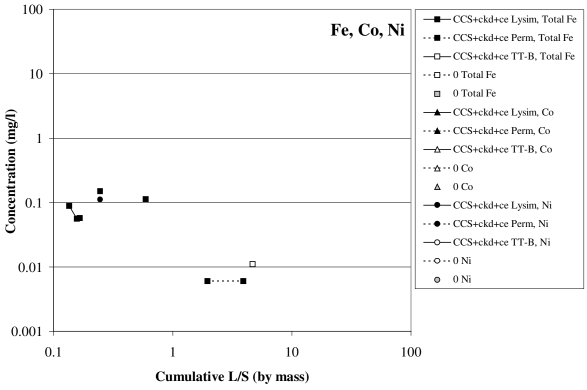




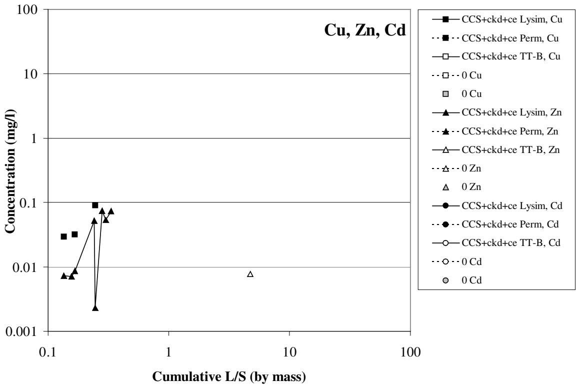


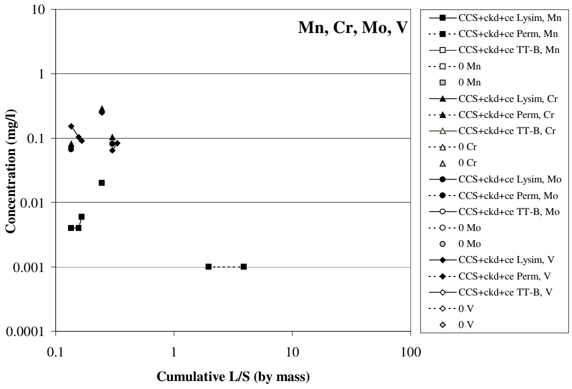


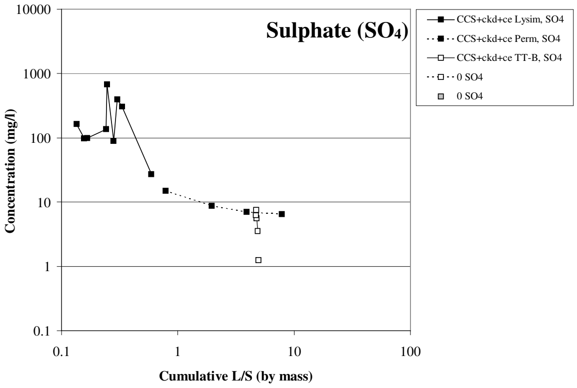


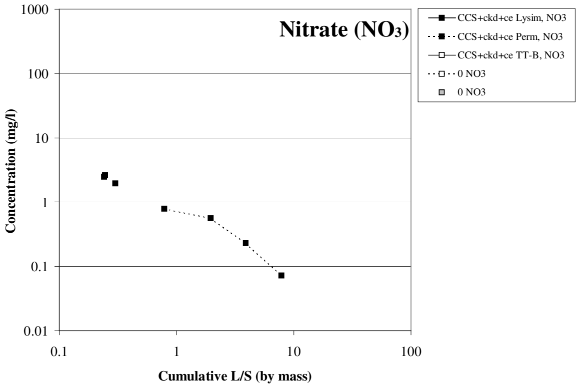


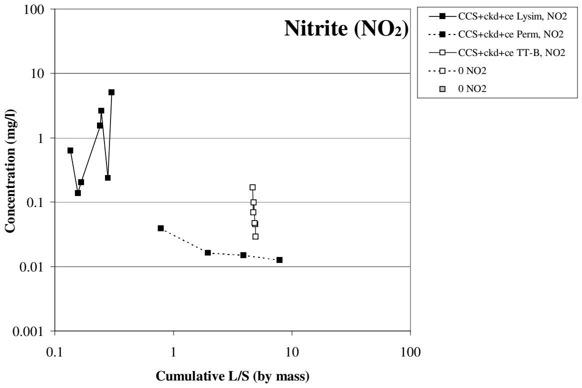
CHINA CLAY SAND + cement kiln dust + cement
Concentrations in the leachate (mg/l) against cumulative L/S ratio

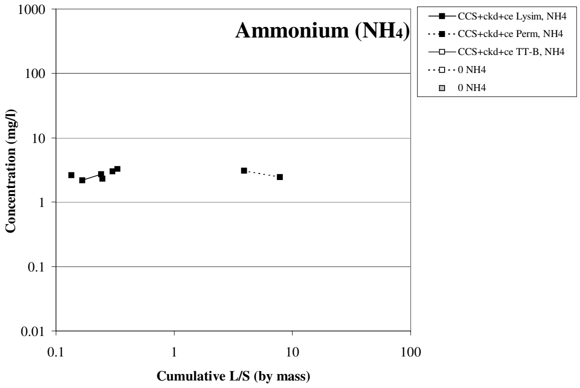


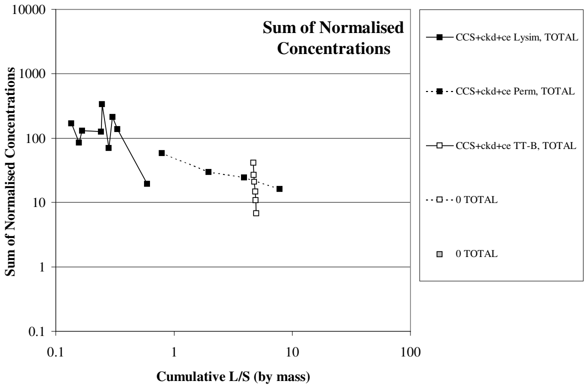


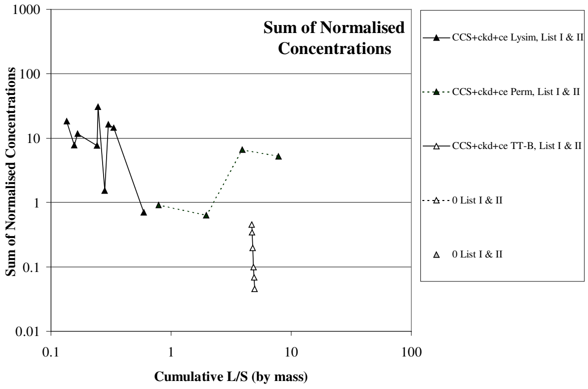




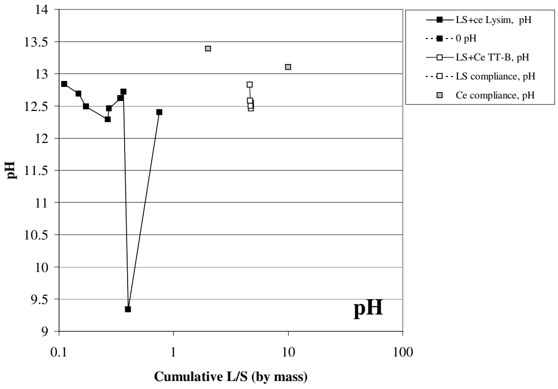


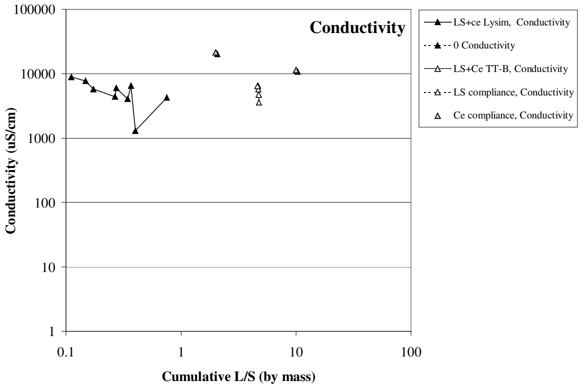


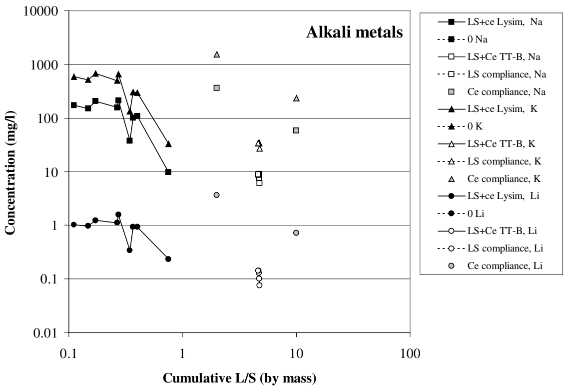


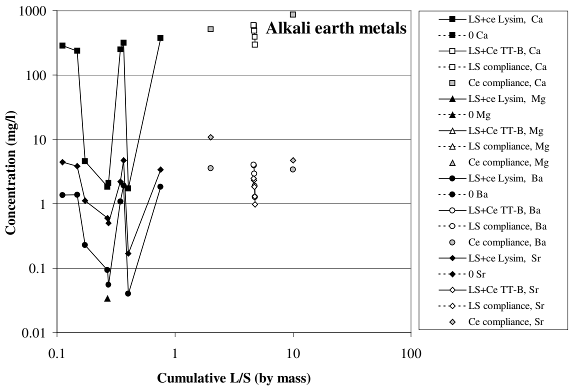


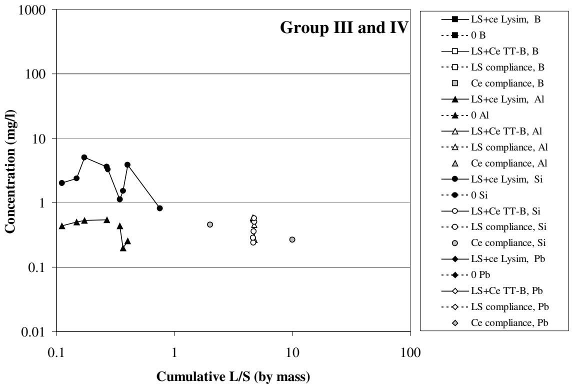
LIMESTONE + cement
Concentrations in the leachate (mg/l) against cumulative L/S ratio

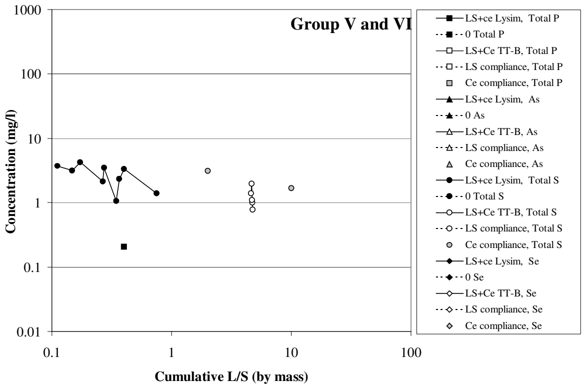


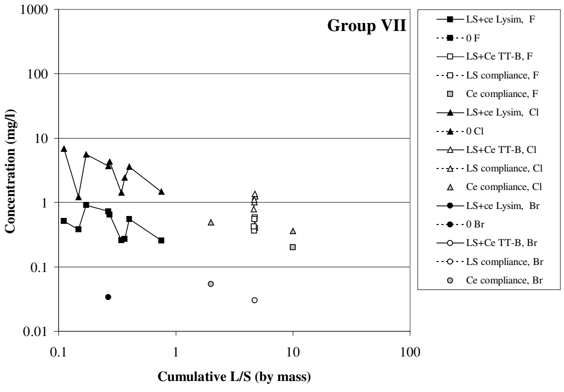


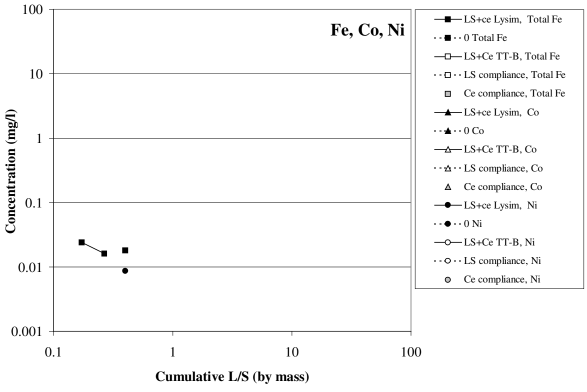




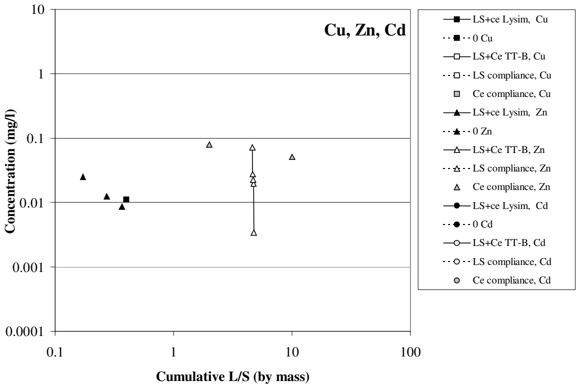


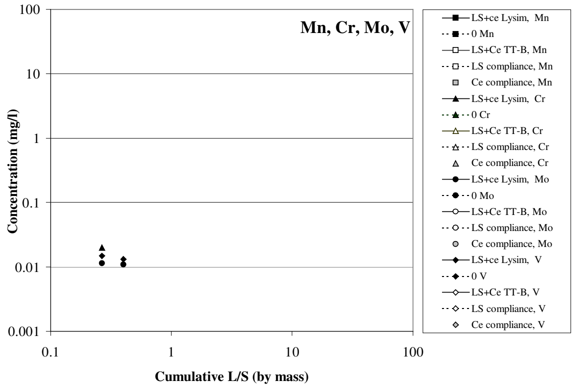


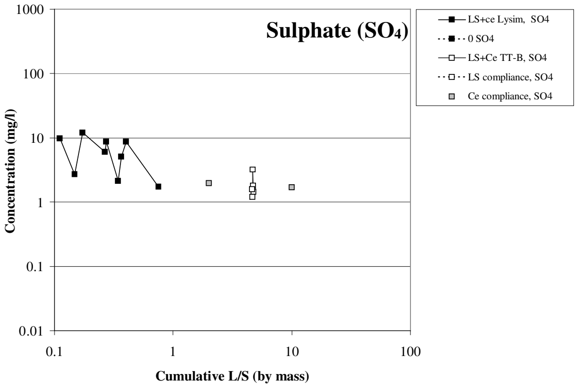


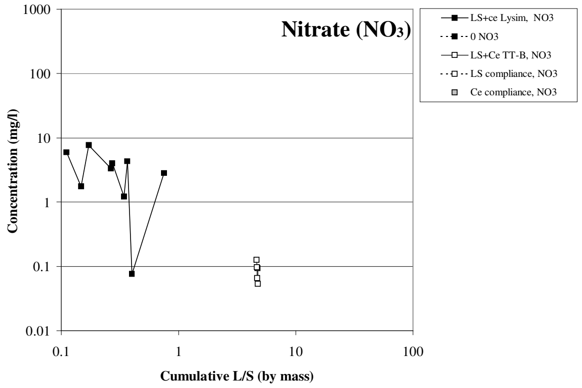


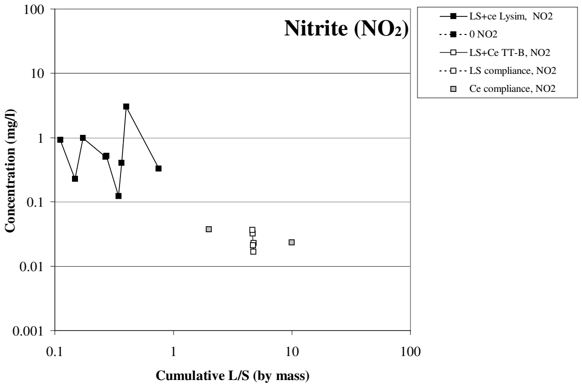
LIMESTONE + cement
Concentrations in the leachate (mg/l) against cumulative L/S ratio

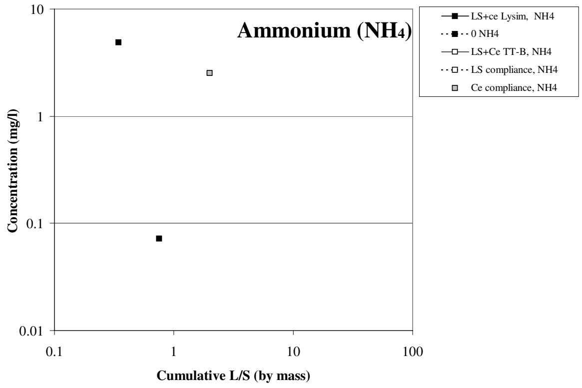


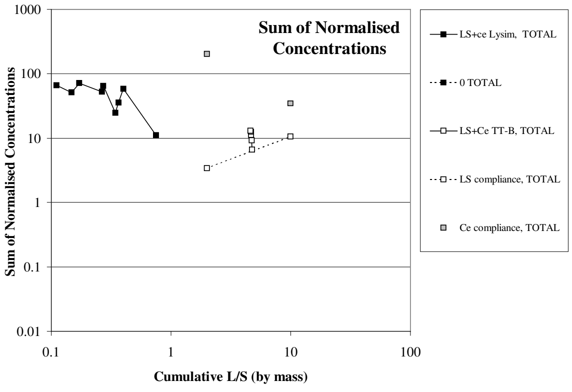


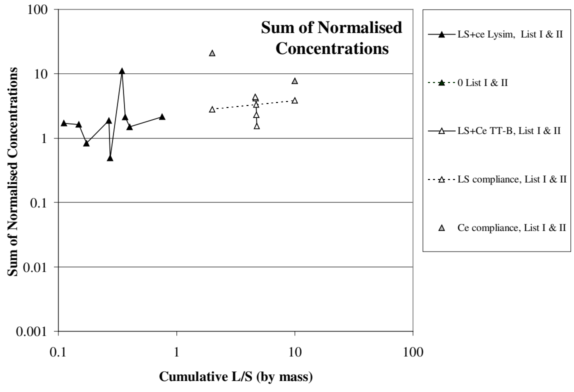




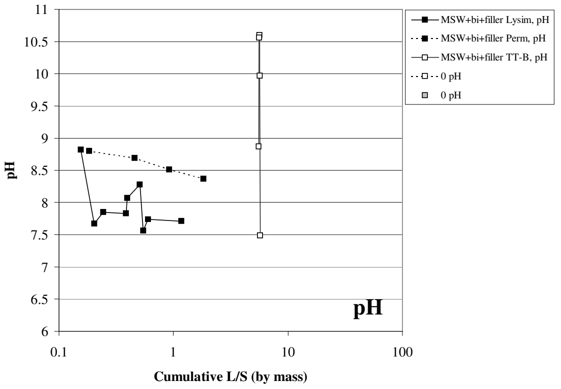


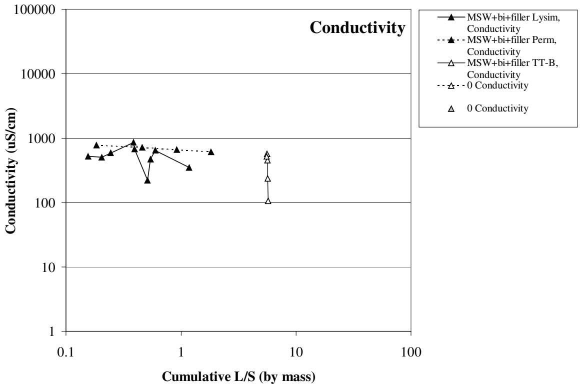


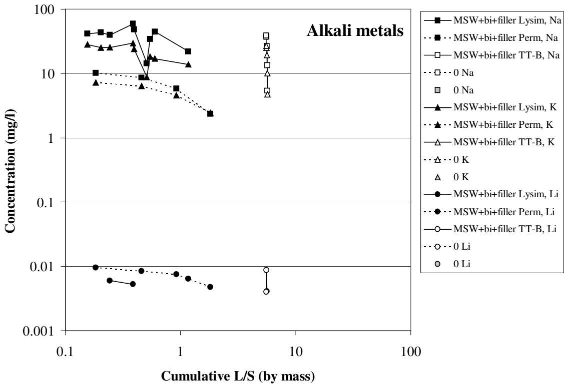


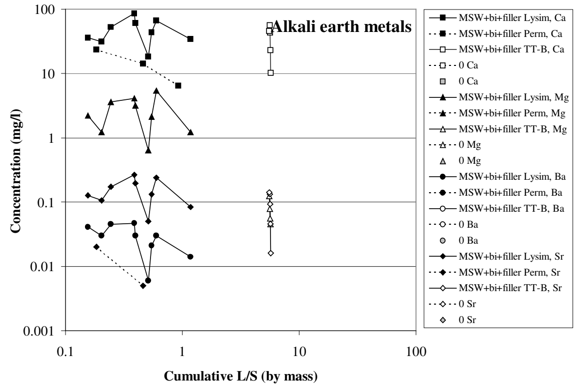


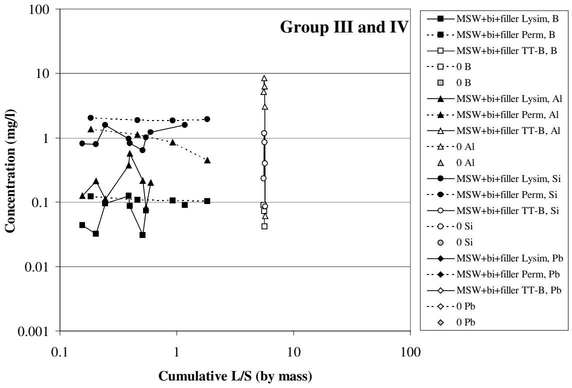
MSW INCINERATOR ASH +bitumen + filler
Concentrations in the leachate (mg/l) against cumulative L/S ratio

