Systematic leaching behaviour of worldwide MSWI bottom ashes in spite of their variability in content

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2013
ECN-B--14-003

IWWG Task group on ???????????????

Sustainable landfilling

edited by Raffaello Cossu, Hans van der Sloot

CISA

Published by CISA, an imprint of Eurowaste Srl
Via Beato Pellegrino 23, 35137 Padova, Italy

First edition 2013

Editors R Cossu, University of Padova, Italy
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Visual organization Daniele Barbiero, Zurich, Switzerland
Typography Cleup sc "Coop. Libraria Editrice Università di Padova"
Printing Cleup sc "Coop. Libraria Editrice Università di Padova"
via G. Belzoni 118/3 – Padova (t. 049 8753496)
www.cleup.it – www.facebook.com/cleup

Paper Munken Linx
Typeface Univers Linotype

ISBN 978-88-6265-005-2

For more information please contact:
CISA Publisher, Via Beato Pellegrino 23, 35137, Padova, Italy
T +39 049 8726986 F +39 049 8726987 www.cisapublisher.com

PREFACE

The International Waste Working Group (IWWG) was established in 2002, following a world-wide demand, to serve as a forum for the scientific and professional community.

The aim of the IWWG is to provide an intellectual platform to encourage and support economical and ecological waste management, and to promote scientific advancement in the field. This aim is being accomplished by learning from the past and by analyzing the present, with a view to developing new ideas and visions for the future.

The objectives of the IWWG are pursued mainly by means of: collecting, developing and disseminating new results and ideas based on Research and Development; promoting discussion on strategic matters, providing and organizing education in waste management and transferring knowledge into practical applications.

To achieve these objectives, the IWWG publishes an international journal (*Waste Management*, Elsevier Publisher), organizes Symposia, Conferences and specialized Workshops, establishes specific Task Groups aimed at discussing the main aspects of waste management and technology, as well as identifying common positions to be proposed subsequently to regulators, decision makers and operators.

In keeping with the aims described in previous paragraphs, the IWWG has opted to publish a series of reference books focusing on the aspects of current interest in the field of waste management and technology.

Contributions to these books originate from relevant papers presented during events organized or promoted by IWWG (in particular the Sardinia Symposia) and from original contributions to the books. The overall intent is to make this wealth of information available to the waste management community in a concise and organized fashion.

The editors of the monographic volumes are internationals experts, generally members of the IWWG Managing Board or Scientific Advisory Panel. Editors perform a variety of tasks including selecting and organizing papers, standardizing the texts and eventually making constructive suggestions when the original manuscript undergoes significant changes. The responsibility for the technical content of the book lies with the individual Authors.

I wish to thank the Editors and all Contributors as well as Roberta Gadia, Tiziana Lai and Paola Pizzardini, for their efforts and support; I trust that the monographic book series and in particular the volume on hand will make a positive contribution towards creating a better understanding of the numerous aspects of waste management and supporting the procedure for making environmentally-safe and economically effective decisions.

Rainer Stegmann, IWWG President

The project for this monograph started a few years ago in the wake of the activities carried out by the Working Group on Sustainable Landfilling, at that time chaired by Raffaello Cossu.

Selected papers from the Proceedings of different Sardinia Symposia have been grouped in order to provide a well-organized and useful volume.

The editing of the work took longer than anticipate, which, combined with the dreadful daily routines we are all affected by, determined a delay in the completion of the final product.

We hope that a widespread interest in the publicaton by the audience can make up for the above delay, for which we sincerely apologize, especially with the authors.

