

Leach testing for pre-selection of waste to be landfilled

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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 international 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 anticipated, 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 publication 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

In this chapter, geochemical speciation modelling is used to determine important chemical processes that determine the release of contaminants from waste materials in short and long term. Moreover, model calculations are performed to show the sensitivity of the waste mixture to changes in the contaminant availability and binding capacity by changing the amounts of hydrated iron oxide (HFO) and organic matter. In

spite of the heterogeneity in waste going to landfill there is a scientific basis for more focused waste selection/acceptance criteria in order to reduce the environmental impact of landfills. The potential pre-selection of waste and the prediction of long term release behaviour of a waste mixture form the basis for the work described in this paper.

Materials and methods

Leaching tests

Laboratory experiments (percolation test – CEN/TS 14405, 2003; pH dependence test – CEN/TS 14429, 2003), lysimeter studies (1-1,5 m³) and a 12,000 m³ pilot demonstration project at landfill site Nauernasche Polder (the Netherlands) were performed. Results from the percolation test, the lysimeter study and the pilot demonstration project are described elsewhere (van der Sloot et al. 2001; van der Sloot et al. 2003).

From all waste samples collected during the filling of the pilot test cell, an integrated waste mixture was prepared by taking the waste mass per waste charge into account. The waste mixture was leached with a pH dependent leaching test (CEN TS 14429). About 80 g portions of the waste mixture was leached with 800 mL water (L/S = 10, corrected for the moisture content of the sample) and equilibrated at pH values between 3 and 12. The pH was adjusted at the start of the experiment and at several times during the experiment (up to 6 hours) with 14.4 M HNO₃ or 10 M NaOH. After 48 hours of equilibration by end over end rotation in PE containers the suspensions were filtered (0.45 µm) and analysed.

Estimation of model parameters

The quantities of “reactive” organic carbon in the solid phase (i.e. HA and FA) were estimated by a batch procedure (van Zomeren and Comans 2007), which is derived from the procedure currently recommended by the International Humic Substances Society (IHSS) for solid samples (Swift 1996). In short, the procedure is based on the solubility behaviour of HA (flocculation at pH < 1) and the adsorption of FA to a polymer resin (DAX-8). The amount of amorphous and crystalline iron (hydr)oxides in

the waste mixture was estimated by a dithionite extraction (Kostka and Luther III 1994). The amount of amorphous aluminium (hydr)oxides were estimated by an oxalate extraction (Blakemore et al. 1987). The extracted amounts of Fe and Al were summed and used as a surrogate for hydrous ferric oxides (HFO) in the model.

Chemical analysis

The leachates and extracts from laboratory tests were analysed for major, minor and trace elements by ICP (Al, As, B, Ba, Ca, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, P, Pb, S, Sb, Se, Si, Sn, Sr, Tl, V, Zn). DOC (dissolved organic carbon) and TIC (total inorganic carbon) were analysed by a Shimadzu TOC 5000a analyser. Cl, F, ammonium and sulphate were analysed by ion-chromatography.

Geochemical speciation and release modelling

Chemical speciation of the solutions was calculated with the ORCHESTRA modelling framework (Meeussen 2003). Aqueous speciation reactions and selected mineral precipitates were taken from the MINTEQA2 database. Ion adsorption onto organic matter was calculated with the NICA-Donnan model (Kinniburgh et al. 1999), with the generic adsorption reactions as published by Milne et al. (Milne et al. 2003). Adsorption of ions onto iron and aluminium oxides was modelled according to the generalized two layer model of Dzombak and Morel (Dzombak and Morel 1990).

The database/expert system LeachXS was used for data management, e.g. pH dependent leaching data, percolation test data, lysimeter and field leachate data and for visualization of the calculated and measured results (van der Sloot et al. 2003; van der Sloot et al. 2001). The coupled LEACHXS - ORCHESTRA combination allowed for very quick data retrieval, automatic input generation for modelling, processing of calculated results and graphical and tabular data presentation.