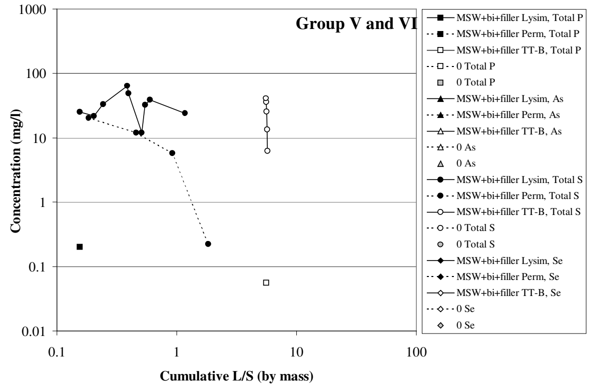


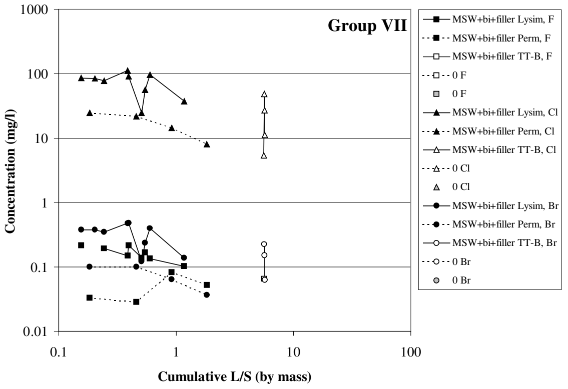


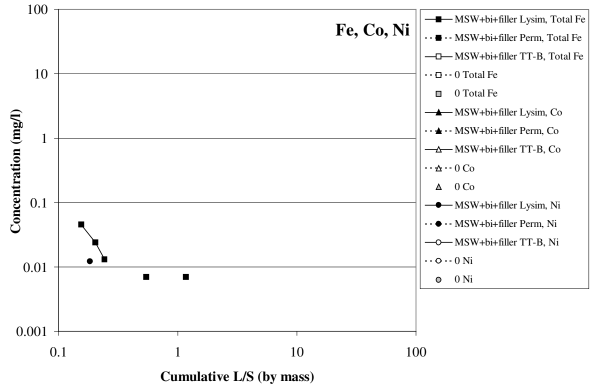




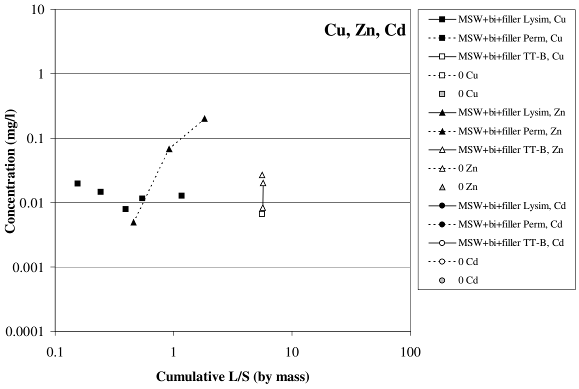


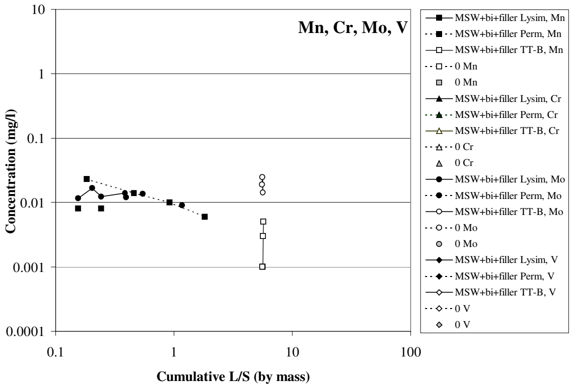


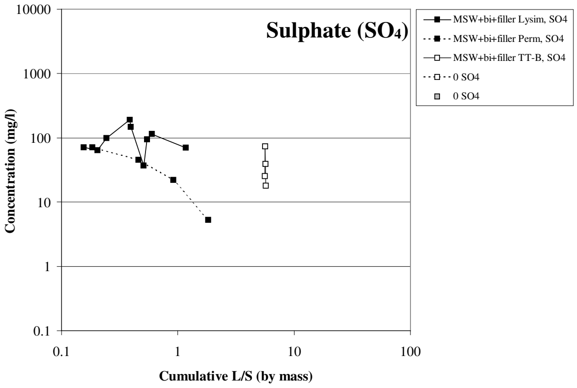


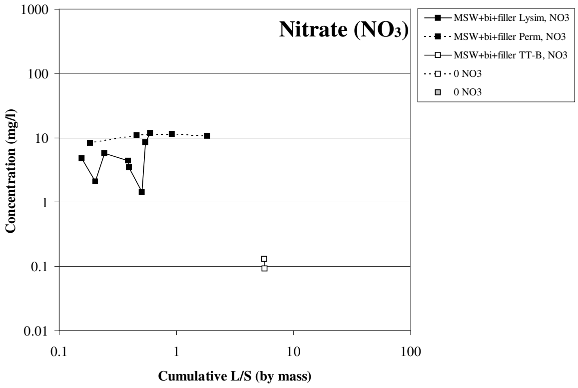


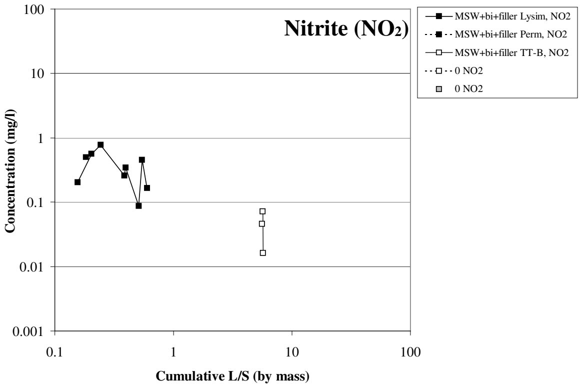
MSW INCINERATOR ASH +bitumen + filler
Concentrations in the leachate (mg/l) against cumulative L/S ratio

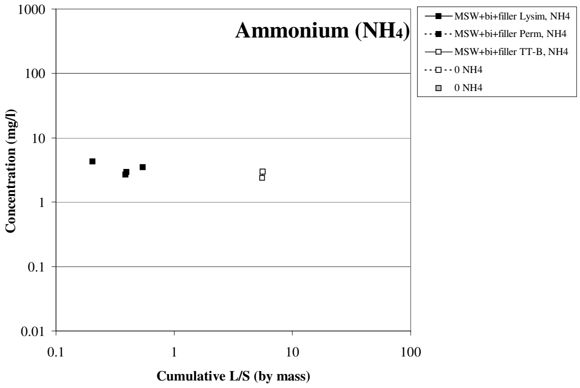


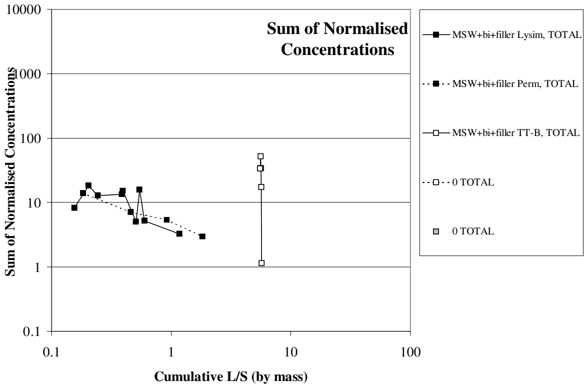


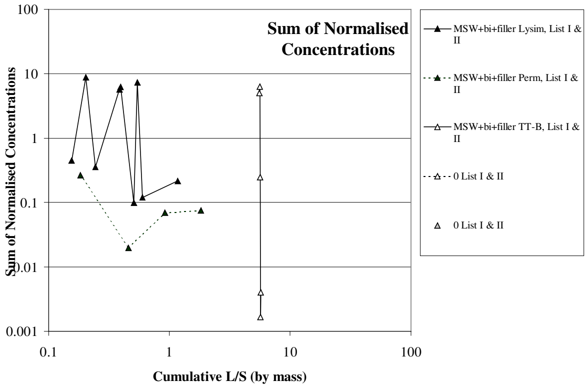










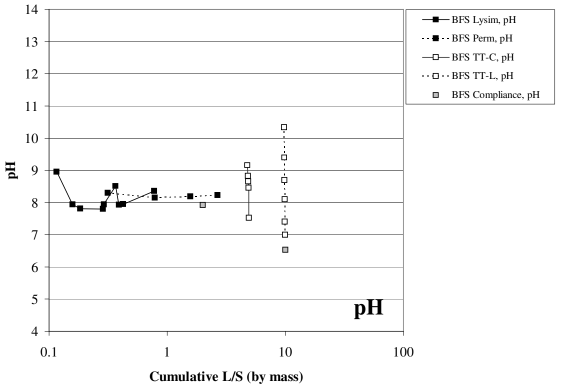


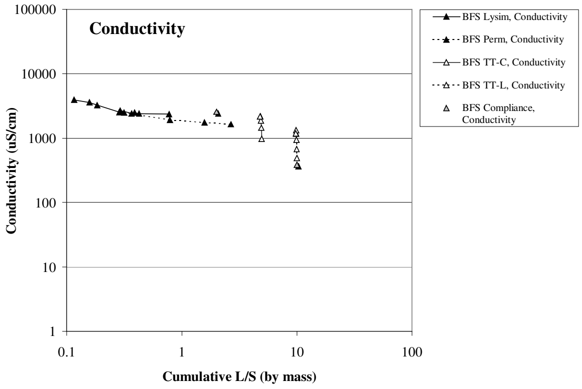
APPENDIX E:

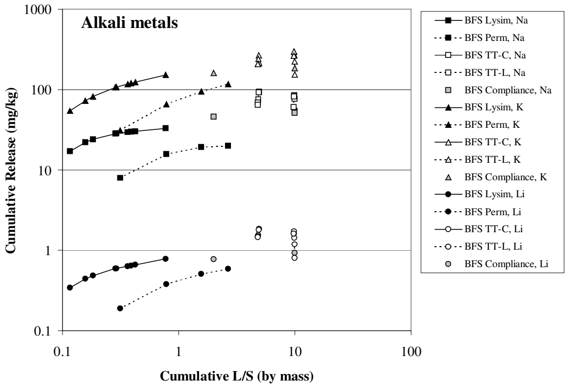
Graphs comparing laboratory leaching tests and lysimeters

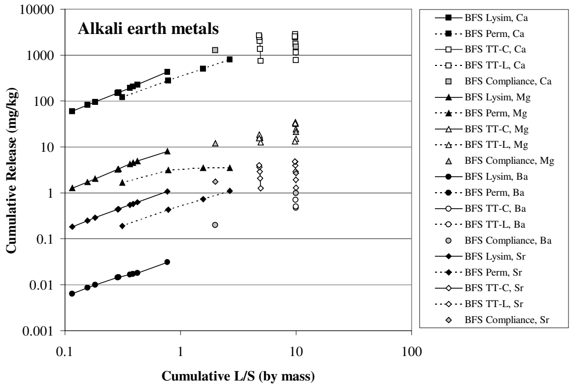
L/S ratio verses cumulative release (mg/kg)

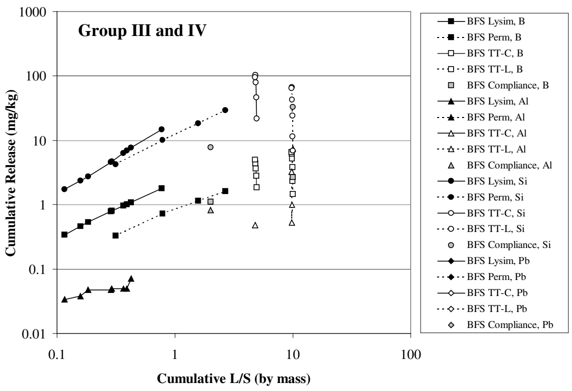
BLASTFURNACE SLAG
Cumulative release (mg/kg) against cumulative L/S ratio

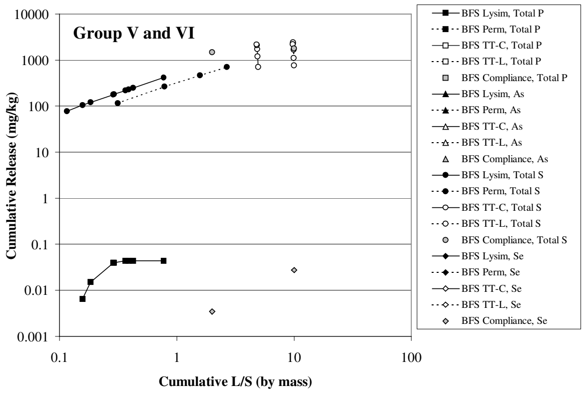


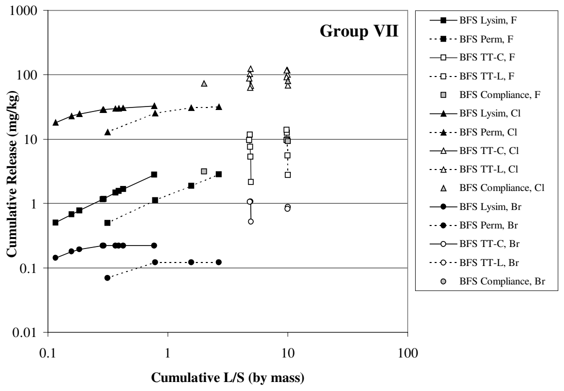


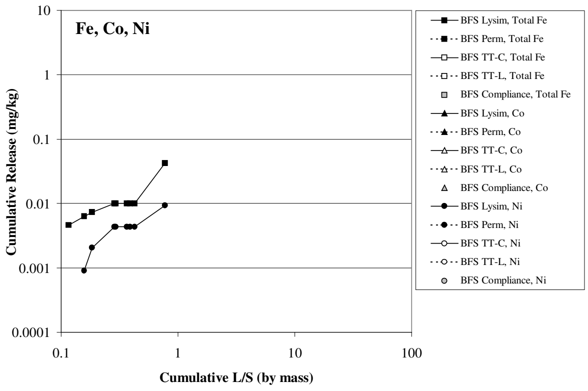




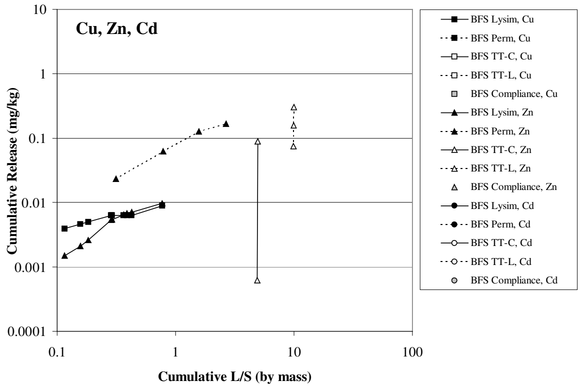


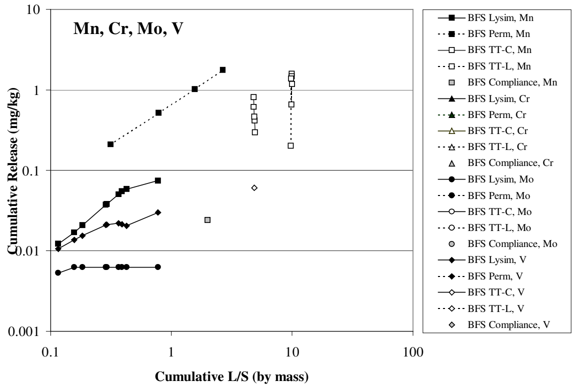


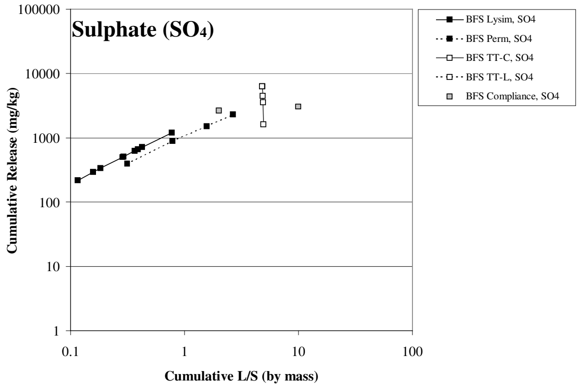


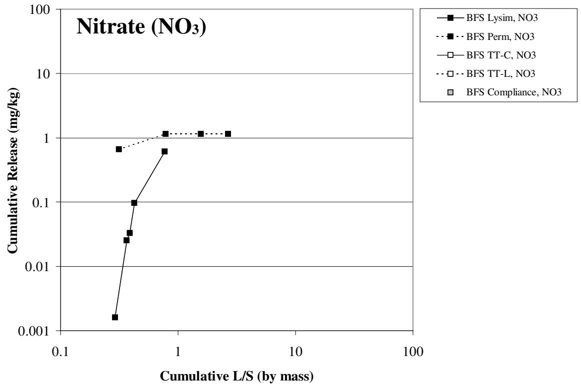


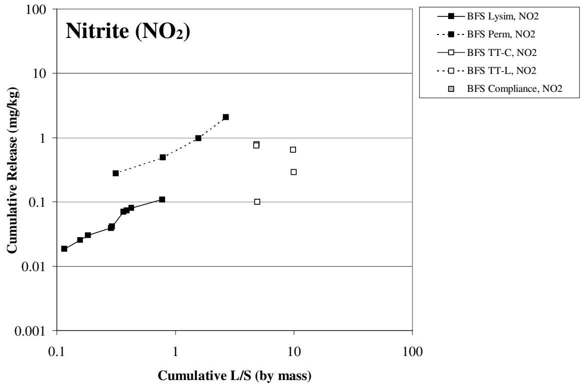
BLASTFURNACE SLAG
Cumulative release (mg/kg) against cumulative L/S ratio

