

Development of a prediction model for the assessment of the middle and long term emission behaviour of inorganic wastes

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Abstract

For the development of a prediction model for the assessment of the middle and long term emission behaviour of inorganic wastes, laboratory tests and the pollutant flux model have to be evaluated. In our project landfill discharge and 24 m³ field lysimeter discharge of weathered inorganic wastes are on the one hand compared to the results of standardized tests and on the other hand to a combination of artificial weathering methods, leaching tests and discharge calculations. Preliminary results indicate that reliable calculation of the middle and long term pollutant flux of a landfill is possible for most components. The calculated flux, using laboratory data, fit the measured landfill lysimeter flux very well. The variation factor is for many components lower than two. Only the redox sensitive elements Fe, Cu, and V show a variation factor of about one order of magnitude. To build a broad basis for our pollutant flux model, eight weathered materials are investigated right now by studying their landfill discharge and performing a variety of laboratory weathering and leaching tests as well as simultaneous geochemical modelling.

Keywords: long term behaviour, inorganic wastes, pollutant flux, weathering, test methods, modelling

1 Introduction

Modern society puts a lot of pollutants in circulation. Many of them are then enriched as wastes in artificial created sinks like multibarrier landfills. Due to the fact, that the technical barrier systems will not last forever, it depends on the middle and long term behaviour of the wastes itself ("inner physico-chemical barrier"), if the landfill will become a source of contamination again. The aim of our study is to develop a practicable prediction model for the assessment of the middle and long term pollutant flux out of inorganic wastes into the drinking water and/or groundwater.

2 Background

In cold and dry climates, where mechanical erosion dominates, pollutant loaded particles are transported by wind. In moderate climates, where chemical weathering dominates, the transport of soluble contaminants by water is crucial. A lot of wastes that are disposed of in (mono-) landfills are residues of thermal processes e.g. waste incineration or metallurgic processes. Due to high temperature processes the wastes are thermodynamically instable (reactive) especially under wet conditions which means they change their mineralogical and physico-chemical properties as long as they are in a "quasi equilibrium" with their environment.

The relatively short initial and instable stage of a landfill, the "aftercare period" which lasts 30 to 50 years, is characterized by the washoff of soluble salts and dropping pH values due to carbonation processes and other internal chemical and physical reactions. This stage is well characterized and datas are available from some field studies (Johnson 1998, Fichtel 1984, Ochs 1999a,b). In the medium term the system is buffered by carbonates at pH values about 8. Depending mainly on the ANC, the Ca content, pCO₂ and the hydrological processes, this period will last for some 1.000 to 10.000 years ("carbonate buffer period"). How the landfill material will behave in the long run, when the climate may have changed, the CO₃ buffer will be exhausted and the pH will drop below 7 is still under investigation. Silicates and Al- and Fe-(hydr)oxides could buffer the system and affect the heavy metal mobility (Zevenbergen 1994).

The aim of this study is to develop a practicable prediction model to assess the pollutant flux out of inorganic wastes into groundwater and/or drinking water with a precision better than one order of magnitude (at this stage). Due to the imminence and duration we focus our investigations on the medium term period ("carbonate buffer period"). The system borders we have chosen are the borders of the landfill itself. Further ongoing processes e.g. interactions with the aquifer and biosphere are not target of this project and have to be worked out in cooperation with hydrogeologists and biologists.

3 Pollutant flux model

The pollutant flux is the quantity of pollutants that is moved over a specific period of time (Equ. 1).

Equ.1: $PF = Q_p \times T_x$ (PF ... pollutant flux; Q_p ... pollutant quantity; T_x ... time period)

The concentrations are governed by the leaching behaviour which will change due to weathering reactions under the influence of water and gas and with time. The pollutant itself is transported by a specific amount of water which results in a **concentration** (C_p). The total amount of discharge over a specific period of time depends on **sub surface discharge (SSD)** of the top soil and the **landfill area (A)** (see Figure 1). So Equ. 1 can be converted into Equ. 2:

Equ. 2: $PF_p = C_p \times SSD \times A = [mg/l] \times [l/m^2/year] \times [m^2] = [mg/year]$

For the implementation of relevant parameters into this prediction model the crucial factors, processes and timescales have to be identified, evaluated and rated and the data sources have to be selected.