R Cossu
H van der Sloot

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Introduction

Municipal Solid Waste Incineration (MSWI) bottom ash is used in several countries as a construction product. The percentage of the total annual production of ash used beneficially for construction purposes in different countries varies substantially (Chandler et al., 1997;CEWEP, 2010). The environmental properties of the material have been studied extensively in the last decade (Chandler et al., 1997; Dijkstra et al., 2002; Dijkstra et al., 2006b; Dijkstra et al., 2006a; Johnson et al., 1996; Meima et al., 1999; Meima and Comans, 1997; Rendek et al., 2006; Yan et al., 1999; Zevenbergen et al., 1994; Chen, 2007; Kalbe, 2007)

The leaching behaviour of Municipal Solid Waste Incinerator bottom ash was already identified as rather systematic (van der Sloot et al., 1996). Dijkstra et al. (2006a; 2008) have shown that leaching tests (pH dependence test (CEN/TS 14429, 2006) and the percolation test (CEN/TS 14405, 2006)) are highly suitable to characterize MSWI bottom ash leaching behaviour.

For a mechanistic interpretation of test results and their translation to field situations, knowledge is required on the geochemical and mass transfer processes that control the leaching of contaminants in a percolation regime. Reactive transport modeling will be a valuable instrument to identify and describe the dynamic leaching processes of constituents from a material (the "source term") as well as their further transport in soil and groundwater.

A key issue for the commercial use of MSWI bottom ash in construction is the type of testing and the frequency of testing required to meet regulatory criteria, such as the Dutch Soil Protection Act (2008). A hierarchy in testing is a very powerful means of providing the detailed knowledge, which it is needed for criteria development and product improvement, and the quick information, which is needed for regular quality control of the final product prior to delivery.

There are developments in Europe in relation to the essential requirement number 3 on health and environment under the Construction Product Directive (1988). This work has just started in CEN/TC 351 with the development of the tools for national regulations to refer to.

The aim of this study is to review current knowledge on leaching test methods and the data-interpretation of MSWI bottom ash test results. It will be shown that the leaching behaviour of MSWI bottom ash is rather systematic within a certain bandwidth and that the remaining variability of results can be explained by (the extent of) chemical processes leading to release. This work on MSWI bottom ash can be shown to be of relevance for the entire range of bottom ashes produced in one installation of relevance for ashes from different Dutch installations as well as bottom ashes produced in different countries. For this the database /expert system LeachXS provides unique capabilities (van der Sloot et al., 2003) allowing such comparison and data evaluation.

Materials and methods

Samples and leaching tests

The test results for the materials presented in this work were obtained from different studies on MSWI bottom ash samples from: The Netherlands (Meima and Comans, 1997; Dijkstra et al, 2006a; Dijkstra et al, 2008), Germany (Kalbe, 2007), France (Rendek et al, 2006), Taiwan (Chen, 2007), Austria (Van der Sloot et al, 2000) and US (Lopez Meza et al, 2008). Standardised test methods and a unified data format are a prerequisite for being able to make comparisons between different materials (van der Sloot et al, 1997). Single step leaching tests alone, such as currently used in a regulatory context, are inadequate for such a comparison. In CEN TC 292 characterisation leaching test methods have been standardised that are suited for this purpose. Several materials have been leached by a percolation test as currently standardised in CEN TC 292 "Waste" (CEN/TS14405, 2005a) to address the long-term behaviour in percolation dominated conditions. To address the factors controlling leaching and to evaluate changes in material properties due to external influences a pH dependent leach test has been applied (CEN/TS14429, 2005b). The pH dependence leaching test consists of a combination of individual batch leaching test, in which pH is controlled (constant L/S = 10 for 48 hours) using pre-determined amounts of acid/base to reach given end point pH values (ANC mode). This method has been applied in most cases in this study. From the acid/base consumption of the material to reach a certain end pH the acid/base neutralisation capacity can be derived (expressed in mol/kg). Both the time dependent leaching as reflected in a percolation leaching test and the pH dependent leaching behaviour are used to predict the release and changes in release of constituents in short and long term due to external influences.

Data-evaluation and geochemical modeling

The geochemical modelling framework ORCHESTRA (Meeussen, 2003), using an extended MINTEQA2 database with thermodynamic constants for inorganic reactions, was coupled to a database/expert system (LeachXS)(van der Sloot et al., 2003) containing the leaching test data for quick data retrieval, processing and data presentation. The amount of amorphous and crystalline iron (hydr)oxides in the waste mixture was estimated by a dithionite extraction described in Kostka and Luther III (Kostka and Luther III, 1994). The extracted amounts were summed and used as an estimate for hydrous ferric oxides (HFO) in the model. DOC was used as input for humic acid modelled with the NICA-Donnan model (Kinniburgh et al., 1999). The availability of all elements was estimated and used as input, the model calculates the predicted concentrations and the speciation in both the leachates and the solid matrix.