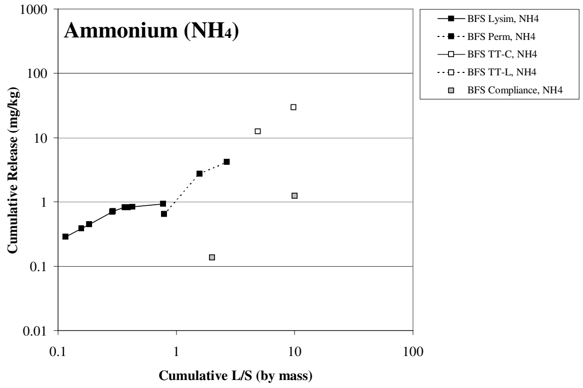




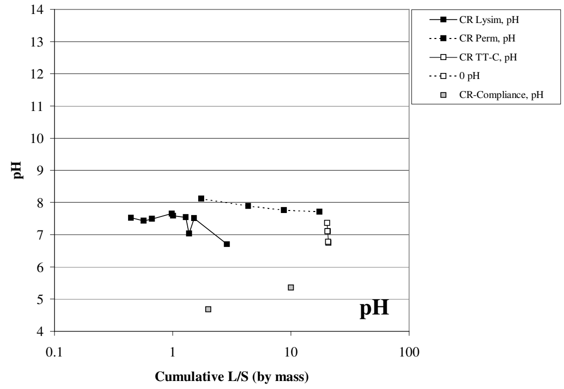


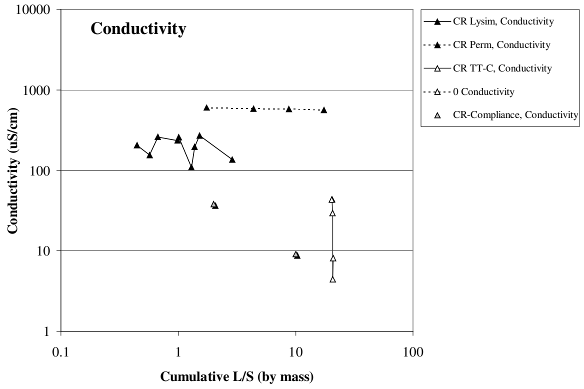


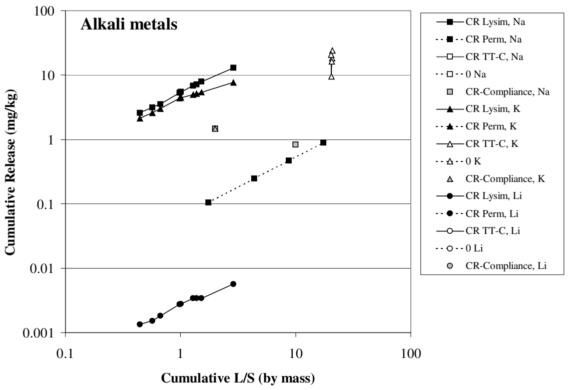


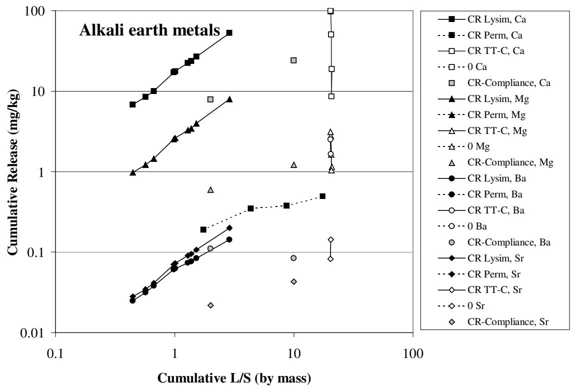


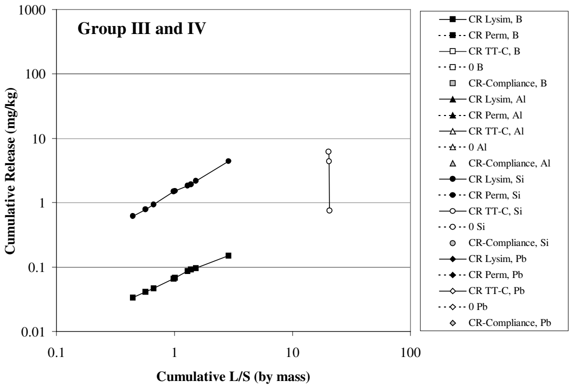
CRUMBED RUBBER (2-8mm)
Cumulative release (mg/kg) against cumulative L/S ratio

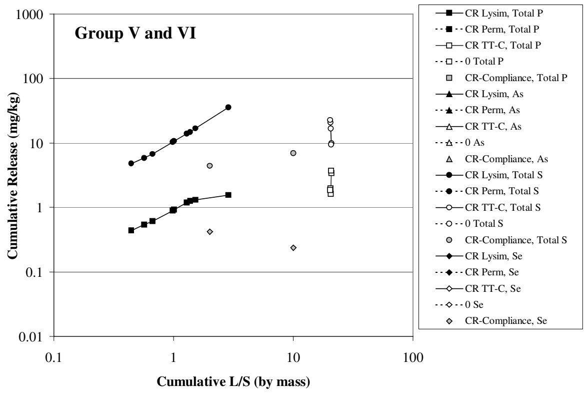


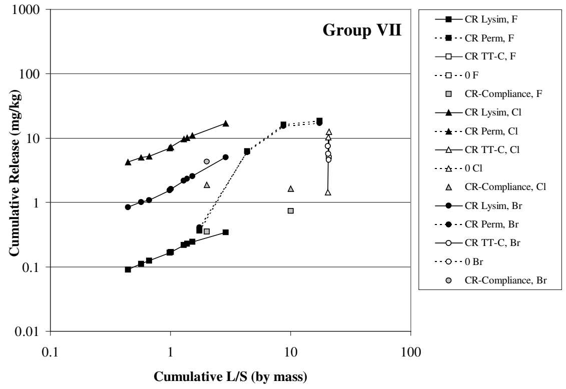


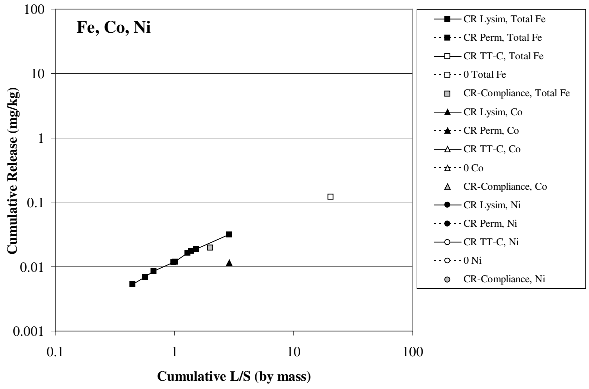




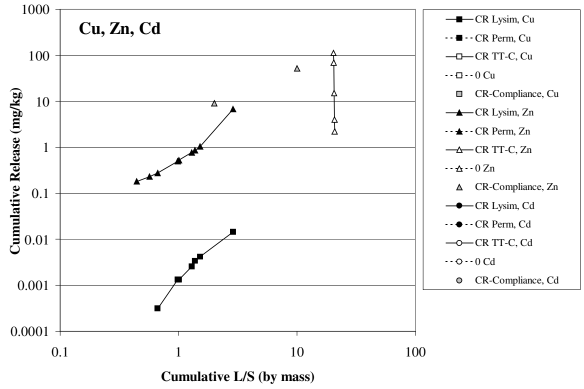


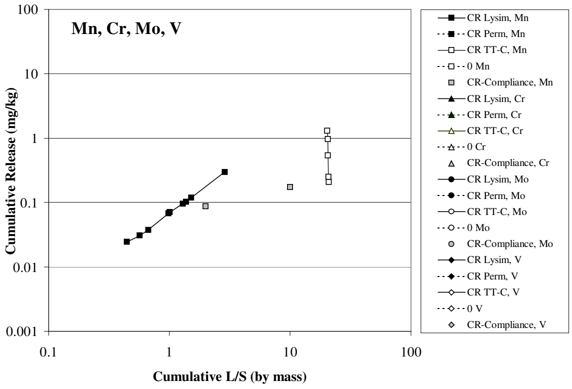


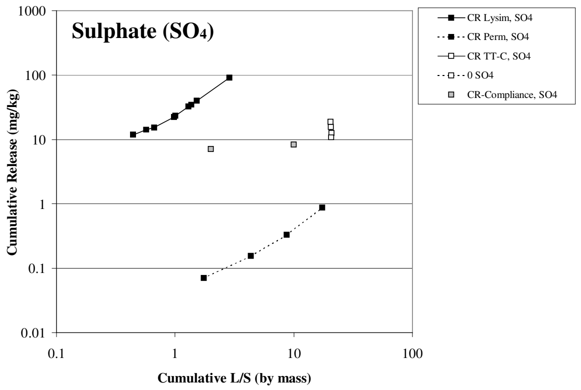


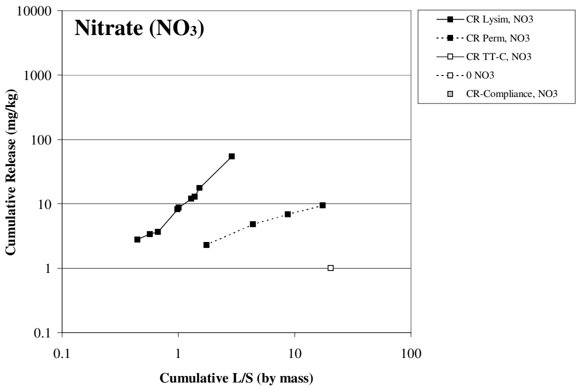


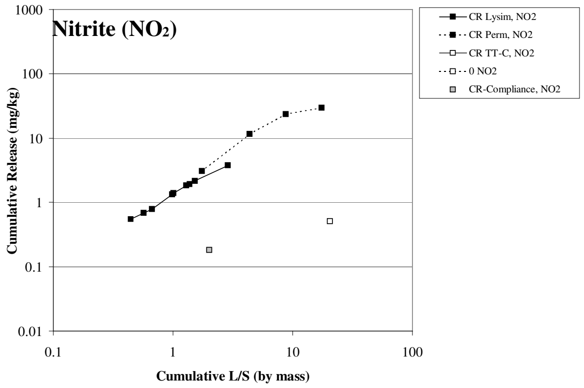
CRUMBED RUBBER (2-8mm)
Cumulative release (mg/kg) against cumulative L/S ratio

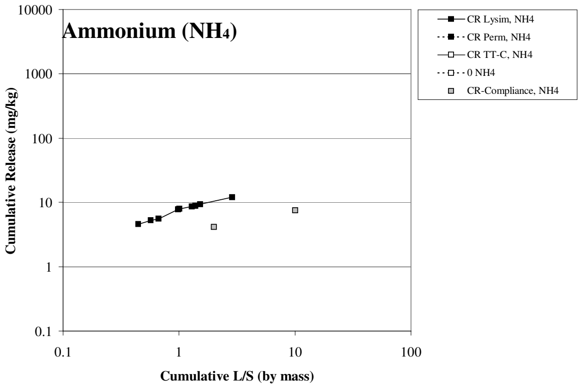




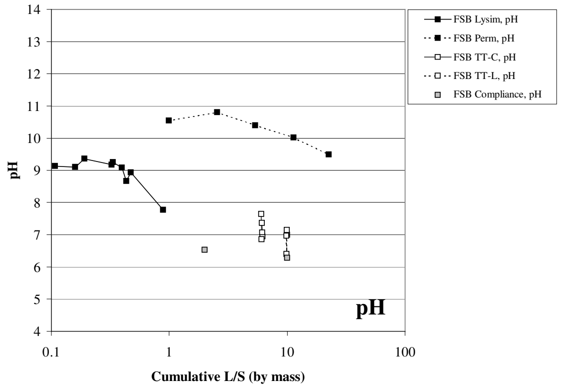


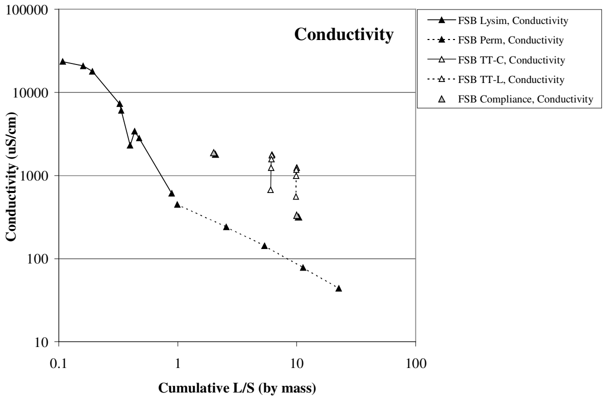


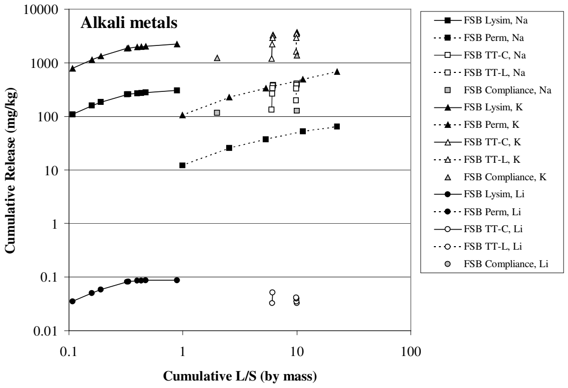


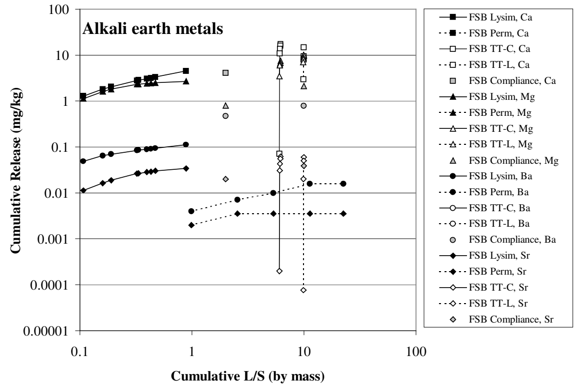


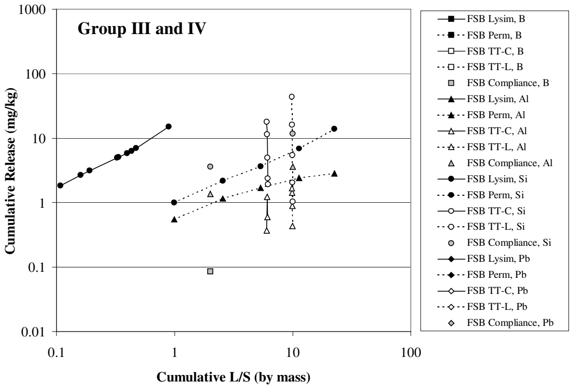
FOUNDRY SAND (PHENOLIC, BRASS)
Cumulative release (mg/kg) against cumulative L/S ratio

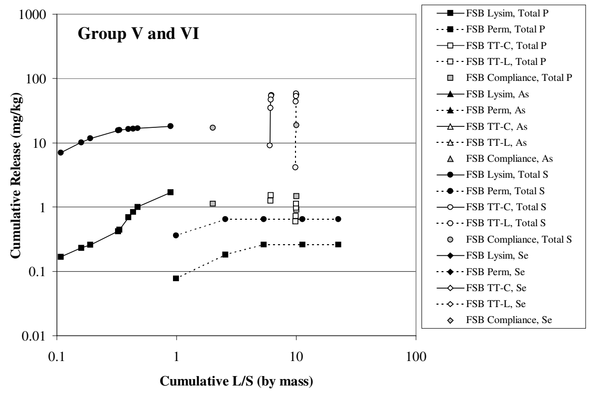


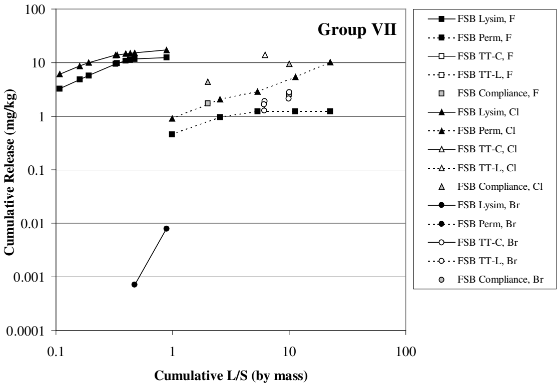


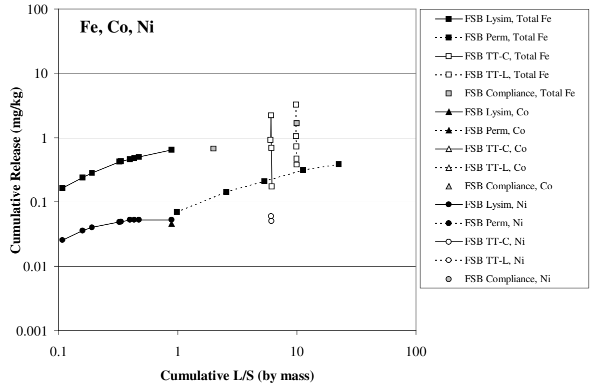




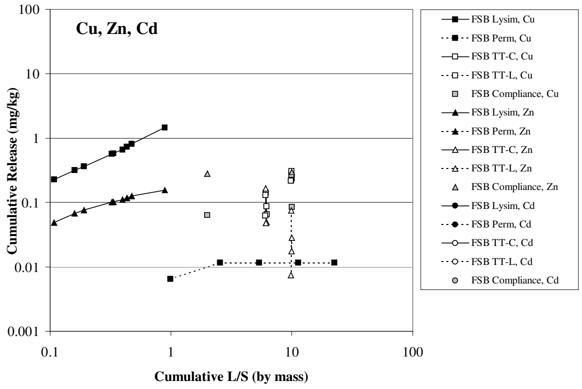


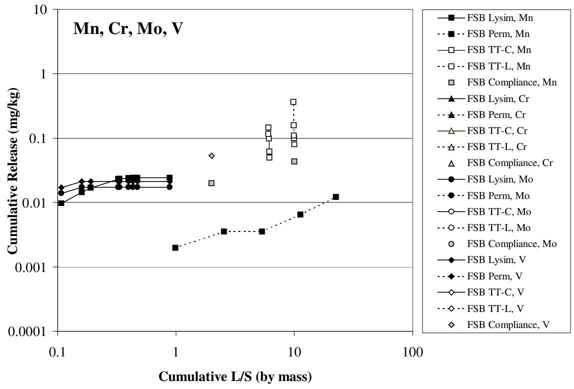


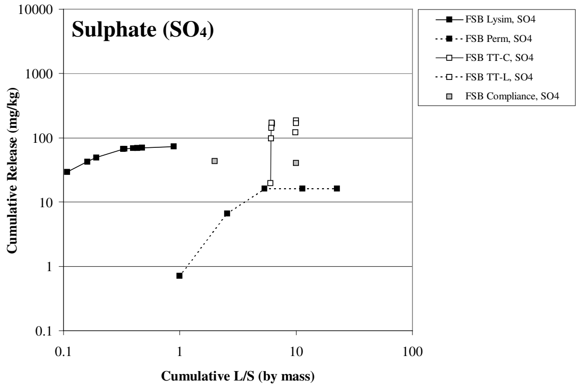


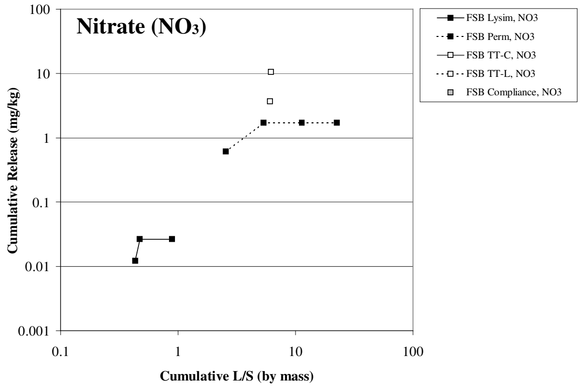


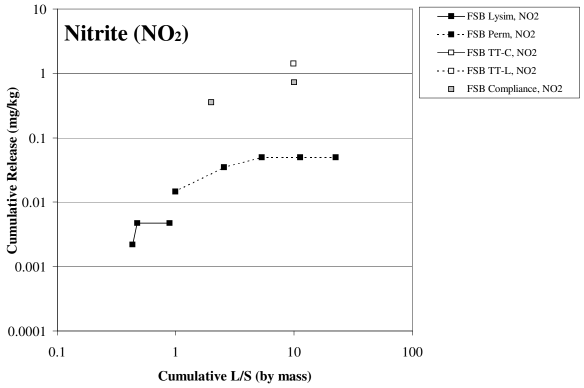
FOUNDRY SAND (PHENOLIC, BRASS)
Cumulative release (mg/kg) against cumulative L/S ratio