The input to the model consists of metal availabilities, selected possible solubility controlling minerals, active Fe- and Al-oxide sites (Fe- and Al-oxides were summed and used as input for HFO as described in (Meima and Comans 1998)), particulate organic matter and a description of the DOC concentration as a function of pH (polynomial curve fitting procedure). Basically, the speciation of all elements is calculated in one problem definition in the model with the same parameter settings. This limits the degrees of freedom in selecting parameter settings considerable, as improvement of the model description for one element may deteriorate the outcome for other elements. As a starting point for the model calculations, the maximum value as obtained in the pH dependence leaching test (between pH 3 and 13) was used as the available concentration. It was found that total (leachable) carbonate concentration plays an important role in the model calculations. This parameter was not measured and was therefore estimated based on the model output. The concentration was adjusted until the major (and some minor) elements showed a reasonably good match with the observed leaching data. There is a clear need for more data on total (available) carbonate concentrations in waste materials to enhance model predictions.

The mineral phases that were allowed to precipitate were selected after calculation of their respective Saturation Indices (SI) in the original pH dependence leaching test eluates. Saturation indices were calculated for all > 650 minerals in the thermodynamic database and a selection of the most likely and relevant phases was made based on the degree of fit over a wider pH range and the closeness of the SI value to 0 and an expert judgement on suitability of possible minerals for the waste mixture (e.g. exclusion of high temperature minerals). Generally, minerals were selected if the SI was in the range of -2 to 2 for more than two pH data points.

Results and discussion

Interpretation of results

The leaching data in Figures 1 to 5 is represented as a function of pH by the data points. The thick solid line represents the predicted total leached concentration of the considered element, which should ideally meet the data points for good understanding of the chemical processes that determine the leaching behaviour. In addition, the Figures show the calculated chemical speciation of the element in both the solid matrix and the sample solution. The predicted leaching behaviour is therefore the intersection between the calculated speciation (minerals, sorption to Fe-oxides and binding to solid organic matter) in the solid matrix (above the line representing the predicted total leached concentration) and in the solution (free + inorganic and complexed by dissolved organic carbon). This type of data presentation integrates the predicted total leached concentration as well as the different species that determine the leached concentrations. In figure 4 the partitioning of Zn between dissolved and particulate phases is shown as a percentage distribution of phases.

Geochemical modelling

The availability of all elements used as input for geochemical speciation modelling is given in Table 1. It should be noted that the availability was determined as the maximum concentration that was obtained in the pH-static leaching test.

DOC is a sum parameter for all organic carbon species. Not all of the DOC is reactive in metal binding, therefore a fraction of total DOC is used in the calculation. Both at low and high pH a larger proportion of DOC is in the reactive form (van Zomeren and Comans, 2004), while at neutral pH the lowest proportion of reactive DOC is found. A varying proportion of reactive DOC is assumed from 50 % at low and high pH to 10 % at neutral pH. This fraction is assumed to consist of humic acid, for which the interaction parameters according to Nica-Donnan are used. The corrected DOC data from the pH dependence test were fitted to a polynomial function in order to describe the pH dependent leaching of DOC for intermediate pH values. ORCHESTRA calculates the geochemical speciation from pH 1 to 14 with intervals of 0.2 pH values. Initial speciation calculations have indicated several possible solubility controlling minerals (SI saturation index close to 0). From this, a set of minerals was selected for the model prediction calculations, of which the minerals listed in table 2 proved relevant.

[Table 1] Available concentrations of elements used in geochemical modelling. Additional parameters to describe binding to the solid phase were: HFO (4.3E-3 kg/kg), total humic acid content (1.03E-3 kg/L) and clay (0.1 kg/kg).

