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# Leaching, geochemical modelling and field verification of a municipal solid waste and a predominantly non-degradable waste landfill

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#### ABSTRACT

In spite of the known heterogeneity, wastes destined for landfilling can be characterised for their leaching behaviour by the same protocols as soil, contaminated soil, sediments, sludge, compost, wood, waste and construction products. Characterisation leaching tests used in conjunction with chemical speciation modelling results in much more detailed insights into release controlling processes and factors than single step batch leaching tests like TCLP (USEPA) and EN12457 (EU Landfill Directive). Characterisation testing also can provide the potential for mechanistic impact assessments by making use of a chemical speciation fingerprint (CSF) derived from pH dependence leaching test results. This CSF then forms the basis for subsequent chemical equilibrium and reactive transport modelling to assess environmental impact in a landfill scenario under relevant exposure conditions, including conditions not readily evaluated through direct laboratory testing. This approach has been applied to municipal solid waste (MSW) and predominantly non-degradable waste (PNW) that is representative of a significant part of waste currently being landfilled. This work has shown that a multi-element modelling approach provides a useful description of the release from each of these matrices because relevant release controlling properties and parameters (mineral dissolution/precipitation, sorption on Fe and Al oxides, clay interaction, interaction with dissolved and particulate organic carbon and incorporation in solid solutions) are taken into consideration. Inclusion of dissolved and particulate organic matter in the model is important to properly describe release of the low concentration trace constituents observed in the leachate. The CSF allows the prediction of release under different redox and degradation conditions in the landfill by modifying the redox status and level of dissolved and particulate organic matter in the model runs. The CSF for MSW provides a useful starting point for comparing leachate data from other MSW landfills.

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#### 1. Introduction

Recent characterisation of materials, such as soil, contaminated soil, sediments, sludge, compost, wood, coal combustion residues, waste and construction products by means of more extended leaching tests and associated chemical speciation modelling has led to much more detailed insights into release controlling pro-

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http://dx.doi.org/10.1016/j.wasman.2016.07.032 0956-053X/© 2016 Elsevier Ltd. All rights reserved. cesses (Kosson et al., 2002, 2009, 2014; Dijkstra et al., 2004; Carter et al., 2009; Schoknecht et al., 2005; van der Sloot et al., 1997, 2007a, 2007c; van der Sloot, 2002; van der Sloot and Dijkstra, 2004; van der Sloot and Eikelboom, 2003). Similarity in release controlling factors across this wide spectrum of materials provides the potential for full mechanistic impact assessments by making use of a chemical speciation fingerprint (CSF) derived from the pH dependence leaching test on each of the materials (van der Sloot et al., 2007a, 2007c; Kosson et al., 2014). In the case of landfilling, the potential prediction of constituent release from a landfill cell or site is complicated by the fact that the material to be evaluated is generally heterogeneous. This heterogeneity may be apparent macroscopically (visual) and in terms of content, but the behaviour of the material may very well be far more consistent in case of leaching (van der Sloot and Dijkstra, 2004) because of volumetric integration by the leaching process (local equilibrium)

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Abbreviations: MSW, municipal solid waste; PNW, predominantly nondegradable waste; CSF, chemical speciation fingerprint; TCLP, toxicity characteristic leaching procedure; LEAF, leaching environmental assessment framework; HA, humic acid; FA, fulvic acid; DHA, dissolved humic acid; SHA, solid humic acid; DOC, dissolved organic carbon; TOC, total organic carbon; POM, particulate organic matter; HFO, hydrated iron oxide; SI, saturation index; L/S, liquid to solid ratio (L/kg); pH+pe, measure of redox state.

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and thermodynamically imposed consistency in liquid-solid partitioning at a macro-scale based on the dominant system chemistry.

Improved insights into the release controlling factors are needed to be able to better design and control release from landfills and, where possible, manage wastes in a manner that is more sustainable (Mathlener et al., 2006; Van Vossen et al., 2009; Heimovaara et al., 2013). In the framework of the Dutch Sustainability project (Mathlener et al., 2006) municipal solid waste and predominantly non-degradable waste (subsequently referred to as PNW) were studied in detail at laboratory-scale and using field lysimeters and large pilot-scale test landfill cells. The objectives of this paper are to (i) compare laboratory and field testing results for MSW and non-degradable waste matrices to identify to what extent similarities in leaching behaviour exist, and (ii) illustrate the usefulness of chemical speciation modelling through use of a CSF to evaluate factors controlling leaching. It is important to recognize that after degradation of organic rich waste, such as typical municipal solid waste (MSW), a residual material remains, that has similarities with the predominantly non-degradable waste studied already in detail (van der Sloot et al., 2001a,b; van Zomeren et al., 2005). In the review paper by Kjeldsen et al. (2002), the factors controlling metal release from MSW are discussed and studies by others are highlighted. The overall conclusion is that there are still many unknowns due to multiple interactions. This paper seeks to reduce a number of the uncertainties surrounding release of inorganic substances from MSW landfills. It focuses on calibration of a mechanistic leaching model based on pH static experiments; the application of the calibrated model to column leaching tests (with a further limited calibration step); the application of the calibrated model to assess data obtained from field sites; and finally the application of the calibrated model to assess effects of redox and variation in DOC levels resulting from organic matter degradation.