Worldwide a large number of leaching test methods are applied. However, different types of data representations currently used might be confusing to the end-users as data are not directly comparable. Therefore, a unified presentation of leaching data has been developed in LeachXS, allowing results from many different leaching tests to be placed in perspective. The database/expert system uses Orchestra (Meeussen, 2003) to enable geochemical speciation and reactive transport modeling. Through this approach the consistency in the release behaviour can be shown and remaining variability largely explained in terms of specific release controlling factors. In addition this unified data presentation format allows a variety of conclusion to be drawn from the same basic set of test results.

Results and discussion

Comparison of total composition versus leaching of contaminants

In the framework of the validation of EN 12457 parts 1–4 test methods (van der Sloot et al., 2001) the sub-sampling of MSWI bottom ash was evaluated by total content analysis and by leaching. The results of total composition analysis and leaching tests (EN 12457-2) for 15 sub-samples are shown in Table 1.

The results illustrate that sampling for leaching tests is less critical than for total composition as the inherent variability, in particular for metal composition, is rather high. This is due to occasional metal parts (staples, wire and solder) that can be included in a test portion during sampling. This can not be avoided even after very rigorous sample pre-treatment. The concentration levels of leaching are orders of magnitude lower than the content, which implies that only a minor fraction of the material is leached.

The leaching results do show some relatively large uncertainties, but these are invariably associated with very low concentration levels in the eluate. The concern for environmental impact at these levels might be far less than at higher concentrations. Therefore, the variability is acceptable given the low concentrations around the detection limits of most analytical instruments. The impact level at which a decision based on leaching is taken in comparison with a decision based on content is far more protective for the environment. It is a misconception that quality control through leaching is more complicated than total composition and more costly.

Comparability of test methods

The uniform data presentation is given for Zn leaching from MSWI bottom ash in Figure 1. The vertical lines of the box in figure 1 (top left) indicates the pH domain relevant for the application under consideration. The horizontal lines represent the regulatory limit value and the detection limit respectively. The solid line in figure 1 (top right) represents the regulatory limit value.

[Table 1] MSWI Bottom ash Composition versus leaching (repeatability, n = 15).

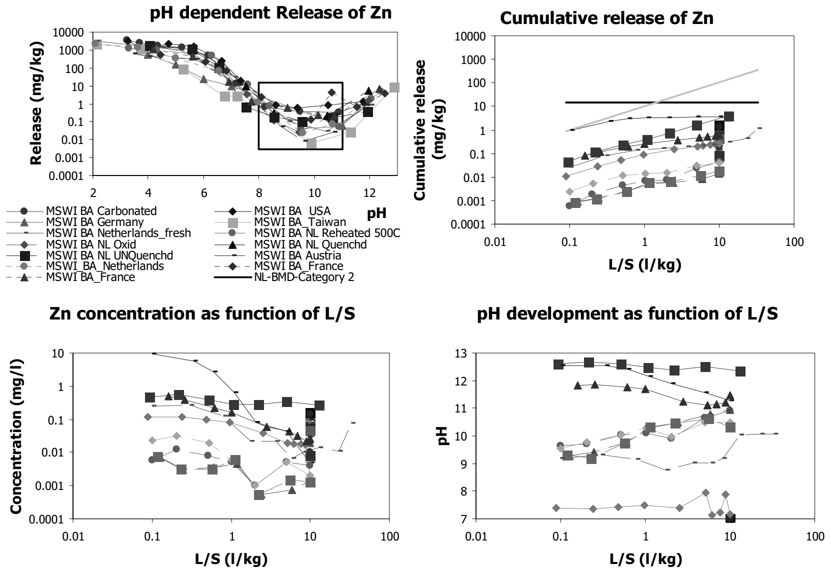
	Total content (mg/kg)			Leaching (mg/kg bij L/S=10)			Ratio	Ratio
Element	Average	St. dev	Stdev. in %	Average	St. dev	Stdev. in %	Total/Leaching	Absolute SD's
As	13.9	1.7	12.4	0.0086	0.0057	66.7	1626	301
Ba	713	207	29.0	0.11	0.0044	4.1	6629	47390
Cd	4.4	0.87	19.9	0.0007	0.0002	31.5	5916	3746
Cr	193	31	15.9	0.018	0.0077	43.2	10923	4031
Cu	3652	2815	77.1	0.55	0.057	10.3	6652	49539
Mo	4.4	1.9	42.9	0.051	0.0053	10.3	87	359
Ni	158	63	39.9	0.0030	0.0013	43.7	53402	48759
Pb	1218	524	43.0	0.010	0.0078	75.9	119002	67453
SO4 as S	2885	240	8.3	104	9.16	8.8	28	26
Sb	28	4.3	15.0	0.030	0.0040	13.5	945	1054
Sr	199	38	19.0	0.95	0.071	7.4	210	536
V	24	5.4	22.2	0.0014	0.0004	26.9	17416	14375
Zn	2275	1045	45.9	0.017	0.0025	14.9	135306	416360
pH				11.0	0.059			