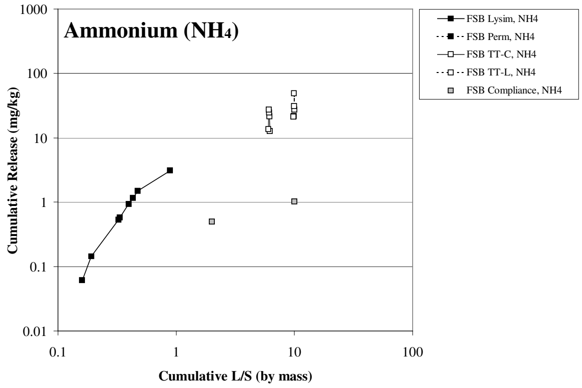




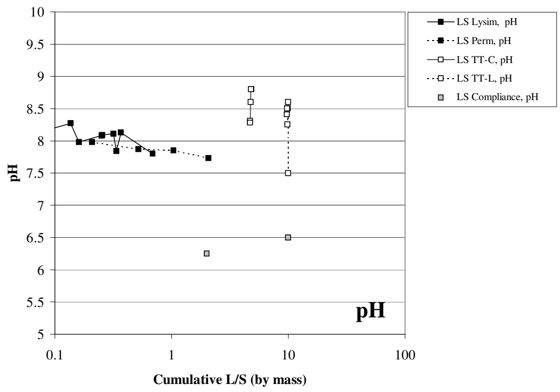


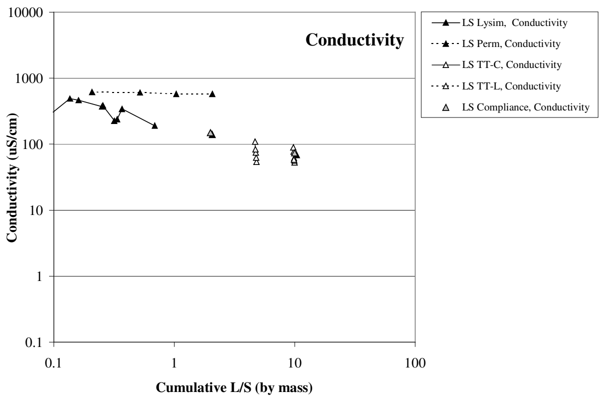


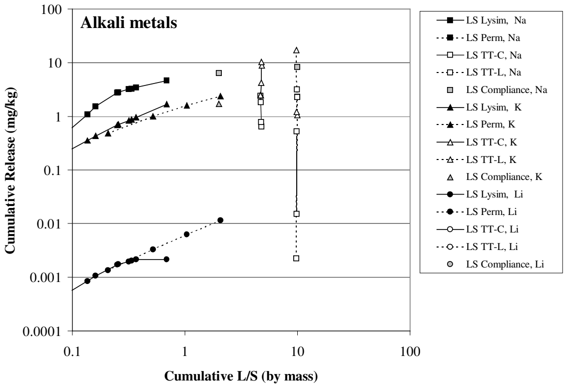


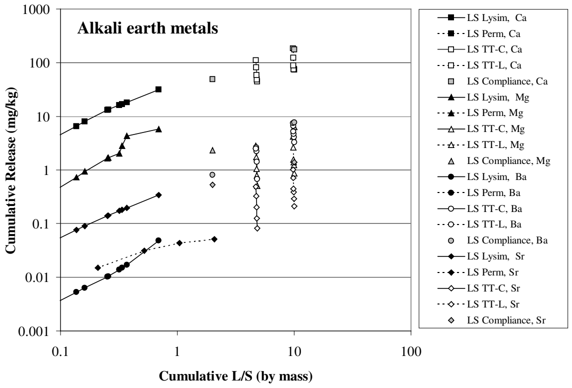


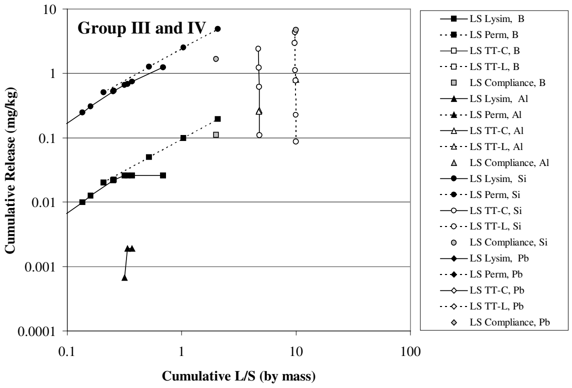
LIMESTONE
Cumulative release (mg/kg) against cumulative L/S ratio

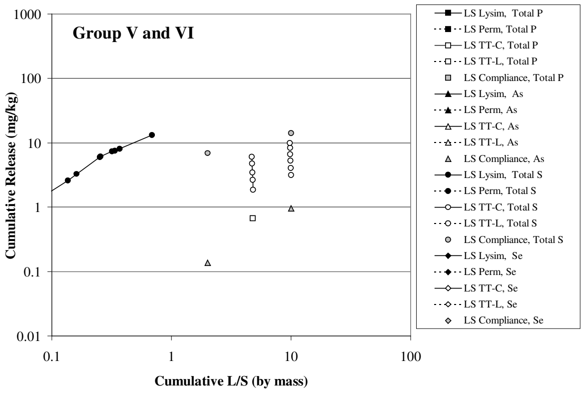


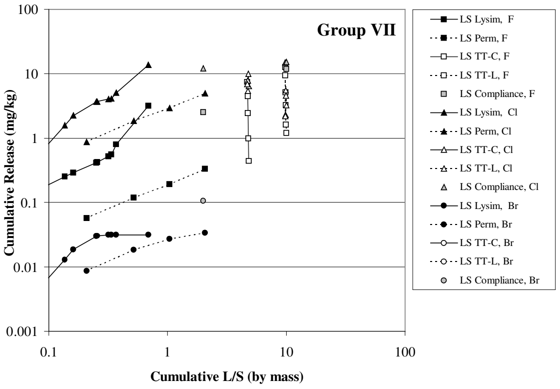


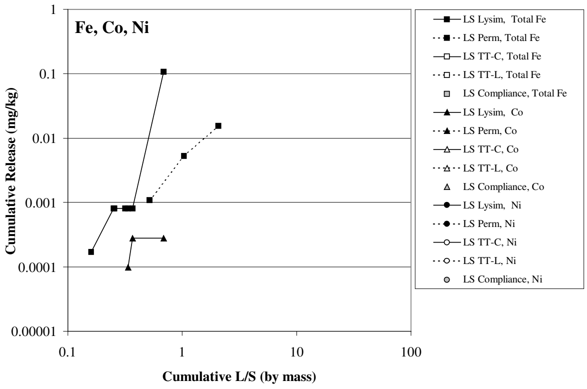




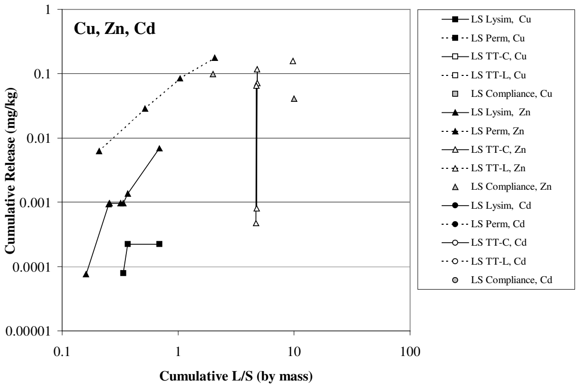


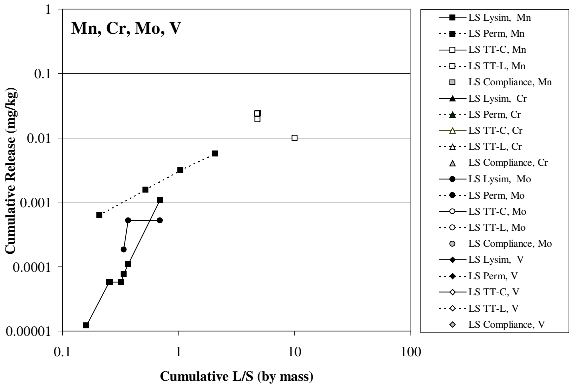


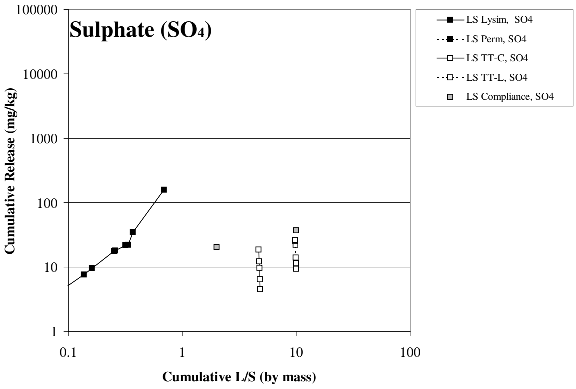


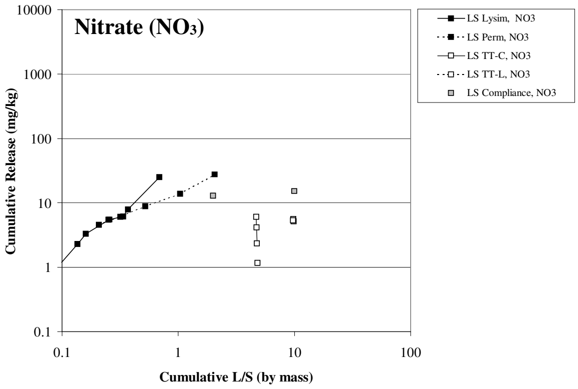


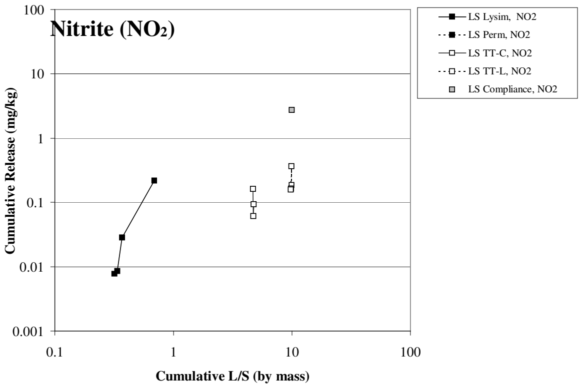
LIMESTONE
Cumulative release (mg/kg) against cumulative L/S ratio

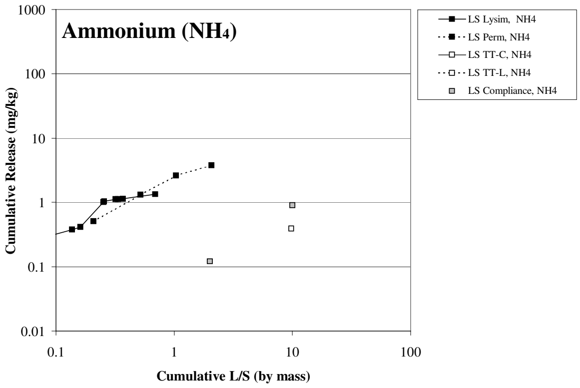




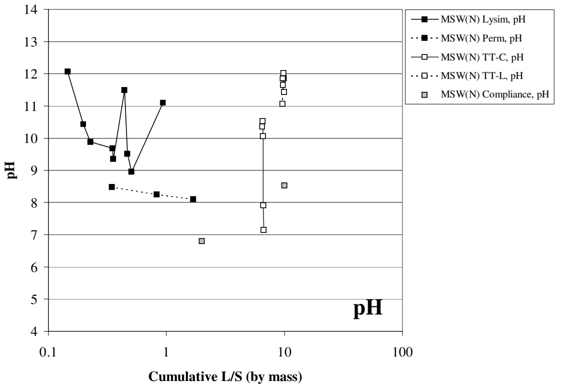


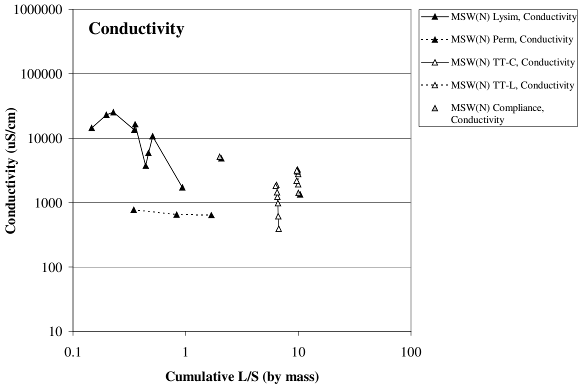


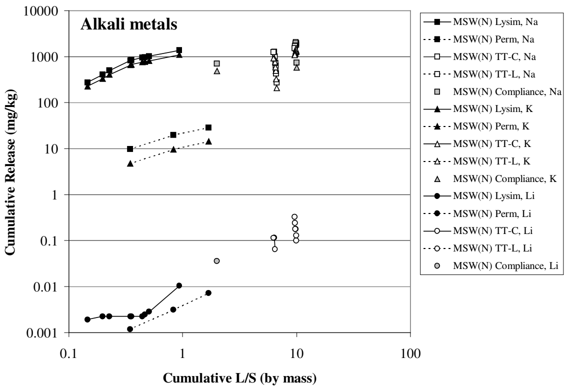


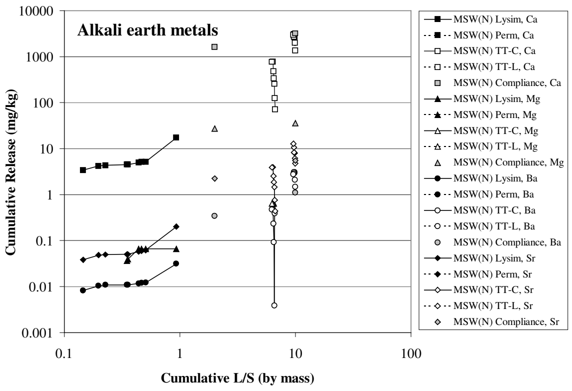


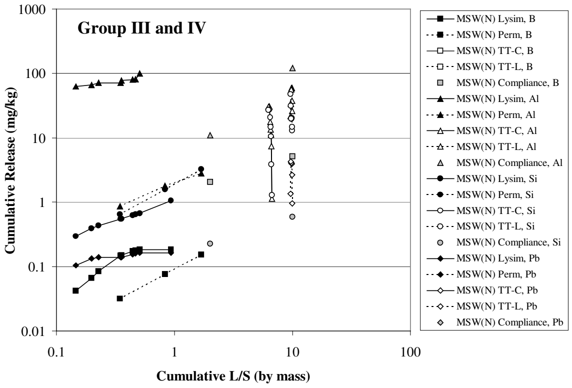
MSW INCINERATOR ASH
Cumulative release (mg/kg) against cumulative L/S ratio