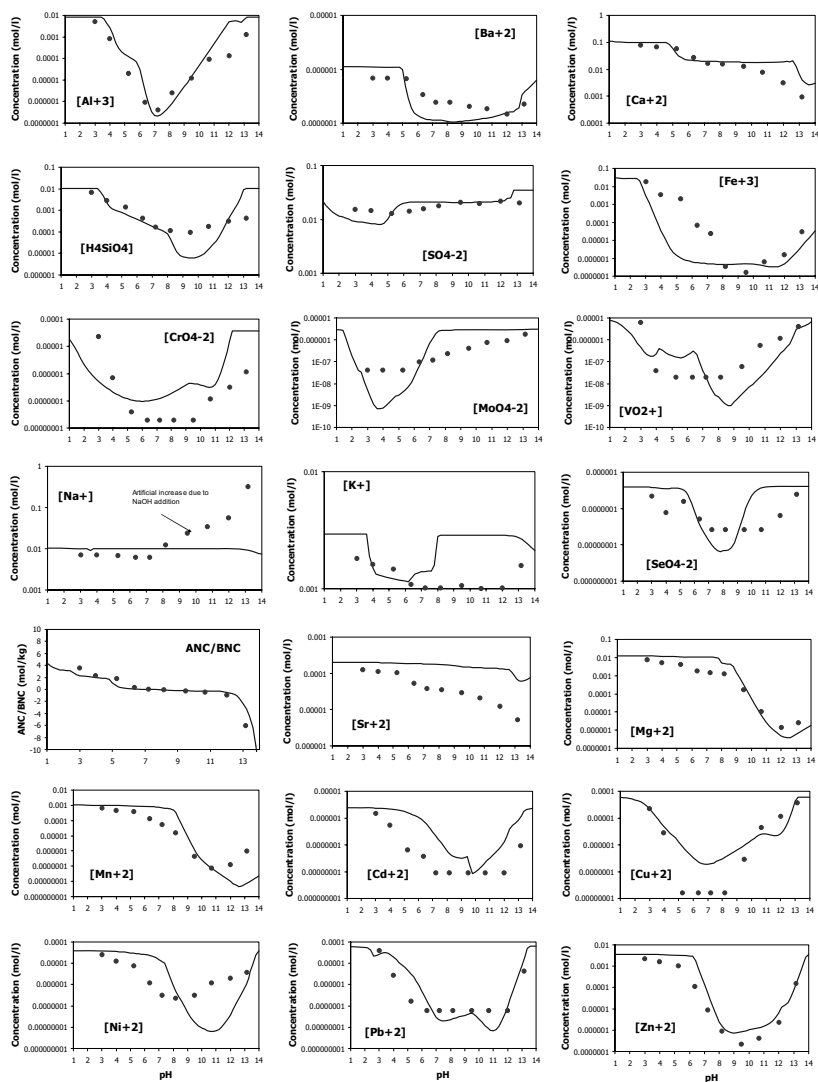
Element	Availability (mg/kg)	Element	Availability (mg/kg)	Element	Availability (mg/kg)	Element	Availability (mg/kg)
Al	3000	Mg	3002	CO3-2	54000	SO4-2	33621
As	2.57	Mn	574	Cr	19.2	Sb	0.39
B	18.7	Mo	2.87	Cu	39.8	Se	0.32
Ba	1.54	Na	2360	F	50	Si	3015
Br	34.5	NH4	609	Fe	16360	Sr	176
Ca	50150	Ni	23.2	K	1158	V	5.22
Cd	2.76	PO4-3	50	Li	3.02	Zn	2401
Cl	5268	Pb	129				

[Table 2] Selected mineral phases.

Calcite	Albite[low]	Ca2Cd[PO4]2	Cu[OH]2[s]	Pb[OH]2[C]	Willemite
CO3-hydrocalcite	Ba[SCr]	CaZincate	Fluorite	Pb2V2O7	
	O4[77%SO4]				
Fe[OH]3[microcr]	BaSrSO4[50%Ba]	Cd[OH]2[A]	Illite[2]	PbMoO4[c]	
Gypsum	Boehmite	Corkite	MnHPO4[C]	Portlandite	
Magnesite	Brucite	Cr2O3	Ni2SiO4	Talc	

The geochemical model results for several major and minor elements in comparison with the measurements in the pH-static leaching test are given in Figure 1. This gives an overall impression of the degree to which prediction and test results for major, minor and trace elements match in a multi-element model run. For one element proper stability data may be lacking (Sr at pH >6), for an other the stability data as obtained from literature may not be entirely adequate (PbMoO4). The agreement between measured and calculated acid/base neutralisation capacity (ANC/BNC) based on the selected minerals and sorption phases is good indicating that the selection of minerals to describe major element behaviour is not far off.

In figure 2 and 3 the agreement between model and prediction is further specified by the partitioning between dissolved and particulate phases. In general, the model describes the leaching behaviour of the waste mixture quite well, especially when it is realised that changes in input parameters may affect the predicted behaviour of several other elements. This implies that the degrees of freedom to vary input parameters are limited dramatically by taking all elements into account simultaneously. As the model assumes equilibrium and it is known that equilibrium is not reached within 48 hours contact time, kinetics of dissolution and precipitation will be a factor to recon with in judging the results. These effects will result in an apparent deviation of the model prediction, whereas the



[Figure1] Predicted results from simultaneous full element chemical speciation (line) with pH dependence test data for the mixed waste (dots).

leached concentrations might still increase or decrease due to equilibrium and/or kinetic processes. Typical examples of such deviations in figure 1 are Mg and Mn at pH 6-8. Work by Dijkstra et al. (2005) has shown these effects in relation to the own pH of the material, where the system is closest to equilibrium. This implies that there is sufficient understanding of the chemical processes that determine the leaching behaviour in this waste mixture.