The percolation test (TS 14405) is generally used to compare measured emissions (at a cumulative L/S ratio of 10 L/kg) with regulatory criteria. An example of MSWI bottom ash data from several countries is given in Figure 1 (top right graph). The difference between Zn emissions from several bottom ash samples is orders of magnitude. One cannot properly explain the reasons for these differences without comparison of these results with the results from a pH dependence leach test (Figure 1, top left). The native pH of the MSWI bottom ash samples ranges from about 7.5 to 12.5 (Figure 1, bottom right). Using this information, it can be seen that the pH dependent leaching behaviour of Zn also changes orders of magnitude in this pH range. The large difference in Zn emissions in the percolation test can largely be explained by variations in the pH of the samples. In addition, other chemical processes that lead to Zn leaching (complexation to DOC) or to a low solubility (controlled by mineral phases or by adsorption to HFO) can play a role. These chemical processes can only be quantitatively assessed using geochemical modeling (see below). The use of information from both the pH dependence leach test and the percolation test enhances the interpretation of test results and forms the basis for quality improvement of MSWI bottom ash.

Variability in leaching between bottom ash from different sources (Austria, France, Germany, Taiwan, The Netherlands and USA)

Leaching data from different countries have been implemented in the LeachXS database to compare data from the same installation at different production dates, between different installations in a country and between different installations in different countries. In figure 2 this comparison is illustrated for Cu and Mo.

The Cu emissions measured with the percolation test can vary over orders of magnitude. In contrast to the leaching of Zn (Figure 1), this variation cannot be explained by the differences in pH of the samples. The leaching of Cu is only limited dependent on the pH in the relevant pH domain between 7.5 and 12. It has been found that the Cu leaching is



[Figure 1] Integrated leaching behaviour of Zn from MSWI bottom ash from different sources using the pH dependence test (TS14429) and the percolation test (TS 14405).