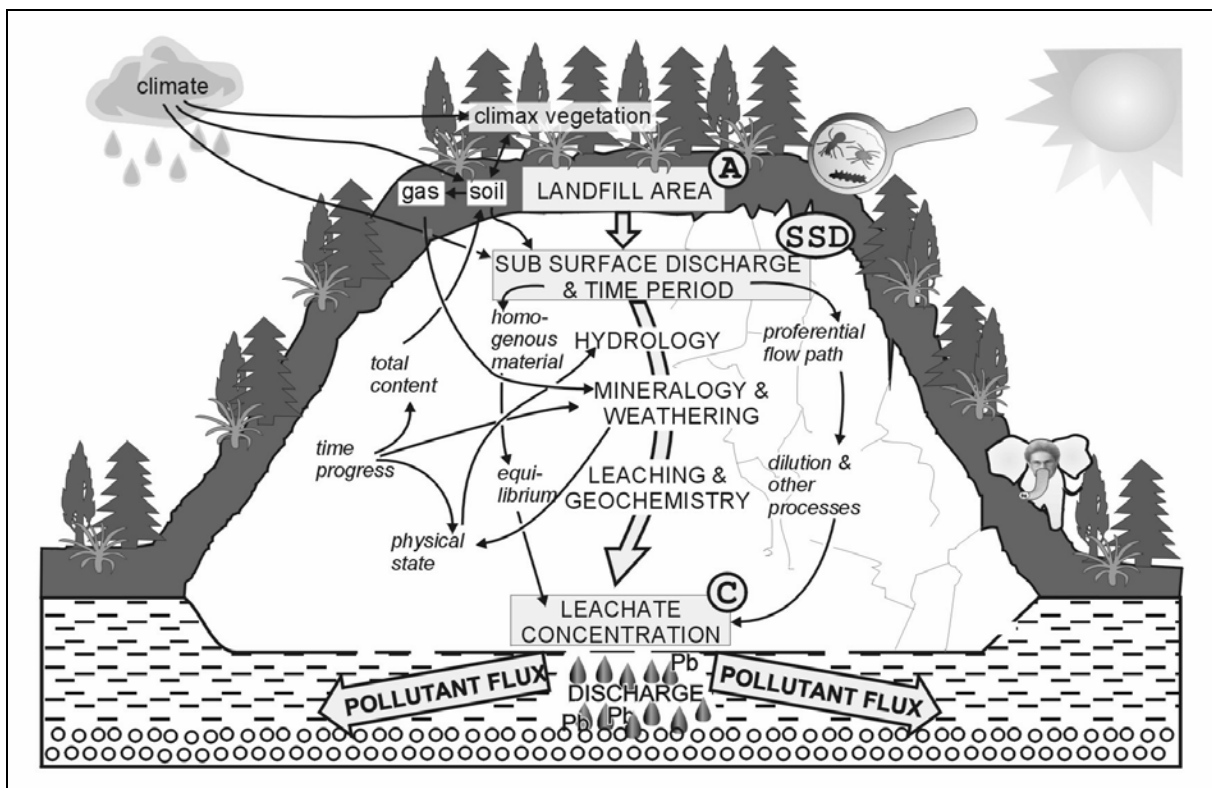


Figure 1: Processes, factors and time influencing the pollutant flux of a landfill / heap.

3.1 Discharge rate

Since water is the transport medium for the pollutants we have to focus on the sources, factors and processes which govern the discharge. Climatic conditions (e.g. precipitation characteristics, temperature and wind) vegetation and the type of surface soil determine the water balance. A significant part of the annual precipitation is evaporated either at the surface (evaporation) or after interception by plants (transpiration). The difference (sub surface discharge, SSD) is crucial for the annual discharge amount. The landfill itself then modifies the sub surface discharge pattern due to the physical and hydrological characteristics of the material. Discharge pattern depend thus also on the permeability and homogeneity of the material and the presence of preferential flow paths. Findings by JOHNSON (1998) show for a 4 years old landfill, that in the presence of preferential flow paths the discharge of a MSWI landfill is characterized by long periods of low and nearly constant flow interspersed with an increase in discharge quantity in response to rain events. Rain events, their intensity, the presence of preferential flow paths and the water content of the landfill determine the higher discharge rates.

With respect to the unsteady pattern of rain events and because preferential flow processes are neither well known nor can be modeled or deduced from experiments until now, discharge fluctuations due to episodic rain events and preferential flow paths can not be implemented into a rather simple prediction model by now.

Annual leachate amounts depend strongly on the surface vegetation of the landfill. In the beginning, evapotranspiration from the uncovered waste itself will primarily define water balance. After a few centuries a sitespecific vegetation will have been established at the landfill site and define the water balance. Coarse discharge estimations, based on a review on water balances of several landfills (Markwart 1990, Brechtel 1984, Clodius 1963, Ehrig 1990, Hartmann F. (2000), Lunzer 1998, Maile & Bidlinmaier 1995, Mostbauer 2000, Marques&Hogland 1999, Nolting 1995), forest areas, grasslands, lysimeters and taking into account own investigations, give an annual discharge amount of 10% to 60 % of the annual precipitation (a.p.), when the a.p. is <700 mm and 20 to 65 %, when the a.p. is >700 mm. Due to the fact that climatic conditions, vegetation and soil properties vary between different landfill sites and the last two factors also vary within time, would make a prediction model, by providing and incorporating all these datas, very complex. Nevertheless it would be afflicted with uncertainties (see chapter 3.3). Therefore we have to simplify and restrict the input datas. Regarding medium and long timescales there will be a constant vegetation cover defined by climatic and soil condition. This can be called a "climax state vegetation" and can be defined as the vegetation that will be established when local climate, soil formation and vegetation have reached a quasi equilibrium state. The type of soil itself depends, among other factors on the primary landfill surface, the vegetation and the climate. The water balance which will be regulated by this natural "**climax state vegetation**" and soil properties will **determine the discharge rate**. Considering a longer time period (some years) the sub surface discharge will be in agreement with the landfill discharge rate. So the annual sub surface discharge related to the "climax state vegetation" under site specific soil and climatic conditions provide the next input parameter for the prediction model and can be found in literature.