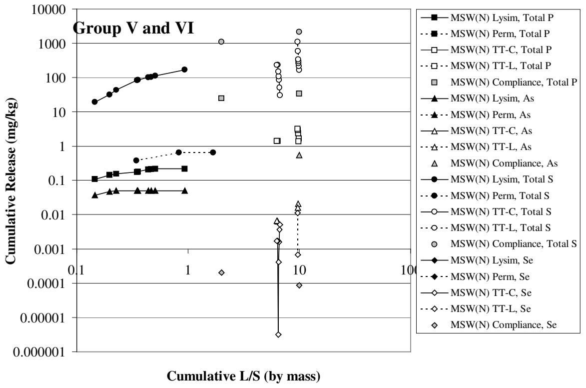


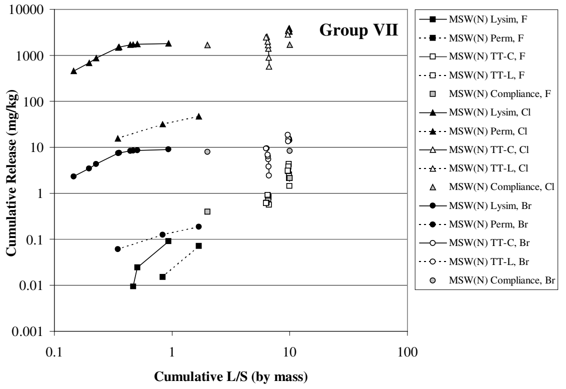


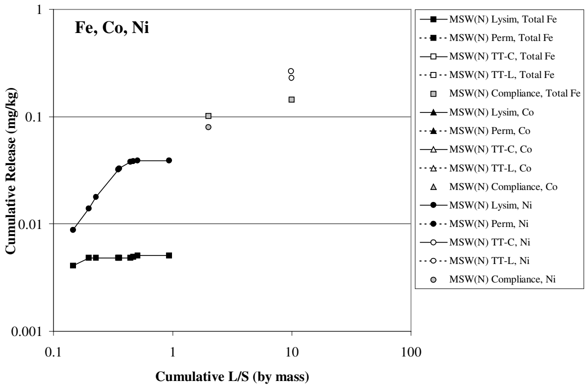




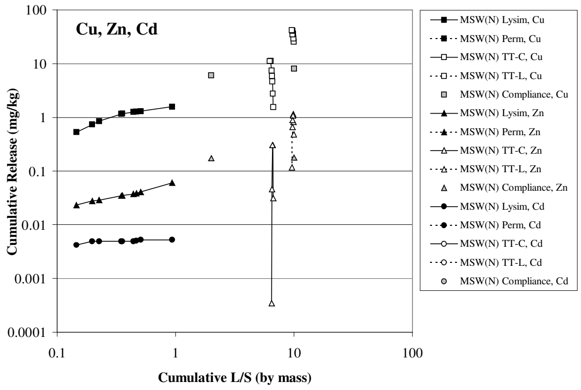


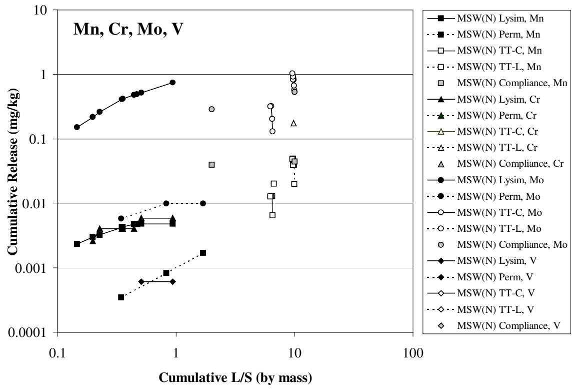


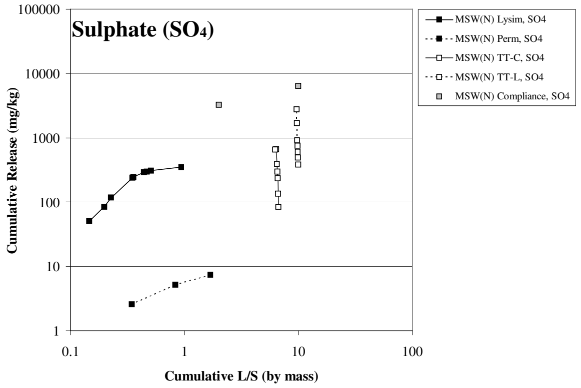


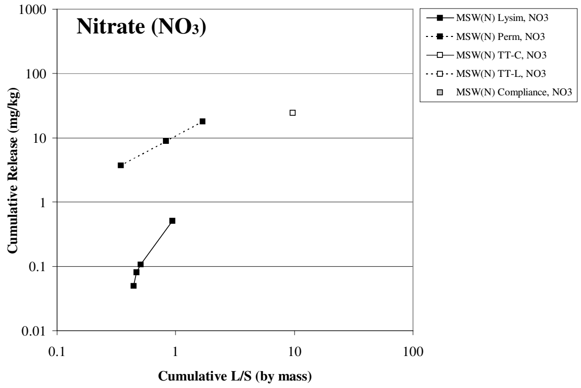


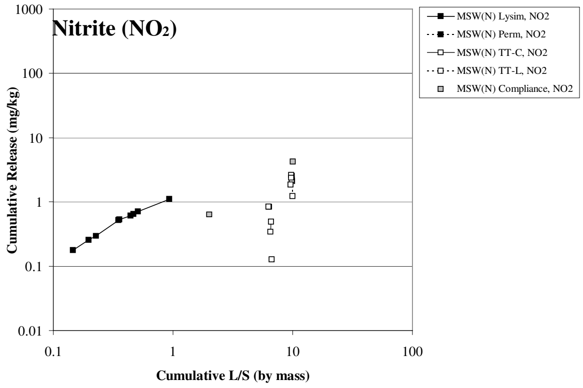
MSW INCINERATOR ASH
Cumulative release (mg/kg) against cumulative L/S ratio

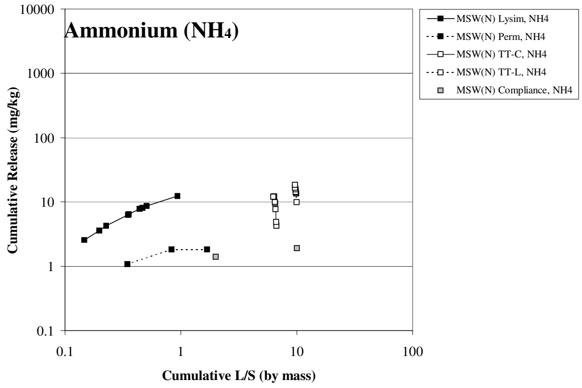




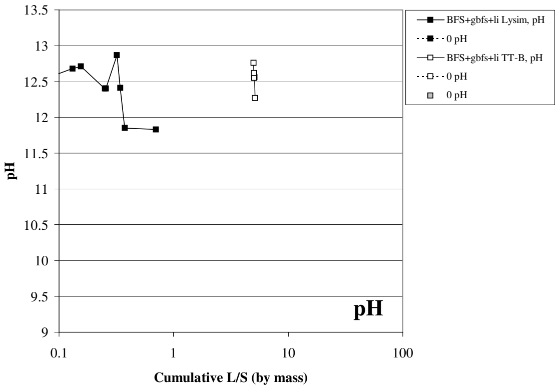


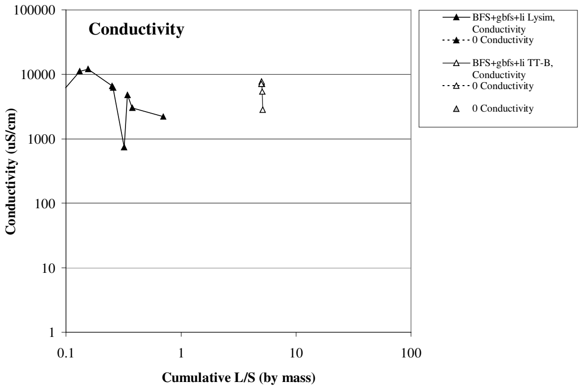


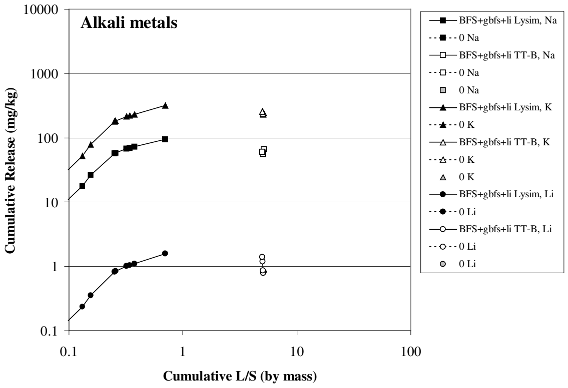


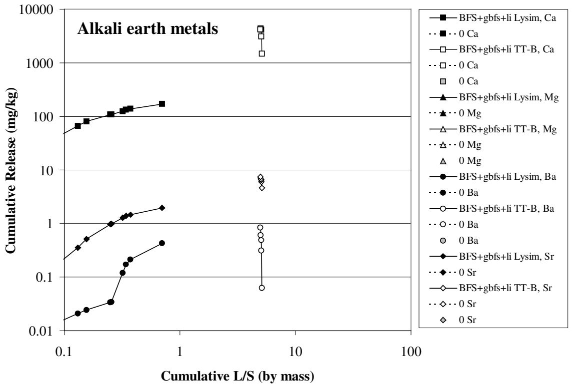


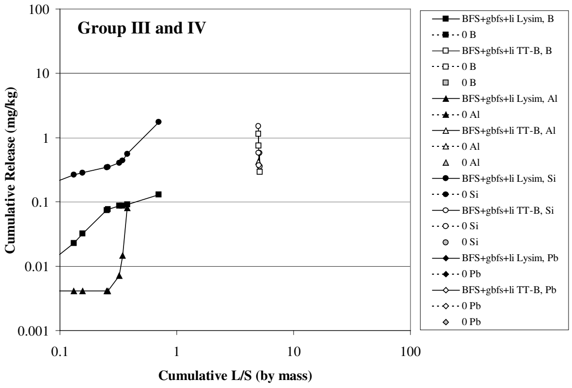
BLASTFURNACE SLAG + granulated blastfurnace slag + lime
Cumulative release (mg/kg) against cumulative L/S ratio

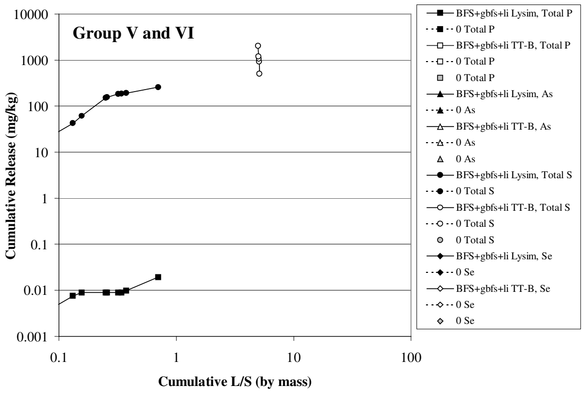


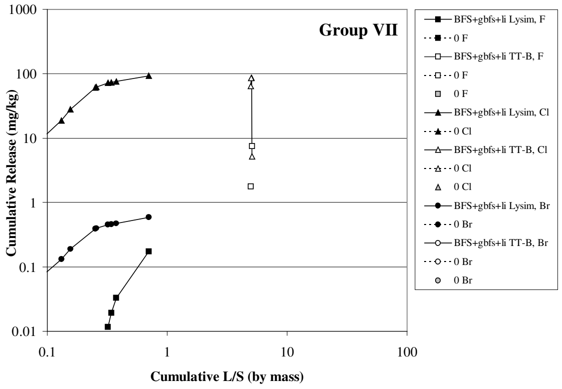


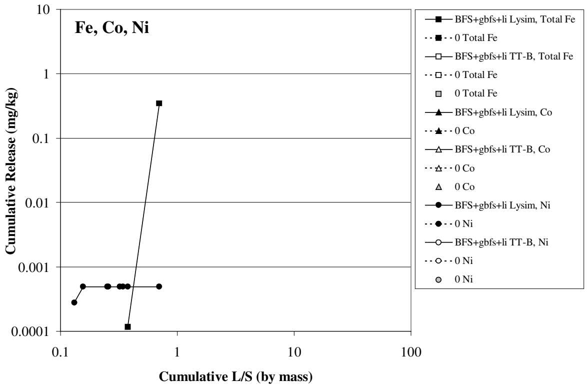




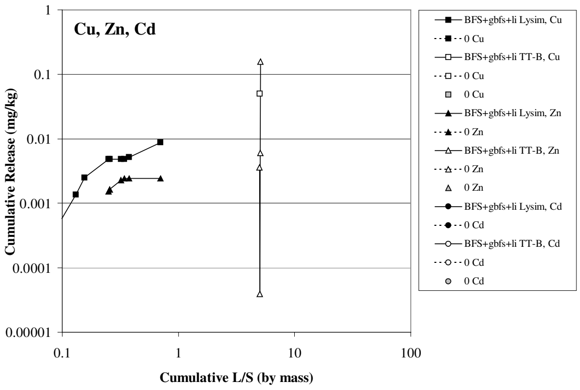


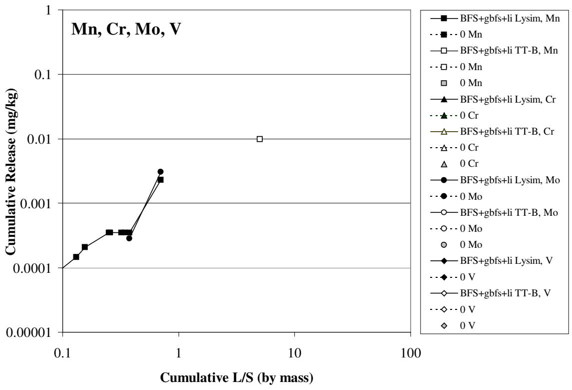


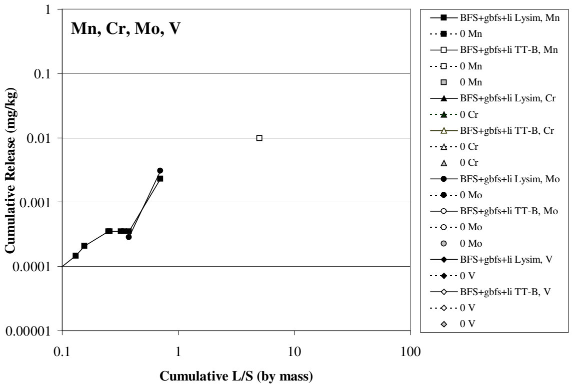


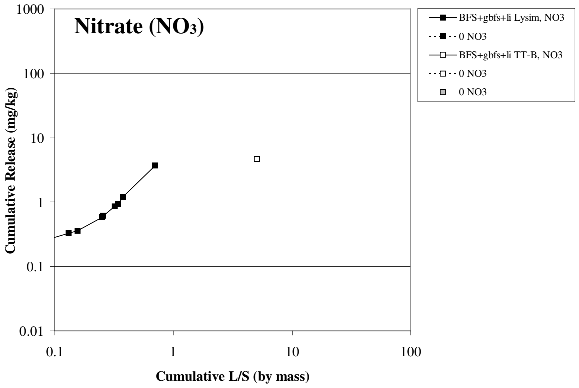


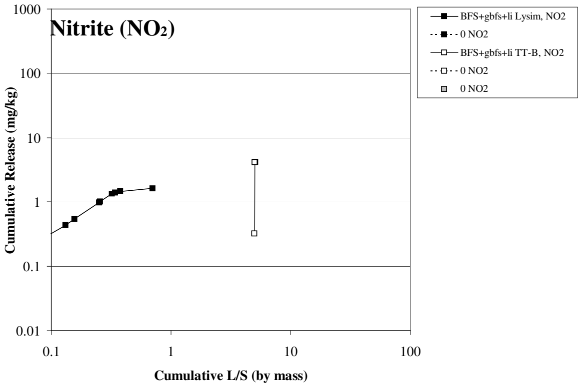
BLASTFURNACE SLAG + granulated blastfurnace slag + lime
Cumulative release (mg/kg) against cumulative L/S ratio

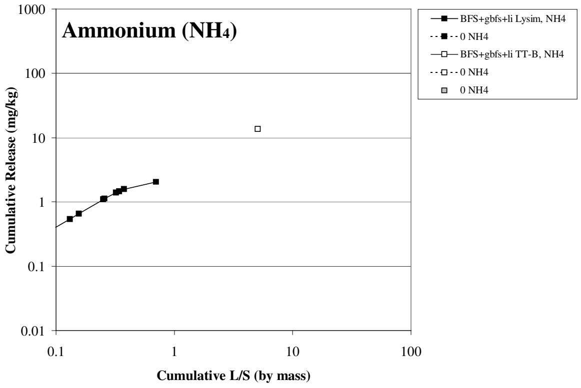




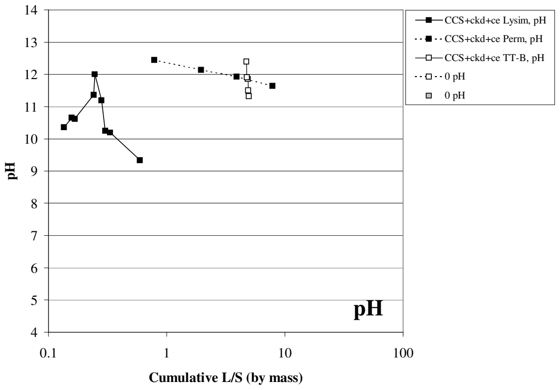


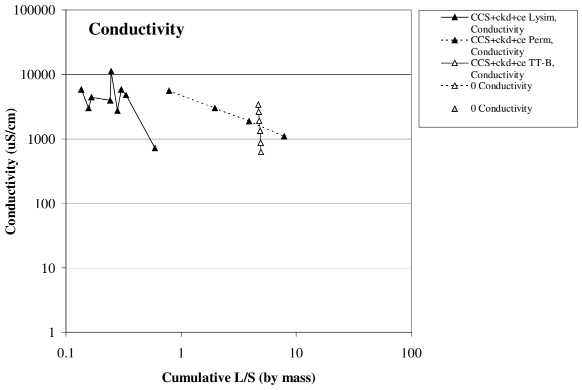


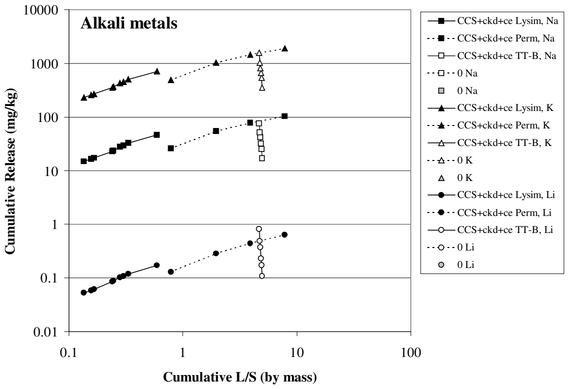


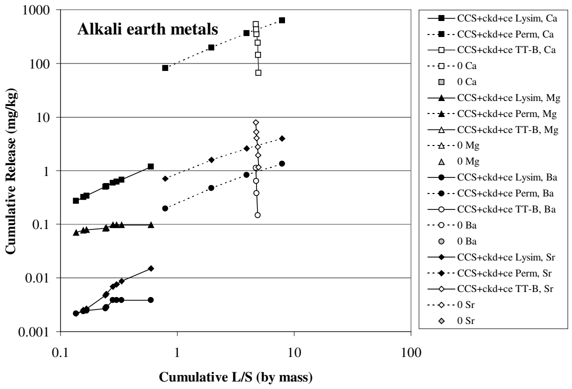


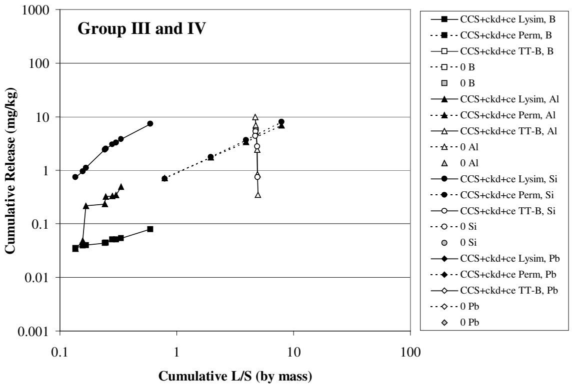
CHINA CLAY SAND + cement kiln dust + cement
Cumulative release (mg/kg) against cumulative L/S ratio