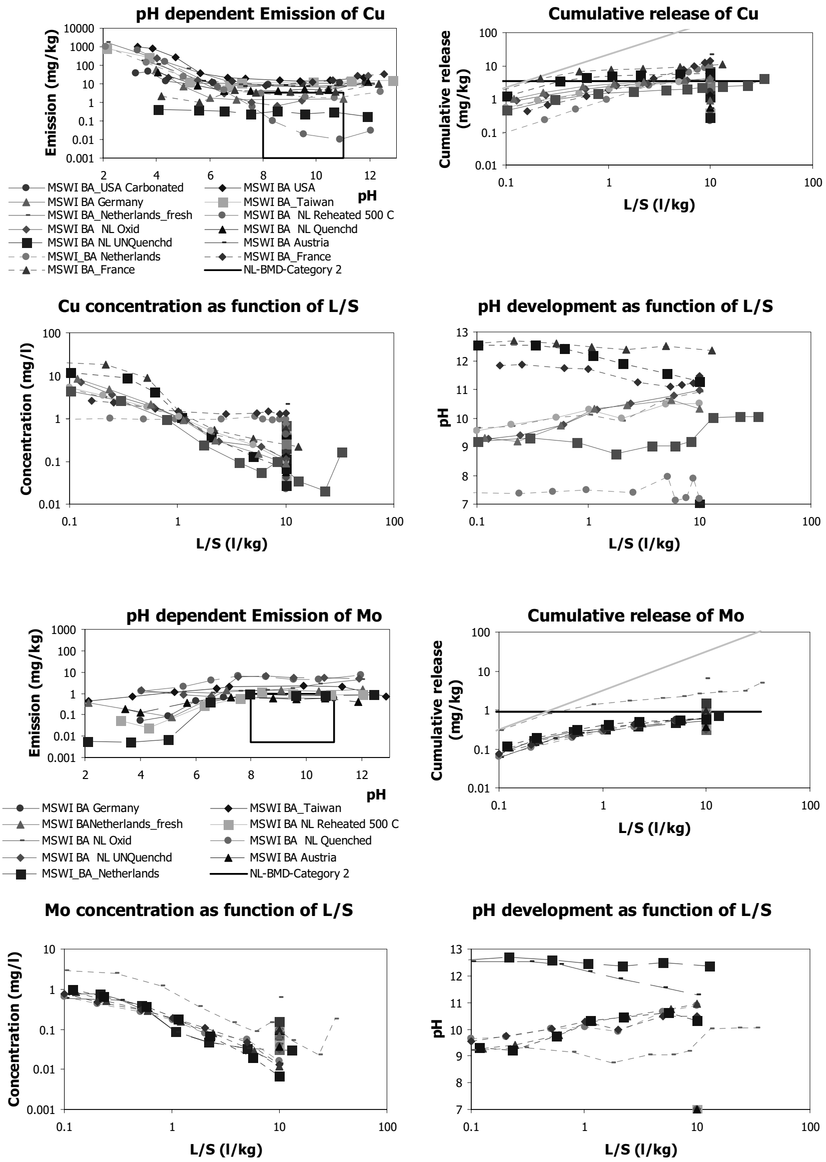
primarily controlled by the availability of fulvic acids in the leachates (van Zomeren and Comans, 2004). Geochemical modeling is needed to quantitatively assess and understand the leaching of Cu from these residues.

The concentration change as a function of the liquid to solid (L/S) ratio in the bottom left graph of figure 2 shows the decrease in concentration with time as (organically complexed) Cu is washed out. The low L/S (0.1-0.2) represents porewater conditions, which is the first leachate from an embankment or similar high volume application scenario.

The cumulative emissions of Mo are also given in Figure 2 and it can be seen that the variation in emissions is generally well within one order of magnitude, despite the wide range of bottom ash sources. The leaching of Mo is almost independent of the pH in the range from 7.5 to 12 and can be explained by weak surface complexation of molybdate (MoO_4^{-2}) to HFO (Dijkstra et al., 2006b).

Modeling of leachability example pH stat

The experimental and modeling approach developed for contaminated soils by Dijkstra et al. (Dijkstra et al., 2004) has also been implemented in LeachXS, this allows easy geochemical modeling and visualization of release controlling phases under a range of pH (and L/S) conditions. Results for Pb from a MSWI bottom ash impact assessment are given as illustration in figure 3. The leaching behaviour as a function of pH can be adequately described (blue line, L/S = 10) with the geochemical model.



[Figure 2] Comparison of the leaching behaviour from MSWI bottom ash from different production dates and from different locations (within one country and between countries).