The pH-static leaching data and geochemical model results for Al, Ca, Fe and Mg are presented in Figure 2. There can be deviations in the model description compared to the actual leaching data, as for instance the leaching of Fe is underestimated for about one to two orders of magnitude in the pH range 5 to 7. Most likely due to not considering the redox status of the waste sufficiently. Despite the model deviations in the leaching of the described elements, the overall prediction of the major element chemistry is an important finding and provides a good basis for the understanding of chemical processes in these apparently very heterogeneous materials.

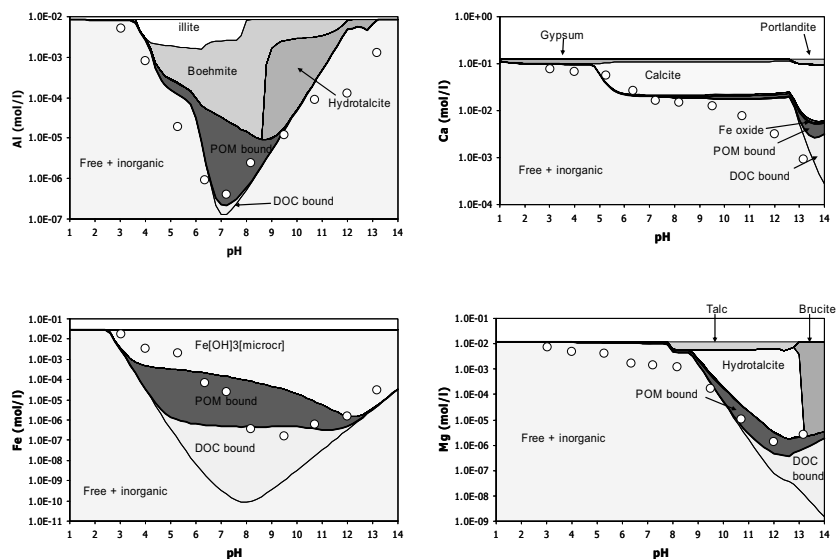
Figure 3 shows the pH dependent leaching behaviour and results from geochemical modelling for Cr, Mo, Pb and Zn. The leaching behaviour of Zn is quite well described. Pb is well described in the neutral pH range, but dissolution kinetics may be the cause for the discrepancy between model and measurement in the mild acidic range. The leaching behaviour of Cr and Mo are reasonable well described. The latter is controlled by Pb and thus any change in Pb partitioning will affect Mo leachability. The thermodynamic database is adequately stocked, but the selection of relevant mineral- or sorptive phases to be included in the predictive modelling requires further work.

The partitioning between dissolved and particulate phases as obtained from the present modelling (Figure 4) provides a significantly increased insight in mutual relationships between elements, which allows prediction of release behaviour under other conditions than those tested in the laboratory.

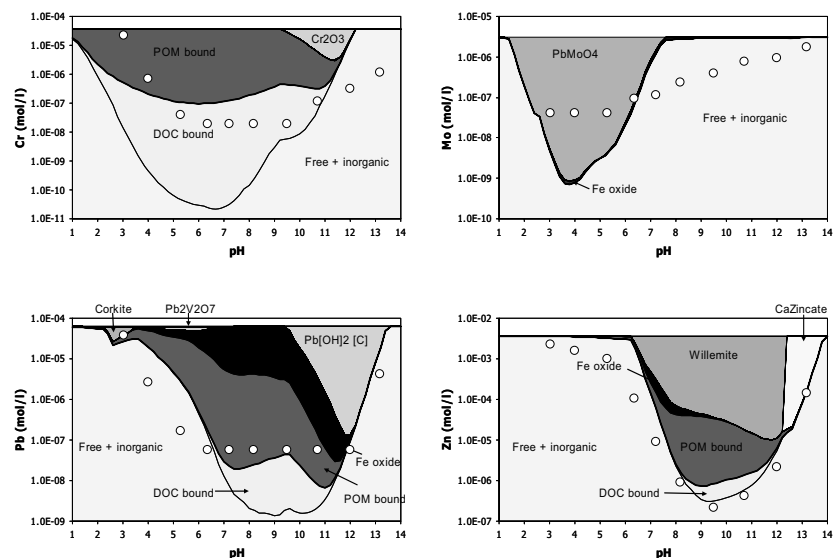
Influence of waste properties on leaching behaviour

The results from geochemical modelling show that there is understanding of the chemical processes that determine the leaching behaviour of a significant amount of elements from the waste mixture. With this information a basis is formed for sensitivity analysis. With sensitivity analysis, influences of changing contaminant availability, amounts of organic matter and/or HFO can be assessed. The outcome can serve as a basis for waste management decisions for landfill owners.