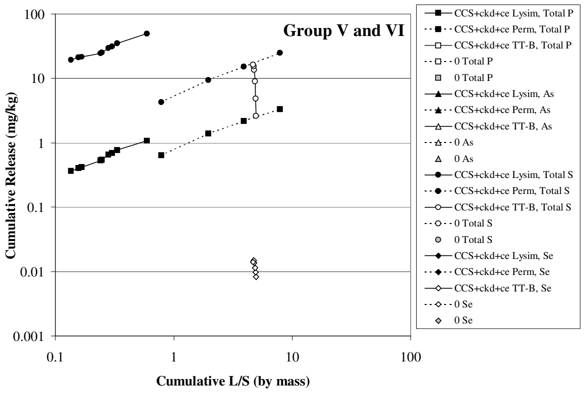


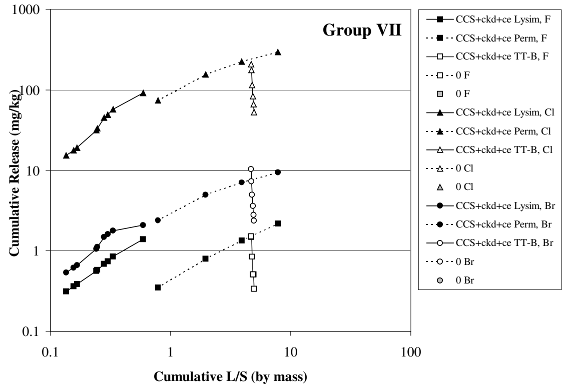


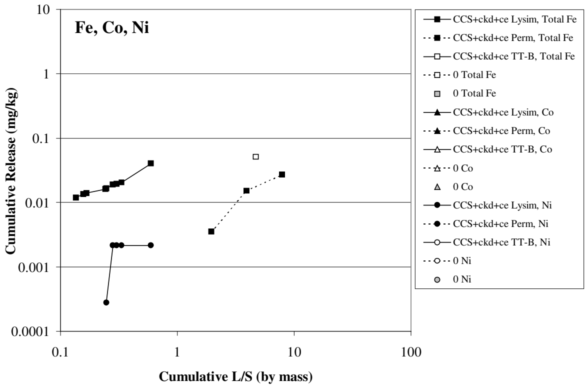




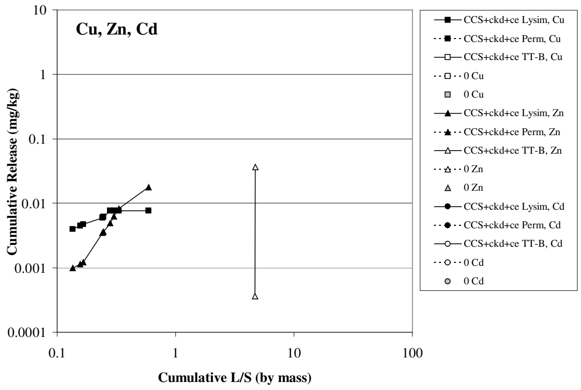


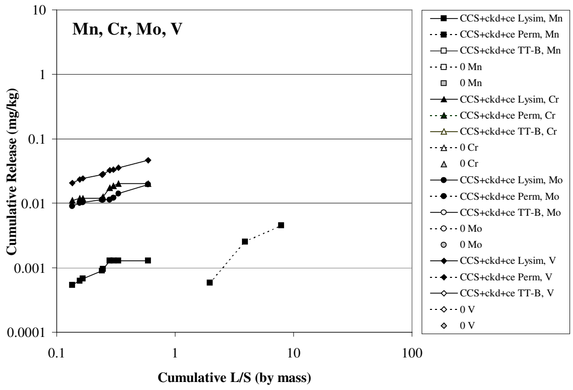


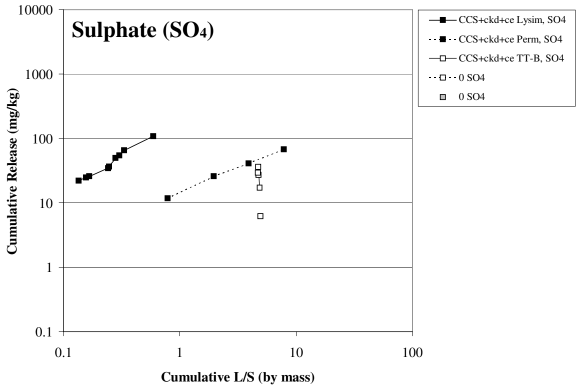


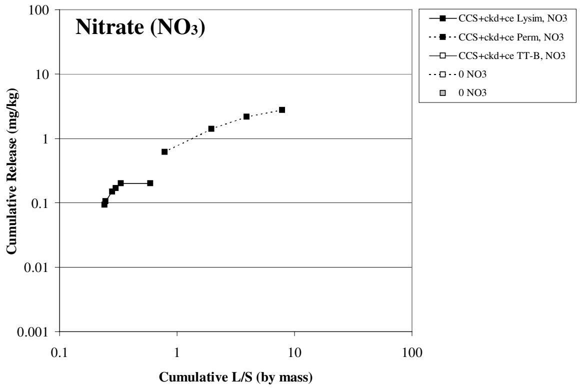


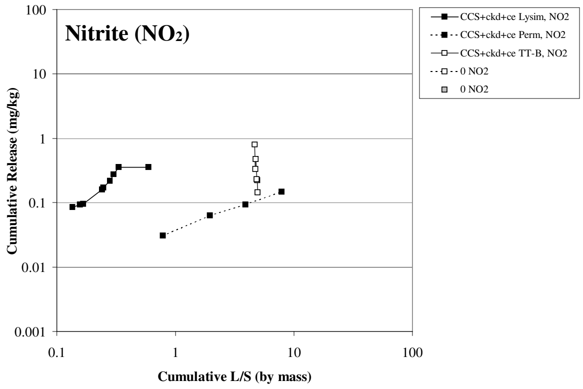
CHINA CLAY SAND + cement kiln dust + cement
Cumulative release (mg/kg) against cumulative L/S ratio

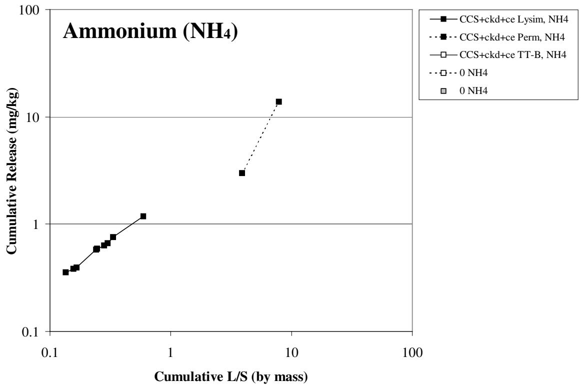




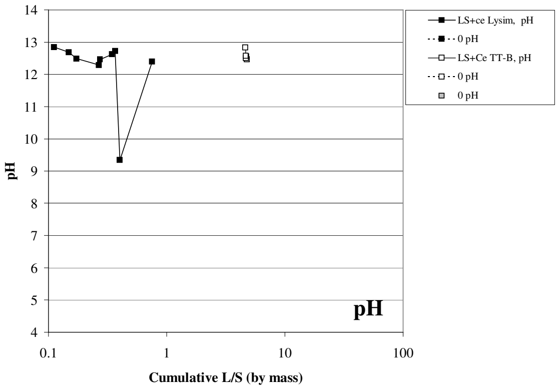


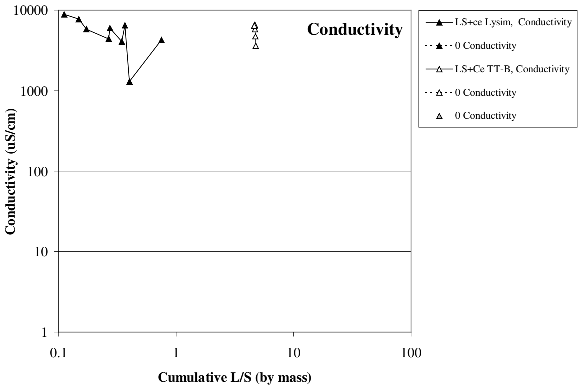


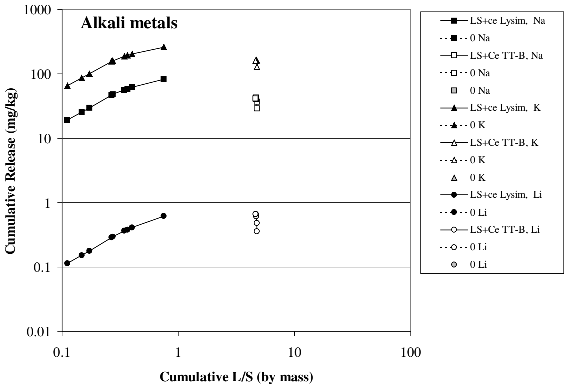


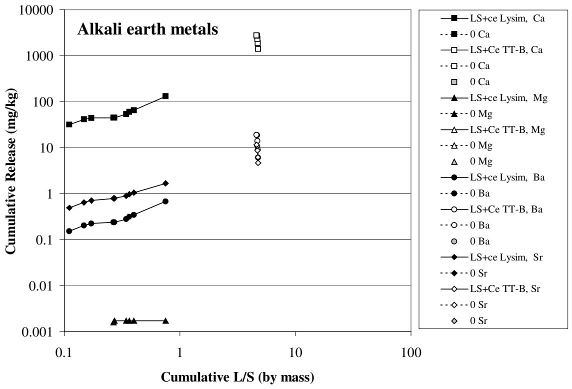


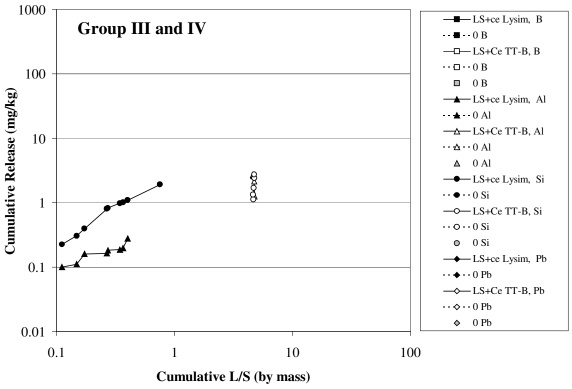
LIMESTONE + cement
Cumulative release (mg/kg) against cumulative L/S ratio

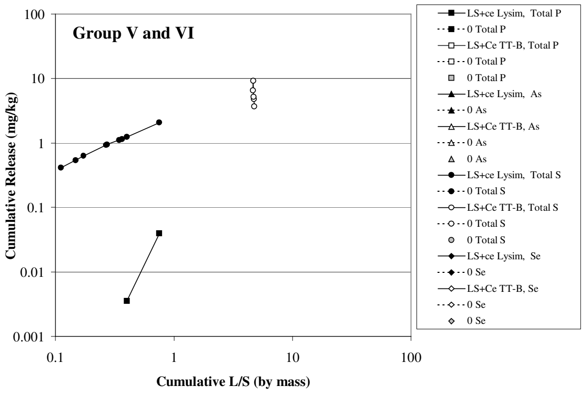


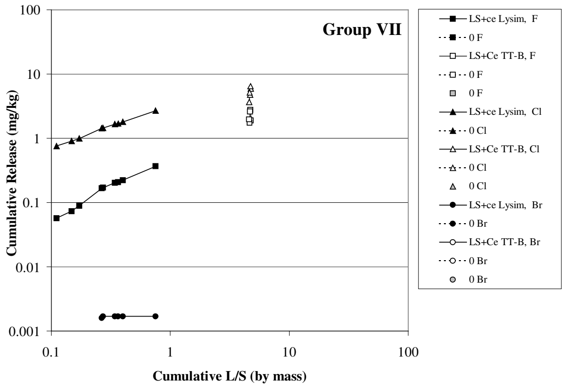


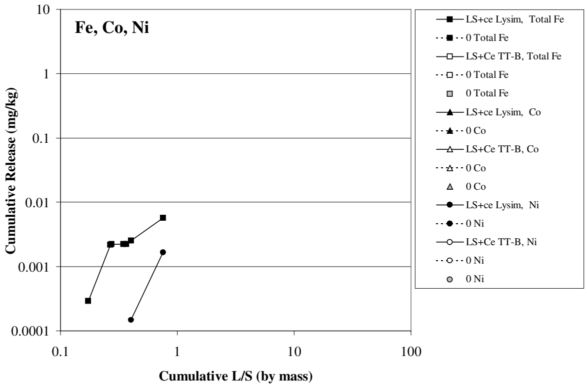




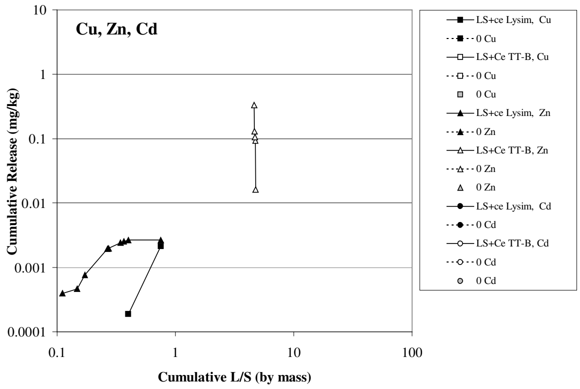


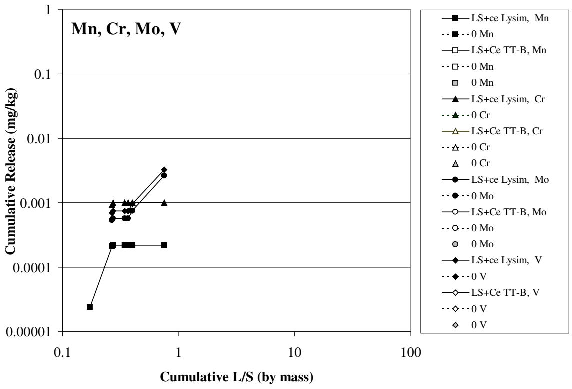


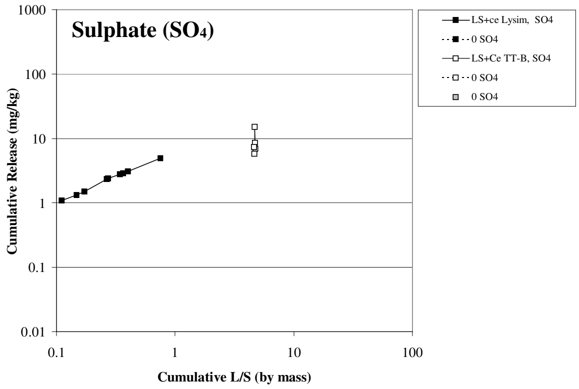


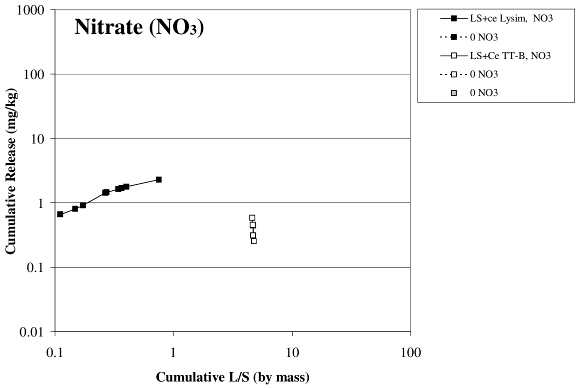


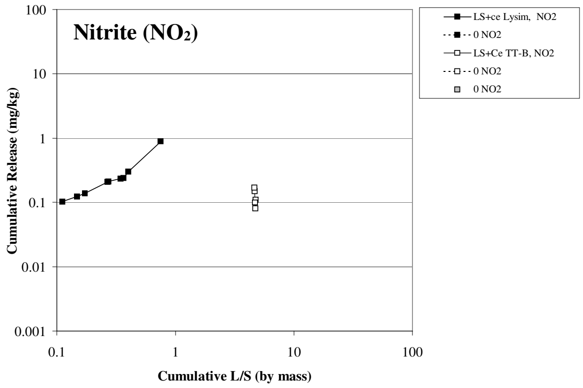
LIMESTONE + cement
Cumulative release (mg/kg) against cumulative L/S ratio

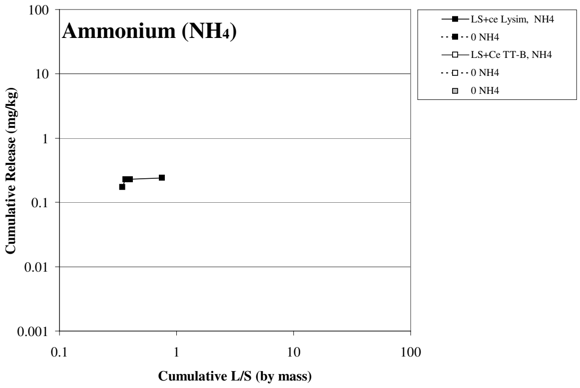




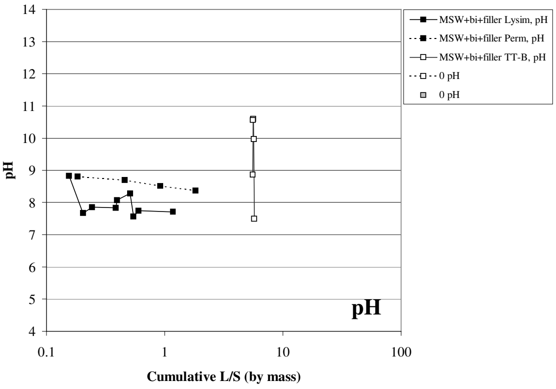


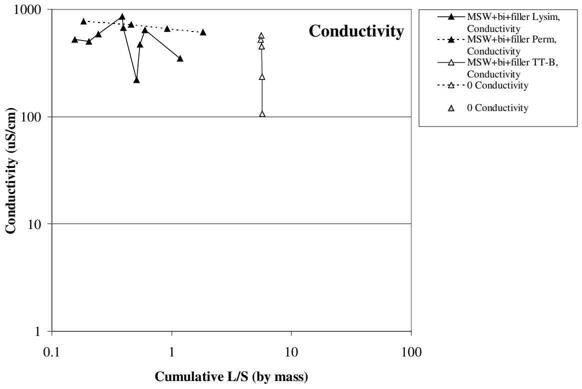


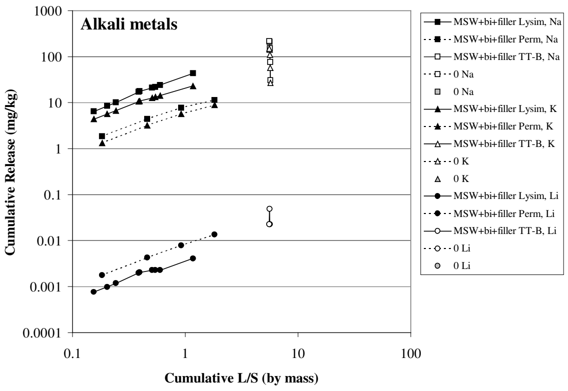


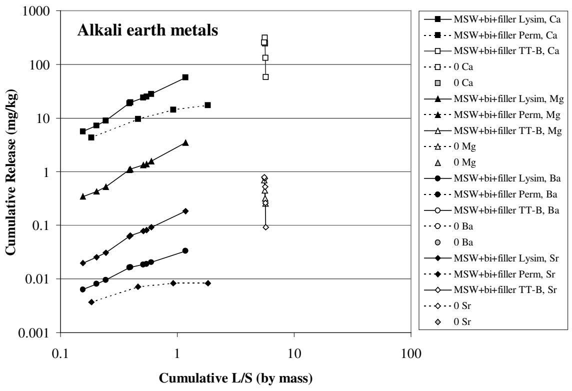


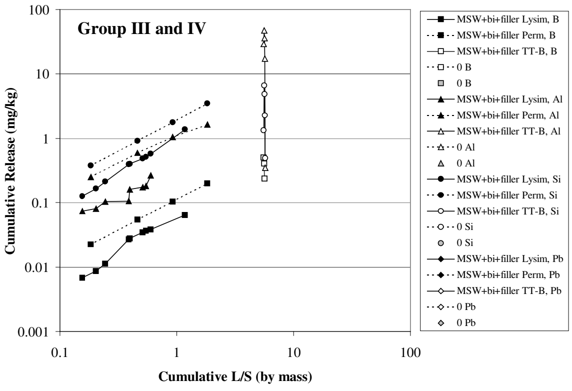
MSW INCINERATOR ASH +bitumen + filler
Cumulative release (mg/kg) against cumulative L/S ratio