#### 2. Experimental

The waste matrices selected are representative of a group of waste mixes covering a significant portion of wastes typically landfilled in practice and for which release behaviour is expected to have many common aspects.

#### 2.1. Materials

Both wastes discussed here were evaluated using leaching characterisation tests and reported earlier (Luning et al., 2006; van Zomeren and van der Sloot, 2006a, 2006b) and are analogous to and directly comparable with the US EPA leaching environmental assessment (LEAF) tests (Garrabrants et al., 2011, 2012). Field test results have been reported in the framework of the Dutch Sustainable landfill project (Mathlener et al., 2006; Oonk et al., 2013). A comparison of laboratory, lysimeter and field scale testing on two waste types was reported in Kosson et al. (2014). The present work focuses on modelling the release behaviour from these different matrices based on characterisation leaching test results in comparison with leachate from MSW and PNW landfills and accounting for differences in conditions between laboratory and field.

#### 2.1.1. MSW

A composite sample of MSW representing organic rich waste was prepared from separately collected samples of waste prior to landfilling for testing the release behaviour from the bioreactor (45,000 m<sup>3</sup>) operated at the Landgraaf landfill (Luning et al., 2006). Test results for this waste have been reported in the context of the Dutch Sustainable Landfill Project (2006). In 2010 the 8 year old pilot cell was dismantled, which provided the opportunity to sample material from different spatially distributed locations within the cell subjected to leachate recirculation and aeration cycles (Oonk et al., 2013). Composite samples were subjected to laboratory characterisation leaching tests (EN 14429, 2015; PrEN 14405, 2015), while individual samples were tested using a single step batch leaching test (EN12457-2, 2002; extraction with deionized water at liquid/solid ratio of 10 mL/g).

#### 2.1.2. Mixed waste

A waste mixture of predominantly non-degradable waste (PNW) was prepared from volumetrically representative portions of the waste delivered at the landfill test cell (Nauerna Landfill, NL). These waste samples were mixed to constitute a composite waste for testing at lysimeter and at lab scale (van Zomeren et al., 2005; van Zomeren and van der Sloot, 2006a). The main components in the waste mixture were soil cleaning residues, contaminated soil, sediments, small industrial waste streams, and construction and demolition waste. Only largely degraded organic matter was allowed in this pilot, hence the term predominantly non-degradable waste (PNW). Leachate from the pilot cell was collected.

#### 2.2. Methods

#### 2.2.1. Leaching tests

The upflow percolation test (PrEN 14405, 2015; continuous elution with deionized water and 7 eluate collection intervals ranging from L/S 0.2–10 mL/g and linear flow velocity of 15 cm/day) and pH dependence leaching test (EN 14429, 2015; parallel batch extraction at L/S 10 mL/g with various acid and base additions to attain specified endpoint pH values) were performed on the composite of collected waste samples. More detailed descriptions of the procedures are given in (van der Sloot et al., 1997). Similar methods and applicable method reproducibility statistics are described in Kosson et al. (2002), Lopez Meza et al. (2008) and Garrabrants et al. (2011, 2012).

#### 2.2.2. 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 recommended by the International Humic Substances Society (IHHS) 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 amounts of amorphous and crystalline iron (hydr)oxides in the waste mixture were estimated by a dithionite extraction (Kostka and Luther, 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. The methods now have been standardised in ISO/TS 12782 parts 1-5 (2011). The clay content of the samples was quantified by a sedimentation method (NEN 5753, 1994).

#### 2.2.3. Chemical analysis

The leachates and extracts from laboratory tests were analysed for major, minor and trace elements by ICP-OES (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 ionchromatography. Unless measurements are close to the detection limits of the analytical methods employed, the measurement error is generally small compared to the uncertainty involved in testing

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