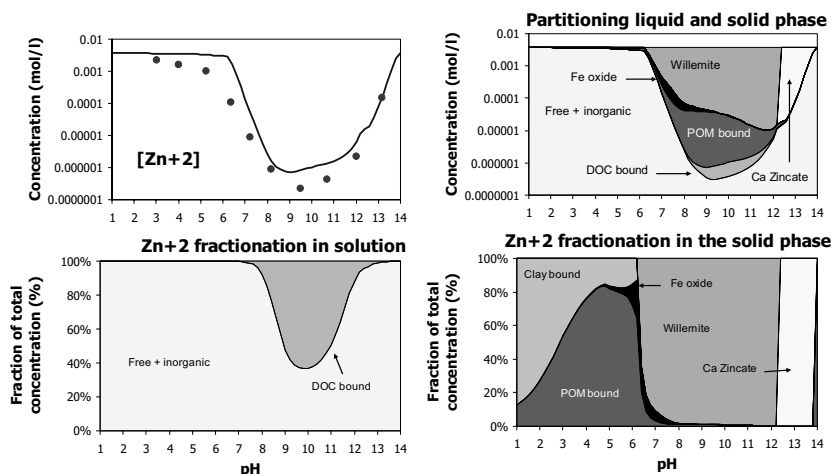
An example has been worked out in detail; it deals with the calculated Pb emissions in the waste mixture as a result of an increased Pb availability (in all cases the measured initial pH dependence test results are shown for reference). In practice, this would imply the effect of adding a waste material with a significantly higher Pb concentration (available for leaching). The initial calculations are given in Figure 5A, it can be seen that Pb is substantially bound to HFO in the solid phase at neutral to slightly acidic pH values. Figure 5B shows the measured and predicted pH dependent leaching behaviour of Pb where the availability of Pb was increased by a factor 10.



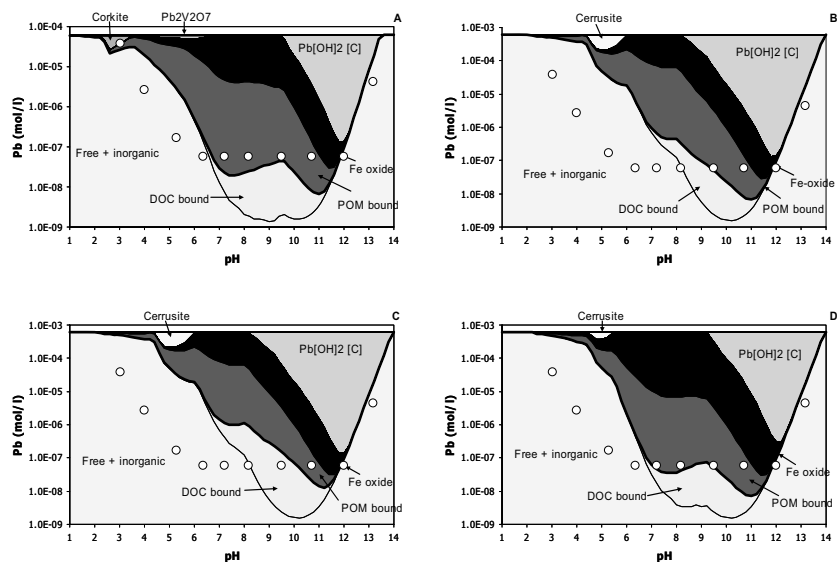
[Figure 2] pH-static results and geochemical modeling for the major elements Al, Ca, Fe and Mg.



[Figure 3] pH-static results and geochemical modelling for the elements Cr , Mo , Pb and Zn.



[Figure 4] Partitioning of Zn in dissolved and particulate phases.