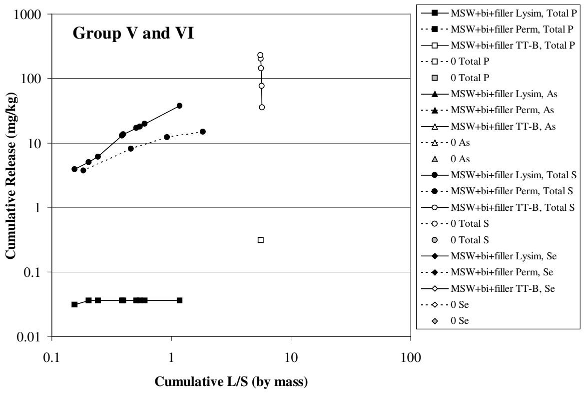


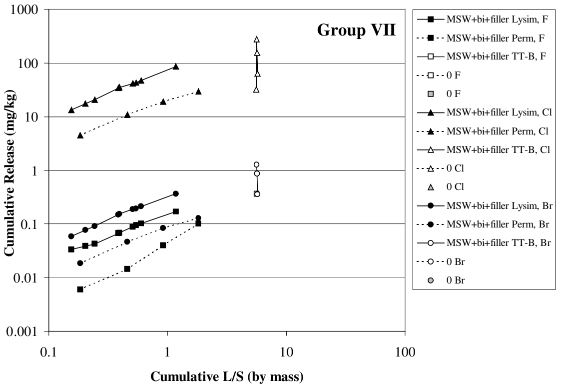


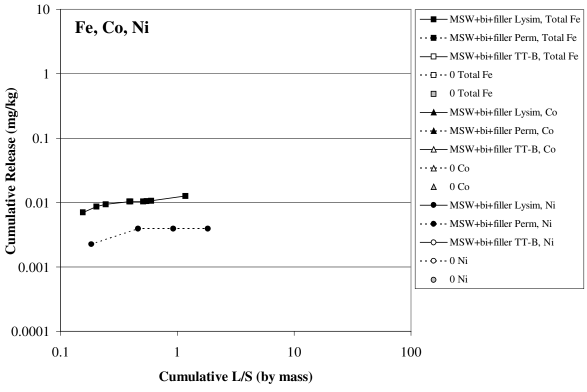




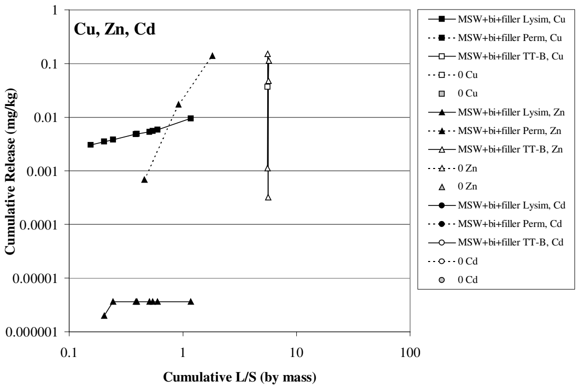


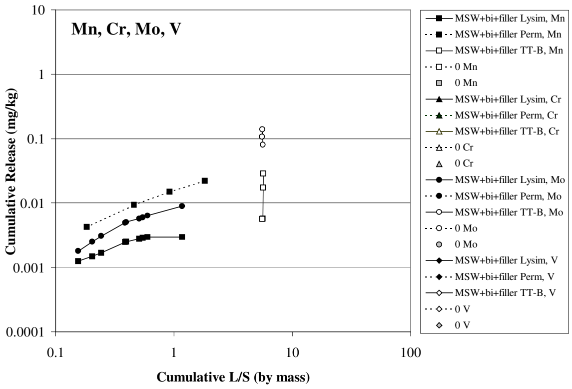


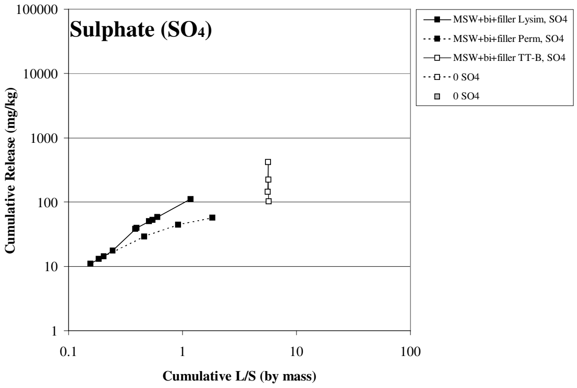


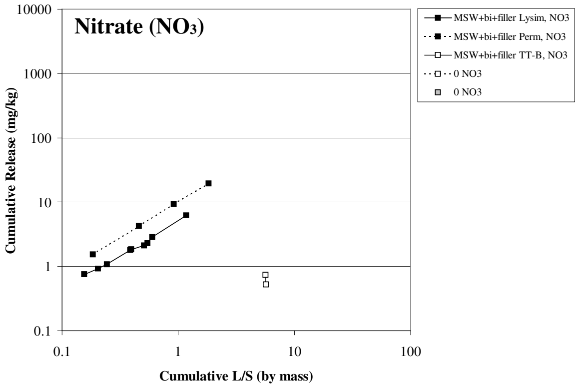


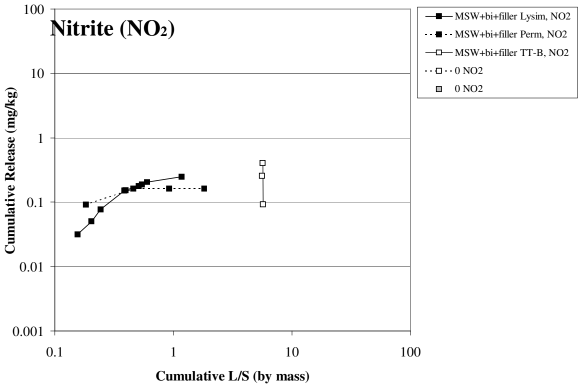
MSW INCINERATOR ASH +bitumen + filler
Cumulative release (mg/kg) against cumulative L/S ratio

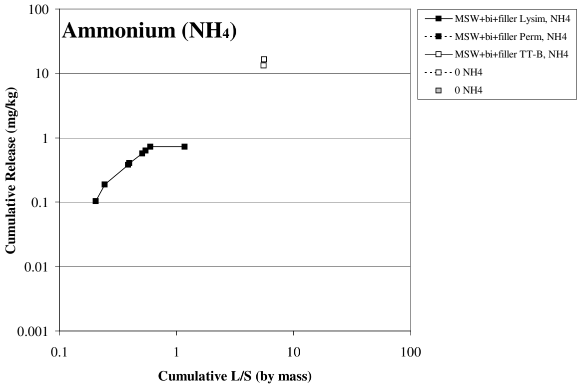












APPENDIX F:

Comparison of laboratory leaching tests and lysimeters

Description of leaching patterns

Leached species concentrations (mg/l) verses L/S ratio

Patterns of Leaching – log concentration (mg/l) verses log L/S (by mass): CCS+ckd+ce

The patterns of leaching for the CCS+ckd+ce mixture are discussed using the data from the following leaching tests:

1. lysimeter
2. permeameter
3. tank-leaching test – bound

Three main patterns of leaching were observed for this material.

CCS+ckd+ce: Pattern 1

Key features:

- The concentrations shown in the leachates from all tests cover a range of 1 to 2 orders of magnitude.
- The three tests show a general overall pattern of gently declining concentrations with increasing L/S (linear on the log-log plot).
- The lysimeter tests show fluctuations in concentration with a general overall slight decline with increasing L/S.
- The highest concentrations are usually shown in the lysimeter test (which is at the lowest L/S). The lowest concentrations are generally shown in the early stages of the tank-leaching test or the later stages of the permeameter test (at the highest L/S). In some cases the permeameter and/or tank-leaching test show concentrations within the range observed in the lysimeter leachate.
- The peak concentrations in the tank-leaching test are higher than the concentration in the permeameter at the same L/S.
- The permeameter test shows a steady decline in concentration with increasing L/S and intersects the plotted concentrations shown in the tank-leaching test at a concentration around the mid-point of the range shown in the tank-leaching test.

Chemical species showing Pattern 1:

- Conductivity (plotted as log conductivity (µS/cm) verses log L/S)
- *Group 1 species:* sodium, potassium and lithium
- *Group VII species:* fluorine, chlorine and bromine
- *Compounds*: HCO_3
- *Other:* Sum of total normalised concentrations

- *Additional chemical species:*

  - Aluminium: shows pattern 1 except that the concentrations measured in the permeameter test are relatively constant in comparison to the declining concentrations observed for the other chemical species.
  - Total phosphorus: shows pattern 1 in the lysimeters and permeameter. The concentrations in the tank-leaching test are below the limits of analytical detection.

CCS+ckd+ce: Pattern 2

Key features:

- The concentrations shown in the leachates from all tests cover a range of up to 3 orders of magnitude.
- The pattern shown in the permeameter and tank-leaching test is the same, relative to each other, as in Pattern 1. The concentration in the permeameter test shows a steady decline with increasing L/S and intersects the plotted concentrations shown in the tank-leaching test at about the mid-point of the range of concentrations shown in the tank-leaching test.
- The concentrations in the lysimeter leachates fluctuate and may show a slight overall decline with increasing L/S.
- The concentrations in the lysimeter leachates are lower than those shown in the laboratory tests by 1 to 2 orders of magnitude, even though the lysimeter is at a lower L/S than the laboratory tests (and theoretically would have higher concentrations).

Chemical species showing Pattern 2:

- pH (plotted as pH verses log L/S, since pH is a logarithmic scale)
- *Group II species:* calcium, strontium and barium

CCS+ckd+ce: Pattern 3

Key features:

- The concentrations shown in the leachates from all tests cover a range of 2 to 3 orders of magnitude.
- The three tests show a general decline in concentrations with increasing L/S.
- The lysimeter test shows fluctuating concentrations that are higher than the concentrations in the laboratory tests by at least 1 order of magnitude.
- The lowest concentrations are generally shown in the early stages of the tank-leaching test or the later stages of the permeameter test (at the highest L/S).
- The permeameter test intersects the plotted concentrations shown in the tank-leaching test.

Chemical species showing Pattern 3:

- *Group 1V species*: Silicon
- *Group V1 species*: Total sulphur
- *Transition elements*: Total iron,
- *Compounds*: SO_4,
- *Other*: TOC, TIC, sum of normalised List I and II species (TIC and sum of normalised List I and II species show fluctuations in the permeameter data that would not be predictable).

- *Additional chemical species:*
  - NO_2: shows a pattern similar to pattern 3 except that the concentrations measured in the tank-leaching test are higher than those in the permeameter test and the concentrations in the lysimeter leachates show a slightly increase with increasing L/S.

CCS+ckd+ce: Chemical species with insufficient data to identify the pattern of leaching

- *Chemical species that were measured above the limits of analytical detection in at least one sample from the lysimeters:*
  - NO_3, NH_4, manganese, zinc, magnesium, nickel, copper, chromium, molybdenum, vanadium and boron.
- *Chemical species that were measured above the limit of analytical detection in at least one laboratory test:*
  - NO_3, NH_4, manganese, zinc and selenium.
- *Chemical species that were not measured above the limits of analytical detection in the leachate from any test:*
  - Cobalt, cadmium, lead and arsenic.

Patterns of Leaching – log concentration (mg/l) verses log L/S (by mass): LS+ce

The patterns of leaching for the LS+ce mixture are discussed using the data from the following leaching tests:

1. lysimeter
2. tank-leaching test – bound

The use of only two tests does limit the discussion and conclusions that can be drawn in terms of the patterns of leaching. However, three main patterns of leaching were observed and are discussed for this material.

LS+ce: Pattern 1

Key features:

- The concentrations shown in the leachates from all tests cover a range of less than 1 order of magnitude.
- The concentrations in the lysimeter leachates show generally small fluctuations (with the odd anomalous data point) and either remain at a fairly constant value or decline very slightly with increasing L/S.
- The concentrations shown in the tank-leaching test are within the range identified in the lysimeter tests (usually towards the upper end of this narrow (1 order of magnitude) range).

Chemical species showing Pattern 1:

- pH (plotted as pH verses log L/S, since pH is a logarithmic scale)
- Conductivity (plotted as log conductivity (µS/cm) verses log L/S)
- *Group II1 species:* Aluminium
- *Group VI species:* Total sulphur
- *Group VII species:* Fluorine, chlorine
- *Compounds*: HCO_3, SO_4

- *Additional chemical species:*
    - Zinc: shows pattern 1 except that the concentrations measured in the tank-leaching test range cover a range of just over 1 order of magnitude and extend above and below the concentrations measured in the lysimeter leachates.
    - Sum of total normalised concentrations: shows pattern 1 except that the lowest concentrations in the tank-leaching test are slightly lower than that of the lysimeter test. This pattern falls between the criteria for pattern 1 and pattern 2.

  - Sum of List I and II normalised concentrations: shows pattern 1 except that the lysimeter data varies by just over 1 order of magnitude and may exhibit a slight increase with increasing L/S. This pattern falls between the criteria for pattern 1 and pattern 3.

LS+ce: Pattern 2

Key features:

- The concentrations shown in the leachates from all tests cover a range of 1 to 2 or 3 orders of magnitude.
- The concentrations in the lysimeter leachates generally show small fluctuations (less than 1 order of magnitude) between samples and the overall trend shows a rapid reduction in concentration of over 1 order of magnitude over the range of L/S shown in the lysimeter test.
- The concentrations shown in the tank-leaching test are either around the lowest concentrations shown in the lysimeter leachates or less, but are at a higher L/S than the lysimeters.

Chemical species showing Pattern 2:

- *Group 1 species:* sodium, potassium and lithium
- *Group IV species*: silicon

- *Additional chemical species:*
  - NO_3 and NO_2: shows pattern 2 except that the decline in concentration shown in the lysimeters is less rapid than that shown for the other chemical species.
  - TOC: shows pattern 2 except that the tank-leaching test data varies over a range of more than 2 orders of magnitude and exhibits concentrations both higher and lower than those shown in the lysimeter.
  - TIC: shows pattern 2 except that the lysimeter shows fluctuations greater than 1 order of magnitude.

LS+ce: Pattern 3

Key features:

- The concentrations shown in the leachates from all tests cover a range of 2 to 3 orders of magnitude.
- The concentrations in the lysimeter leachates show large fluctuations (up to 3 orders of magnitude) between samples and the overall trend shows fairly constant or slightly increasing concentrations with increasing L/S.

- The concentrations shown in the tank-leaching test are around the highest concentrations shown in the lysimeter leachates and may peak at values slightly higher than the peak concentrations shown in the lysimeter.

Chemical species showing Pattern 3:

- *Group 1I species*: calcium, barium and strontium

LS+ce: Chemical species with insufficient data to identify the pattern of leaching

- *Chemical species that were measured above the limits of analytical detection in at least one sample from the lysimeters:*
    - magnesium, Total phosphorus, bromine, Total iron, nickel, copper, chromium, molybdenum, vanadium, NH_4.
- *Chemical species that were measured above the limit of analytical detection in at least one laboratory test:*
    - Bromine.
- *Chemical species that were not measured above the limits of analytical detection in the leachate from any test:*
    - Boron, lead, arsenic, selenium, cobalt, cadmium, and manganese.

Patterns of Leaching – log concentration (mg/l) verses log L/S (by mass): BFS+gbfs+li

The patterns of leaching for the BFS+gbfs+li mixture are discussed using the data from the following leaching tests:

1. lysimeter
2. tank-leaching test – bound

The use of only two tests does limit the discussion and conclusions that can be drawn in terms of the patterns of leaching. However, two main patterns of leaching were observed and are discussed for this material.

BFS+gbfs+li: Pattern 1

Key features:

- The concentrations shown in the leachates from all tests cover a range of approximately 1 order of magnitude.
- The concentrations in the lysimeter leachates generally show only small fluctuations (with the odd anomalous data point) and either remain at a fairly constant value or decline by up to 1 order of magnitude with increasing L/S.
- The concentrations shown in the tank-leaching test are within the range identified in the lysimeter tests.

Chemical species showing Pattern 1:

- pH (plotted as pH verses log L/S, since pH is a logarithmic scale)
- Conductivity (plotted as log conductivity (µS/cm) verses log L/S)
- *Group I1 species:* Strontium
- *Group II1 species:* Boron
- *Group VI species:* Total sulphur
- *Group VII species:* Fluorine
- *Compounds*: HCO_3, NO_3

- *Additional chemical species:*
    - Calcium: shows pattern 1 except that the concentrations in the lysimeter leachates show large fluctuations of over 2 orders of magnitude between measurements.
    - Barium: shows pattern 1 except that the concentrations in the lysimeter leachates show fluctuations and a slight increase with increasing L/S.
    - Transition elements copper, zinc and manganese: indicate that they show pattern 1 although these chemical species are not detected in all samples from each test.

BFS+gbfs+li: Pattern 2

Key features:

- The concentrations shown in the leachates from all tests cover a range of 1 to 3 orders of magnitude.
- The concentrations in the lysimeter leachates show a rapid reduction in concentration of 1 to 2 orders of magnitude over the range of L/S shown in the lysimeter test.
- The concentrations shown in the tank-leaching test are either within the range defined by the lysimeter tests or around the lowest concentrations shown in the lysimeter leachates or less, but are at a higher L/S than the lysimeters.

Chemical species showing Pattern 2:

- *Group 1 species:* sodium, potassium and lithium
- *Group 1V species:* silicon
- *Group VII species:* chlorine, bromine
- *Compounds*: SO_4, NO_2, NH_4
- *Other:* TOC, TIC, sum of total normalised concentrations, sum of List I and II normalised concentrations.

BFS+gbfs+li: Chemical species with insufficient data to identify the pattern of leaching

- *Chemical species that were measured above the limits of analytical detection in at least one sample from the lysimeters:*
    - Aluminium, Total phosphorus, total iron, nickel.
- *Chemical species that were measured above the limit of analytical detection in at least one laboratory test:*
    - Aluminium
- *Chemical species that were not measured above the limits of analytical detection in the leachate from any test:*
    - Magnesium, lead, arsenic, selenium, cobalt, cadmium, chromium, molybdenum and vanadium.

Patterns of Leaching – log concentration (mg/l) verses log L/S (by mass): MSW+bi+filler

The patterns of leaching for the MSW+bi+filler mixture are discussed using the data from the following leaching tests:

1. lysimeter
2. permeameter
3. tank-leaching test – bound

Two main patterns of leaching were observed for this material.

MSW+bi+filler: Pattern 1

Key features:

- The concentrations shown in the leachates from all tests cover a range from less than 1 to approximately 1.5 orders of magnitude.
- The three tests show a general overall pattern of either constant of gently declining (less than 1 order of magnitude) concentrations with increasing L/S (on the log-log plot).
- Fluctuations in the concentrations in the lysimeter in consecutive readings are less than 1 order of magnitude.
- The concentrations in the permeameter leachates are slightly higher (less than 1 order of magnitude) than those in the lysimeter at the same L/S.
- The concentrations in the tank-leaching test are within the range of the lysimeter and permeameter tests.

Chemical species showing Pattern 1:

- Conductivity (plotted as log conductivity (μS/cm) verses log L/S)
- *Group 1 species:* Lithium
- *Group II1 species:* boron
- *Group 1V species:* silicon
- *Transition Elements*: manganese

- *Additional chemical species:*
    - pH (plotted as pH verses log L/S, since pH is a logarithmic scale): shows pattern 1 except that the range of pH shown in the tank-leaching test covers 3 pH units and is higher than that of the lysimeter and permeameter tests.
    - Aluminium: shows pattern 1 except that in the tank-leaching test the concentration varies over 2 orders of magnitude and extends above that measured in the lysimeter and permeameter tests.

  - NO3: shows pattern 1 except the concentrations in the tank-leaching test are lower by approximately 1 order of magnitude.

MSW+bi+filler: Pattern 2

Key features:

- The concentrations shown in the leachates from all tests cover a range of 1 to 2 orders of magnitude.
- The lysimeter and permeameter tests show a general declining in concentrations (less than 1 order of magnitude) with increasing L/S (on the log-log plot).
- Fluctuations in the concentrations in the lysimeter in consecutive readings are less than 1 order of magnitude.
- The concentrations in the permeameter leachates are slightly lower (up to 1 order of magnitude) than those in the lysimeter at the same L/S.
- The concentrations in the tank-leaching test are within the range of the lysimeter and permeameter tests.