The chemical processes that determine the leaching behaviour of Pb can be seen from the speciation graphs in Figure 3. Around the native pH of many fresh bottom ash samples (pH 10-11), leaching of Pb is primarily controlled by the solubility of $Pb(OH)_2$. In the leachate, Pb is mainly complexed to DOC (fulvic acids). The modeling of the pH dependence leaching test data provides a chemical speciation fingerprint that can be used in subsequent modeling actions involving transport. This implies that a very wide range of possible exposure conditions is covered.

It is important to know in what chemical form Pb or any other relevant element is released as that determines the possible impact. Free Pb will, when leached from ash be retained in the subsoil readily, whereas DOC-bound Pb can be transported over larger distances from the source. On the other hand DOC-bound Pb is less likely to lead to effects on soil organisms as only the free ionic Pb forms can be taken up by most organisms.

Modeling of release in a percolation test

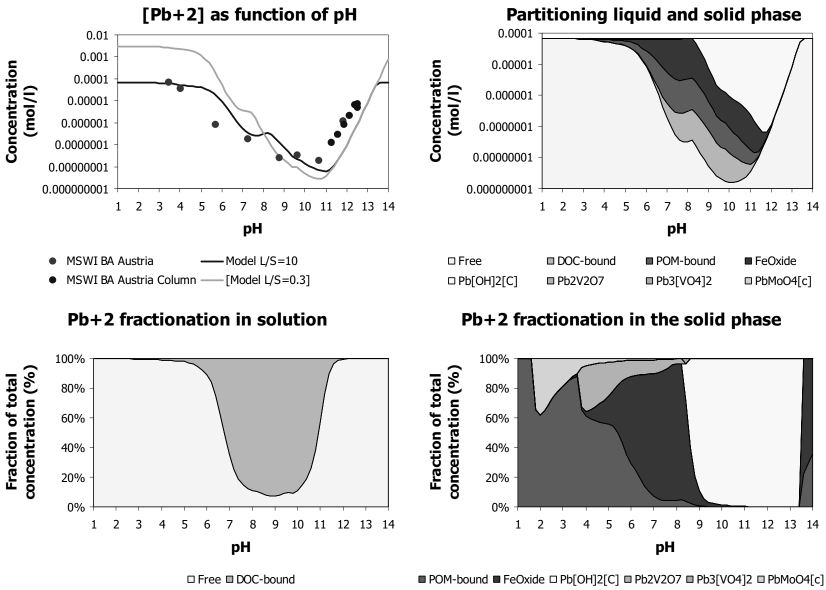
A next step is to model the release in a percolation leaching test based on the chemical speciation fingerprint as obtained from the modeling of pH dependent leaching test data.

A dual porosity model is applied to account for non-equilibrium conditions during percolation. Additional factors are column dimension, porosity and mass transfer between the stagnant and the mobile phase. The results of release modeling from the percolation leaching test is given in Figure 4. The closed circles represent the actual concentrations measured in the percolation test. The drawn line is the predicted concentration change as a function of L/S from the multi-element dual-porosity chemical reaction/transport model. The open circles are the calculated averaged concentrations over the L/S interval for which eluate was collected and these points should be compared directly with the closed circles. The release of these contaminants as a function of the L/S ratio is adequately predicted by the model. The results imply that the physical and chemical processes in MSWI bottom ash are sufficiently understood and this knowledge can be used in (long-term) environmental impact assessments of this material.

Some leaching behaviour aspects relevant for product quality

A number of relevant factors have been identified affecting the leaching behaviour of specific constituents from MSWI bottom ash. In the Netherlands, the leaching of particularly molybdenum and copper does not always comply with environmental regulations, which requires special (and expensive) measures to reduce contact with water. In order to understand the cause of the enhanced concentrations of these contaminants in MSWI bottom ash leachates it is necessary to identify the underlying geochemical processes.