[Figure 5] Prediction of pH dependent Pb leaching from a waste mixture under varying conditions. **A:** Initial calculations; **B:** Pb availability increased by a factor 10; **C:** Pb availability increased by a factor 10 and DOC increased by a factor 3; **D:** Pb availability increased by a factor 10 and HFO increased by a factor 5. Open circle: pH dependence test data on waste mix (in B, C and D only shown for reference to A starting condition).

In Figure 5C, the Pb availability and the DOC concentration is increased in the model by a factor 10 and 3, respectively. It can be seen that the predicted Pb emissions in the neutral to acidic pH region significantly increase as a result of the increased availability. This implies that increasing Pb availability under unchanging conditions of the waste mixture will directly result in an increasing Pb emission. Figure 5C shows the effect of increased DOC concentrations on the predicted Pb emissions. It can be seen that Pb emissions become higher, mainly as a result of organically complexed Pb. Increase of both the Pb availability as well as the HFO content of the waste mixture will lead to a subsequent reduction of the Pb emissions (Figure 5D) to about the level of initial emission in the neutral pH range. Only at pH values lower than 6, an increase in the emissions is observed as already shown in the initial calculations (Figure 5A). Binding to HFO will not be the dominant solubility controlling mechanism at these conditions. However, these low pH values will probably not be relevant in practice.

Conclusions

In this study, geochemical speciation modelling was used to identify important chemical processes that determine leaching from mixed waste materials in short and long term. pH-static leaching tests in combination with model predictions of the leaching behaviour and the speciation of contaminants in both the solid phase and solution provides detailed knowledge of chemical processes in these apparent heterogeneous materials. Significant progress in geochemical modelling has been made, whereas many elements can be adequately predicted simultaneously by model calculations. This is particularly true for the pH domain around the own pH of the material (neutral pH). There are gaps between data and model predictions for a number of constituents, which are due to missing thermodynamic data, as yet unidentified mineral phases or kinetics of dissolution and precipitation.

The advantage of the integral approach applied here is that more guidance can be derived as to which factors need further work. Kinetics have been shown to be of relevance in the type of test used in this work, which may help to decide at what level a match between measurement and prediction is sufficiently accurate for a subsequent decision. The potential to predict behaviour under conditions that have not yet been tested before (e.g. low L/S, imposed redox condition, increased contamination) provide important insight on how to design verification experiments. In general, the results show that the approach of characterisation and geochemical modelling provides an increased level of understanding the relationships between major, minor and trace elements, which significantly helps to make choices through the acceptance of waste to reach a more sustainable landfill practice. Based on a database containing information on individual wastes, the type of modelling of mixtures as described here can be applied to assess long term release behaviour for a projected waste mix considered relevant for a given site. In addition, it allows verification of streams that disproportionably would influence the leachate quality.

From the modelling it has become very clear that the interactions between major, minor and trace elements forms a very significant limitation for studies in which only a limited set of elements is evaluated, let alone the unjustified omission of major elements, which dictate the leaching conditions that are imposed on trace constituents of concern. The parameter settings for the prediction will be used in subsequent work on combined chemical reaction/transport to predict percolation behaviour at lab, lysimeter and field scale.

If the relevant parameters (sum of dissolved and particulate organic matter, sum of Fe/Al oxide surfaces, relevant minerals) describing the leaching behaviour of the landfilled mix can be derived from the mass contribution and properties of disposed waste in a cell, a reasonably accurate prediction of leachate quality may prove feasible for that cell.

The sensitivity analysis that can be made with this type of modelling allows decisions on acceptability of waste affecting the behaviour of the landfill as a whole. Also changes in waste input to the landfill can be assessed, as well as effects of contaminant loading of waste being accepted for landfilling. In comparison with the current approach to waste acceptance, which is based on judging individual waste arriving at the gate, it will be possible to make a judgment on the behaviour of an entire cell and verify that against leachate quality from that cell. In the current system of waste acceptance wastes are rejected, which pose no threat to the release from the landfill as a whole (for example high pH waste exceeding criteria for Pb will after neutralisation by the co disposed waste pose no risk of Pb release from the landfill cell).

This type of prediction will also be useful for decisions about the level of aftercare measures to be defined at the time of landfill closure. Thus the preliminary sensitivity analysis of the leaching behaviour under varying conditions indicates that there is a scientific basis for more focused waste selection/acceptance criteria in order to reduce the environmental impact of landfills, thereby creating a landfill with minimal impact which in that way can help to reduce or eliminate the need for long-term aftercare.

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