Chemical species showing Pattern 2:

- *Group 1 species:* sodium and potassium
- *Group I1 species:* calcium and strontium
- *Group VI species:* Total sulphur
- *Group VII species:* fluorine, chlorine and bromine
- *Compounds*: SO4
- *Other:* Sum of total normalised concentrations

- *Additional chemical species:*
  - Magnesium: shows pattern 2 except that the concentrations in the tank-leaching test are approximately 1 order of magnitude lower than expected and the concentration in the permeameter test was below the limits of analytical detection.
  - Copper, HCO3, NO2, TOC, TIC and NH4: show pattern 2 except that the concentrations in the permeameter test are below the limits of analytical detection.
  - Sum of List I and II normalised concentrations: shows pattern 2 except that the lysimeter data shows large fluctuations between consecutive readings of up to 2 orders of magnitude, and the tank-leaching test data varies over a range of 3 orders of magnitude.

MSW+bi+filler: Chemical species with insufficient data to identify the pattern of leaching

- *Chemical species that were measured above the limits of analytical detection in at least one sample from the lysimeters:*
    - Barium, total phosphorus, total iron, nickel and molybdenum.
- *Chemical species that were measured above the limit of analytical detection in at least one laboratory test:*
    - Total phosphorus, zinc and molybdenum.
- *Chemical species that were not measured above the limits of analytical detection in the leachate from any test:*
    - Lead, arsenic, selenium, cobalt, cadmium, chromium and vanadium.

Patterns of Leaching – log concentration (mg/l) verses log L/S (by mass): foundry sand (phenolic, brass)

The patterns of leaching for the foundry sand (phenolic, brass) are discussed using the data from the following leaching tests:

1. lysimeter
2. permeameter
3. tank-leaching test – compacted
4. tank-leaching test – uncompacted
5. rapid leaching characterisation test

Two main patterns of leaching were observed for this material.

FSB: Pattern 1

Key features:

- The concentrations shown in the leachates from all tests cover a range of between 2 and 4 orders of magnitude.
- The general overall pattern shows a rapid decline in concentrations in the leachates with an increase in L/S (linear on the log-log plot).
- The lysimeter tests show a rapid reduction in concentration of 1 to 2 orders of magnitude over the L/S range of the test of 0.9.
- The concentrations in the permeameter test continue from the end of the lysimeter test. The concentrations continue to decline over about 1 order of magnitude from L/S of 1 to 23.
- The concentrations in the tank-leaching tests range over less than 1 order of magnitude that show rapid establishment of concentrations that are maintained or even decrease with increasing time.
- The tank-leaching tests and the rapid leaching characterisation test represent the ‘average’ concentrations of the lysimeter and permeameter tests over the L/S range between 0 and the L/S of the tank-leaching test or rapid leaching characterisation test.
- Concentrations in the tank-leaching tests and rapid leaching characterisation test are within the range shown in the lysimeter and permeameter tests.
- The gradient shown by the concentrations in the rapid leaching characterisation test is similar to the gradient of the lysimeter and permeameter tests.

Chemical species showing Pattern 1:

- Conductivity (plotted as log conductivity (µS/cm) verses log L/S)

- *Group 1 species:* sodium, potassium and lithium [except lithium shows no permeameter or rapid leaching characterisation test concentrations above the limit of analytical detection].
- *Group II species:* calcium, magnesium, strontium and barium [none of the Group II species show concentrations above the limit of analytical detection in the permeameter tests]
- *Group VI species:* total phosphorus [except that the lysimeter concentrations remain relatively constant but fluctuate over less than 1 order of magnitude]
- *Group VI species:* total sulphur
- *Group VII species:* fluorine and chlorine
- *Transition Elements*: total iron, manganese and zinc [except no permeameter data for zinc]
- *Compounds*: HCO_3, SO_4 [except no permeameter data], TOC [except very steep lysimeter and permeameter concentrations] and TIC
- *Other:* sum of total normalised concentrations, sum of List I and II normalised concentrations [except very steep decline in permeameter]

- *Additional chemical species:*
  - pH (plotted as pH verses log L/S, since pH is a logarithmic scale): shows a pattern similar to pattern 1 except that the pH in the permeameter tests is 3 to 4 pH units above that which is expected for this pattern.

FSB: Pattern 2

Key features:

- The concentrations shown in the leachates from all tests cover a range of 2 to 3 orders of magnitude.
- The concentration in the lysimeter leachates is 1 to 2 orders of magnitude higher than in the laboratory leaching tests and the concentration remains relatively constant over the whole L/S ratio of the test with only small (less than 0.5 orders of magnitude) fluctuations.
- The concentrations in the permeameter tests are also relatively constant or decline slightly with increasing L/S.
- The permeameter concentrations intersect the range of tank-leaching test concentrations at the same L/S.
- The tank-leaching test concentrations peak at a concentration between that shown in the lysimeter and permeameter tests.

- The concentrations in the rapid leaching characterisation test are within the range of the lysimeter and permeameter tests.

Chemical species showing Pattern 2:

- *Group II1 species:* aluminium [except the concentrations in the lysimeter are below the limits of analytical detection]
- *Group 1V species:* silicon
- *Transition Elements*: copper

FSB: Chemical species with insufficient data to identify the pattern of leaching

- *Chemical species that were measured above the limits of analytical detection in at least one sample from the lysimeters:*
  - Bromine, cobalt, nickel, molybdenum, vanadium, NO_2 and NH_4
- *Chemical species that were measured above the limit of analytical detection in at least one laboratory test:*
  - Boron, arsenic, selenium, bromine, nickel, vanadium, NO_3, NO_2 and NH_4
- *Chemical species that were not measured above the limits of analytical detection in the leachate from any test:*
  - Lead, cadmium and chromium

Patterns of Leaching – log concentration (mg/l) verses log L/S (by mass): MSW incinerator ash (new incinerator)

The patterns of leaching for the MSW incinerator ash (new incinerator) are discussed using the data from the following leaching tests:

1. lysimeter
2. permeameter
3. tank-leaching test – compacted
4. tank-leaching test – uncompacted
5. rapid leaching characterisation test

Three main patterns of leaching were observed for this material.

MSW(N): Pattern 1

Key features:

- The concentrations shown in the leachates from all tests cover a range of between 2 and 3 orders of magnitude.
- The general overall pattern shows a decline in concentrations in the leachates with an increase in L/S.
- The lysimeter tests show a rapid reduction in concentration of 1 to 2 orders of magnitude over the L/S range of the test of 0.9. The concentrations in the lysimeters fluctuate by up to 1 order of magnitude between consecutive readings.
- The concentrations in the permeameter test are lower than the lowest concentrations in the lysimeter test. The concentrations in the permeameter show a slight decline with increasing L/S.
- The concentrations in the tank-leaching tests generally range over less than 1 order of magnitude.
- The compacted tank-leaching test has a lower peak concentration than the uncompacted tank-leaching test.
- The tank-leaching tests and the rapid leaching characterisation test represent the 'average' concentrations of the lysimeter test over the L/S range between 0 and the L/S of the tank-leaching test or rapid leaching characterisation test.
- Concentrations in the tank-leaching tests are within or below the range shown in the lysimeter and permeameter tests.
- Concentrations in the rapid leaching characterisation test are within the range shown in the lysimeter and permeameter tests.
- The gradient shown by the concentrations in the rapid leaching characterisation test is similar to the gradient of the lysimeter test.

Chemical species showing Pattern 1:

- Conductivity (plotted as log conductivity (µS/cm) verses log L/S)
- *Group 1 species:* sodium and potassium
- *Group II1 species:* aluminium
- *Group VI species:* total sulphur
- *Group VII species:* chlorine, bromine and fluorine [except the fluorine shows increasing concentrations with increasing L/S in the permeameter]
- *Transition Elements*: copper, manganese and molybdenum
- *Compounds*: SO_4 and NH4. HCO_3, NO2, TOC and TIC [except no permeameter data]
- *Other:* sum of total normalised concentrations, sum of List I and II normalised concentrations

- *Additional chemical species:*
    - pH (plotted as pH verses log L/S, since pH is a logarithmic scale): shows a pattern similar to pattern 1 except that the data is spread over a wider range than that expected for this pattern with the tests covering a range of 5 pH units.

MSW(N): Pattern 2

Key features:

- The concentrations shown in the leachates from all tests cover a range of approximately 1 order of magnitude.
- The general overall pattern shows relatively constant concentrations in the leachates with increasing L/S.
- The lysimeter and permeameter tests show fairly steady or slightly increasing concentrations with increasing L/S.
- The concentrations in the tank-leaching tests generally range over less than 1 order of magnitude. The peak concentrations in the tank-leaching tests are similar to the concentrations in the lysimeter.

Chemical species showing Pattern 2:

- *Group I species*: lithium
- *Group II1 species:* boron
- *Group 1V species:* silicon
- *Group V species:* total phosphorus
- *Transition Elements*: zinc

MSW(N): Pattern 3

Key features:

- The concentrations shown in the leachates from all tests cover a range of 2 to 3 orders of magnitude.
- The concentrations in the lysimeter leachates are lower than in the tank-leaching tests and rapid leaching characterisation test. The concentrations in the lysimeter leachates fluctuate by up to 2 orders of magnitude between consecutive readings and may show a general slight increase with increasing L/S.
- The minimum concentration in the lysimeter leachates is up to 2 orders of magnitude lower than in the minimum in the tank-leaching tests. The maximum concentration in the lysimeter leachates is less than 1 order of magnitude lower than the peak concentrations in the tank-leaching tests.
- The chemical species are all at concentrations below the limit of analytical detection in the permeameter tests.
- The concentrations in the tank-leaching tests generally range over less than 1 order of magnitude.
- The compacted tank-leaching test has a lower peak concentration than the uncompacted tank-leaching test.

Chemical species showing Pattern 3:

- *Group II species:* calcium, strontium and barium

MSW(N): Chemical species with insufficient data to identify the pattern of leaching

- *Chemical species that were measured above the limits of analytical detection in at least one sample from the lysimeters:*
    - Magnesium, lead, arsenic, total iron, nickel, cadmium, chromium, vanadium and NO_3
- *Chemical species that were measured above the limit of analytical detection in at least one laboratory test:*
    - Magnesium, lead, arsenic, selenium, total iron, nickel, chromium, vanadium and NO_3
- *Chemical species that were not measured above the limits of analytical detection in the leachate from any test:*
    - Cobalt

Patterns of Leaching – log concentration (mg/l) verses log L/S (by mass): limestone

The patterns of leaching for the limestone are discussed using the data from the following leaching tests:

1. lysimeter
2. permeameter
3. tank-leaching test – compacted
4. tank-leaching test – uncompacted
5. rapid leaching characterisation test

Three main patterns of leaching were observed for this material.

LS: Pattern 1

Key features:

- The concentrations shown in the leachates from all tests cover a range of between 1 and 2 orders of magnitude.
- The general overall pattern shows relatively constant concentrations in the leachates with a slight decline of approximately 1 order of magnitude with an increase in L/S.
- The lysimeter tests show a slight reduction in concentration of up to 1 order of magnitude over the L/S range of the test of 0.7. The concentrations in the lysimeters fluctuate by up to 1 order of magnitude between consecutive readings.
- The concentrations in the permeameter test are either relatively constant or show a slight decline with increasing L/S. The concentrations in the permeameter are generally similar to those of the lysimeter test but may be slightly higher or lower.
- The concentrations in the tank-leaching tests generally range over less than 1 order of magnitude. In a number of cases that range may extend to 2 orders of magnitude.
- The compacted tank-leaching test generally has a peak at a similar or slightly higher concentration than in the uncompacted tank-leaching test.
- The tank-leaching tests and the rapid leaching characterisation test represent the approximate concentrations that would be predicted for the lysimeter test at the same L/S. This may also represent the ‘average’ concentrations since the gradient of the lysimeter data is shallow.
- The gradient shown by the concentrations in the rapid leaching characterisation test is similar to the gradient of the lysimeter test.

Chemical species showing Pattern 1:

- Conductivity (plotted as log conductivity (μS/cm) verses log L/S)
- *Group 1 species*: potassium and lithium

- *Group 1I species*: calcium [except no permeameter data] and strontium [except the permeameter data is lower than expected]
- *Group II1 species*: boron
- *Group IV species:* silicon
- *Group VII species:* chlorine and fluorine
- *Transition Elements*: zinc and manganese
- *Compounds*: NO_2, and HCO_3 and TIC [except no permeameter data for HCO_3 and TIC]
- *Other:* sum of total normalised concentrations, sum of List I and II normalised concentrations

- *Additional chemical species:*
  - pH (plotted as pH verses log L/S, since pH is a logarithmic scale): shows a pattern similar to pattern 1 except that the data is spread over a wider range than that expected for this pattern with the tank-leaching tests approximately 1 pH unit above expected and the rapid leaching characterisation test approximately 1 pH unit below expected.

LS: Pattern 2

Key features:

- The concentrations shown in the leachates from all tests cover a range of between 2 and 3 orders of magnitude.
- The general overall pattern shows a rapid decline in concentrations in the leachates with an increase in L/S.
- The lysimeter tests show a rapid reduction in concentration of 1 to 2 orders of magnitude over the L/S range of the test of 0.7.
- The concentrations in the permeameter test are mainly below the limits of analytical detection. Where chemical species are detected they are of a similar concentration to that of the lysimeter at the same L/S.
- The concentrations in the tank-leaching tests range over less than 1 order of magnitude and show rapid establishment of concentrations.
- The tank-leaching tests and the rapid leaching characterisation test represent the ‘average’ concentrations of the lysimeter test over the L/S range between 0 and the L/S of the tank-leaching test or rapid leaching characterisation test if the lysimeter test data is extrapolated to such L/S.

- The gradient shown by the concentrations in the rapid leaching characterisation test and the peak concentrations in the tank-leaching tests is similar to the gradient of the lysimeter test (on the log-log plot).

Chemical species showing Pattern 2:

- *Group I species*: sodium
- *Group II species*: magnesium
- *Group V1 species*: total sulphur
- *Compounds*: SO_4, NO_3 and NH_4
- *Other*: TOC

LS: Pattern 3

Key features:

- The concentrations shown in the leachates from all tests cover a range of 1 to 2 orders of magnitude.
- The general overall pattern shows an increase in concentrations in the leachates with an increase in L/S.
- The lysimeter tests show an increase in concentration of approximately 1 order of magnitude over the L/S range of the test of 0.7.
- The chemical species in the permeameter tests are at concentrations below the limit of analytical detection.
- The concentrations in the tank-leaching tests range over less than 1 order of magnitude.
- The compacted tank-leaching test has a lower peak concentration than the uncompacted tank-leaching test.
- The gradient shown by the concentrations in the rapid leaching characterisation test and the peak concentrations in the tank-leaching tests is similar to the gradient of the lysimeter test (on the log-log plot).

Chemical species showing Pattern 3:

- *Group II species:* barium

LS: Chemical species with insufficient data to identify the pattern of leaching

- *Chemical species that were measured above the limits of analytical detection in at least one sample from the lysimeters:*
    - Bromine, total iron, cobalt, copper and molybdenum.

- *Chemical species that were measured above the limit of analytical detection in at least one laboratory test:*
    - Aluminium, total phosphorus, arsenic, selenium, bromine and total iron.
- *Chemical species that were not measured above the limits of analytical detection in the leachate from any test:*
    - Lead, nickel, cadmium, chromium and vanadium.

Patterns of Leaching – log concentration (mg/l) verses log L/S (by mass): blastfurnace slag

The patterns of leaching for the blastfurnace slag are discussed using the data from the following leaching tests:

1. lysimeter
2. permeameter
3. tank-leaching test – compacted
4. tank-leaching test – uncompacted
5. rapid leaching characterisation test

Two main patterns of leaching were observed for this material.

BFS: Pattern 1

Key features:

- The concentrations shown in the leachates from all tests cover a range from less than 1 to approximately 1.5 orders of magnitude.
- The general overall pattern shows relatively constant concentrations in the leachates with a slight decline of approximately 1 order of magnitude with an increase in L/S.
- The lysimeter tests show relatively constant concentrations with a slight reduction of up to 0.5 orders of magnitude over the L/S range of the test of 0.8. The concentrations in the lysimeters only show small fluctuattions between consecutive readings.
- The concentrations in the permeameter test show a slight decline with increasing L/S. The gradient may be similar to the lysimeter or slightly steeper.
- The concentrations in the permeameter test are generally up to 0.5 orders of magnitude lower than in the lysimeter test at the same L/S.
- The concentrations in the tank-leaching tests generally range over less than 1 order of magnitude.
- The compacted tank-leaching test has a slightly higher peak concentration than the uncompacted tank-leaching test.
- The tank-leaching tests and the rapid leaching characterisation test represent the approximate concentrations that would be predicted for the lysimeter and permeameter tests at the same L/S. This may also represent the 'average' concentrations since the gradient of the lysimeter and permeameter data is shallow.
- The gradient shown by the concentrations in the tank-leaching tests and rapid leaching characterisation test is similar to the gradient of the lysimeter and permeameter tests.

Chemical species showing Pattern 1:

- Conductivity (plotted as log conductivity (µS/cm) verses log L/S)
- *Group 1I species*: calcium, strontium, magnesium and barium
- *Group II1 species*: aluminium and boron
- *Group IV species:* silicon
- *Group VI species:* total sulphur
- *Group VII species:* fluorine and bromine
- *Transition Elements*: zinc [except no rapid leaching characterisation test data] and manganese
- *Compounds*: HCO_3, SO4, NO_2, NH4, TOC and TIC
- *Other:* sum of total normalised concentrations, sum of List I and II normalised concentrations

- *Additional chemical species:*
  - pH (plotted as pH verses log L/S, since pH is a logarithmic scale): shows a pattern similar to pattern 1 except that the data is spread over a wider range than that expected for this pattern with the tank-leaching tests up to 2 pH units above expected and the rapid leaching characterisation test up to 1 pH unit below expected.

BFS: Pattern 2

Key features:

- The concentrations shown in the leachates from all tests cover a range of between 2 and 3 orders of magnitude.
- The general overall pattern shows a rapid decline in concentrations in the leachates with an increase in L/S.
- The lysimeter tests show a rapid reduction in concentration of approximately 1 order of magnitude over the L/S range of the test of 0.8.
- The concentrations in the permeameter test decline rapidly by approximately 1 order of magnitude. The concentrations in the permeameter are similar to those in the lysimeter at the same L/S.
- The concentrations in the tank-leaching tests generally range over less than 1 order of magnitude and shows rapid establishment of concentrations that are maintained.
- The compacted tank-leaching test has a higher peak concentration than the uncompacted tank-leaching test.
- The gradient shown by the concentrations in the tank-leaching tests and rapid leaching characterisation test is similar to the gradient of the lysimeter and permeameter tests.

- The tank-leaching tests and the rapid leaching characterisation test represent the 'average' concentrations of the lysimeter and permeameter tests over the L/S range between 0 and the L/S of the tank-leaching test or rapid leaching characterisation test.

Chemical species showing Pattern 2:

- *Group I species*: sodium, potassium and lithium
- *Group VI1 species*: chlorine

BFS: Chemical species with insufficient data to identify the pattern of leaching

- *Chemical species that were measured above the limits of analytical detection in at least one sample from the lysimeters:*
    - Total phosphorus, total iron, nickel, copper, molybdenum, vanadium and NO_3
- *Chemical species that were measured above the limit of analytical detection in at least one laboratory test:*
    - Selenium, vanadium and NO_3
- *Chemical species that were not measured above the limits of analytical detection in the leachate from any test:*
    - Lead, arsenic, cobalt, cadmium and chromium