We have found that dissolved natural organic matter is responsible for enhanced copper leaching from MSWI bottom ash. More specifically, we determined that the leached copper concentrations are for 80-100% bound to FA (Dijkstra et al., 2006b; van Zomeren and Comans, 2004). Heat treatment (500 °C) to burn organic matter away or the leaving the ash unquenched, leads to lower Cu leachability because of reduction of



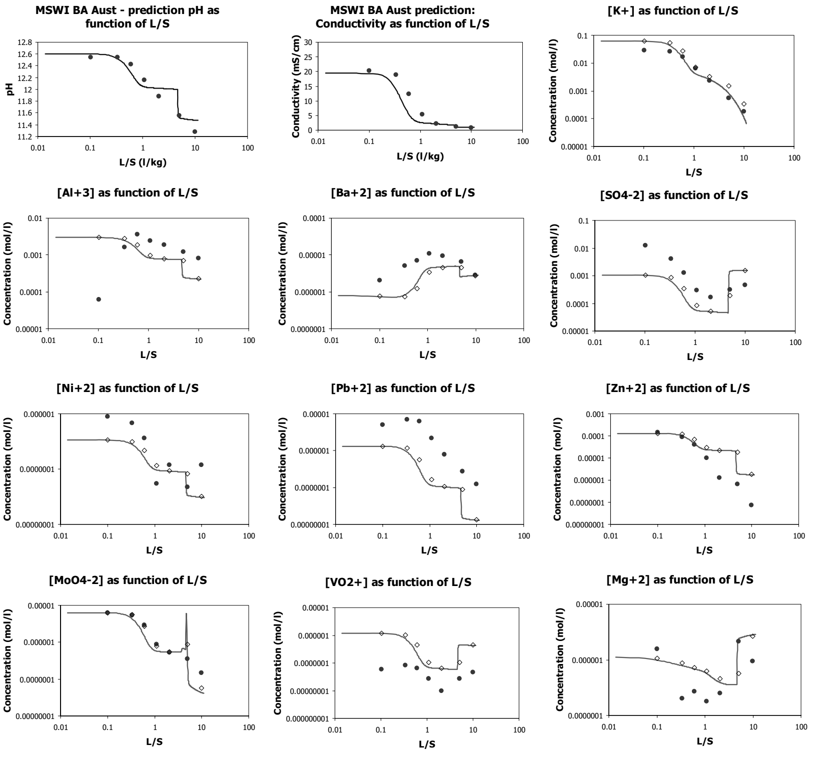
[Figure 3] Chemical speciation modeling of Pb in MSWI bottom ash illustrating the partitioning between dissolved (free and DOC-bound) and particulate forms of Pb (Pb-minerals, Feoxide bound and particulate organic matter bound).

the organic matter in the bottom ash. In the case of heat treatment the Cr leachability increases, as Cr III is oxidized to Cr VI. Generally the concentration level stays below the critical regulatory limit.

Carbonation of bottom ash (pH 12 to pH 10) has a positive effect on the leachability of Pb and Zn. It has also been found that the leaching of Cu and Mo can be reduced as a result of accelerated ageing with CO_2 enriched air. New Fe- and Al(hydr-)-oxides surfaces can adsorb organically complexed Cu and Mo anions (Dijkstra et al., 2006b). The change in pH also leads to increase of other elements (e.g. Sb). A high pH in ash may be caused by a high gypsum input (e.g. plaster board) or shells.

Conclusions

The leaching behaviour of MSWI bottom ash is very consistent and worldwide results are quite comparable. This implies that the release controlling processes are the same or at least very similar for nearly all MSWI bottom ashes. From chemical speciation modeling work these controlling phases can be quantified and model predictions fit with actual practice rather well. This creates opportunities for understanding product quality, when deemed necessary. Placing QC data (usually single step tests) in perspective with generic characterization data for MSWI bottom ash will provide a sound basis for judgment of material properties and production variability



[Figure 4] Predicted and measured release in a percolation test TS14405 illustrating the level of prediction possible using a mechanistic modeling approach.

and variability between installations. Relationships between total content and leachability only exist for Cl, Br, Na, K and possibly to some extent for Mo. For all other elements chemistry controls release and relationships between waste input and release is not easily made. A few mechanistic scenario descriptions for different applications of MSWI bottom ash are in advanced state of preparation.

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