3.2 Weathering

Besides the chemical composition, the physical properties, and the environmental forces the mineralogical composition particularly affects the leaching behaviour of inorganic wastes. During ageing processes thermodynamical equilibrium with the environment is strived for, whereby a sequence of secondary mineral neof ormations and transformations (oxide → hydroxide → carbonate → silicate → ...) is passed through. In the short and medium term hydration, oxidation and carbonation of the major minerals and components (Si, Ca, Al, Fe) are the dominating processes which among others determine the porewater pH and thus also the dissolution/precipitation, sorption and complexation reactions of the heavy metal cations (e.g. cations of Pb, Cd, Zn and Cu) or mobility of heavy metal anions (e.g. CrO_4^{2-} , MoO_4^{2-} or HVO_4^{2-}). The dominating weathering reaction path is described in detail elsewhere (Rautner 2000). All these mineralogical and chemical changes are accompanied by physical changes like pore cementations, changes in grain size and pore size distribution and consequently changing hydrological characteristics.

Since these weathering reactions and states affect the leaching and discharge particularely, they have to be considered in a prediction model whereby many **weathering processes** can be simulated and **incorporated into laboratory tests** (Rautner 2000).

3.3 Leaching behaviour

The leaching behaviour of wastes depends on mineralogical, geochemical and hydrological processes and will change due to weathering reactions under the influence of water and gases. Johnson (1998) showed for a MSWI bottom ash landfill that the discharge composition is quite constant and in quasi equilibrium with the material most of the time. In this case, discharge represents the leachate (porewater) composition and can be partially described by thermodynamic calculations. During rain events the leachate is mixed with rainwater followed by rapid (hours to days) increase in discharge volume and synchronous decrease of the electrical conductivity which indicates the presence of preferential flow path. Major discharge constituents and most heavy metals behave conservatively and show a strong correlation to the decreasing electrical conductivity. A few components such as Al and Cu do not follow this behaviour which indicates other overlying processes than dilution. Isotope studies and tracer methods in combination with a simple dilution model showed that rainwater, which passes through the landfill with little interactions with the material make up 20% to max. 80 % of the discharge volume during summer rain events and around 10 % in winter months.

The discharge quality of long and constant low flow periods under "quasi equilibrium" conditions is "predictable" by tests and modelling and show higher concentrations for almost all elements compared to the discharge in response to rain events. Therefore, with respect also to the unsteady pattern of rain events and because preferential flow processes are not well known until now, "**quasi equilibrium discharge**" should be used as a parameter to specify the environmental compatibility of the total discharge. Since there exist just a few datas for some types of wastes in a weathered state, datas have to be compiled from **laboratory tests**. The strategy (methodology) for these tests has to consider as many factors and processes as possible.

3.4 Time

Two types of "time factors" have to be considered. On the one hand, we have to know how fast the physico chemical state of the wastes changes (time progress) and how long defined weathering states (e.g. "carbonate buffer period") will last. The duration of a weathering state can be estimated by the knowledge of the ANC and the sub surface discharge to landfill (material) ratio (L/S). The "**time progress**" is accompanied by changes in behaviour, therefore has to be incorporated into the test procedure and is represented by an **increasing liquid to solid (L/S) ratio**. On the other hand we focus on the flux, respectively which quantity of contaminants will be moved within a specific "**time period**", which is connected to the **sub surface discharge rate** (see chapter 3.1).

3.5 Landfill size

If we state quasi geochemical equilibrium conditions in the medium and long term, average porewater concentrations do not change with depth (below a certain depth). The volume and thus the height of the landfill only influences the duration of each period (e.g. exhaustion of salts or carbonate buffer) and can be deduced e.g. from the ANC and calculated L/S ratios. This means that the **landfill area**, as a fixed size, provide an additional input parameter.

4 Methodology

Leachate concentrations (C) for the pollutant flux model are provided by laboratory weathering methods and leaching tests. Therefore standardized and modified test methods have to be evaluated, to what extent they reflect natural discharge behaviour of longterm weathered wastes.

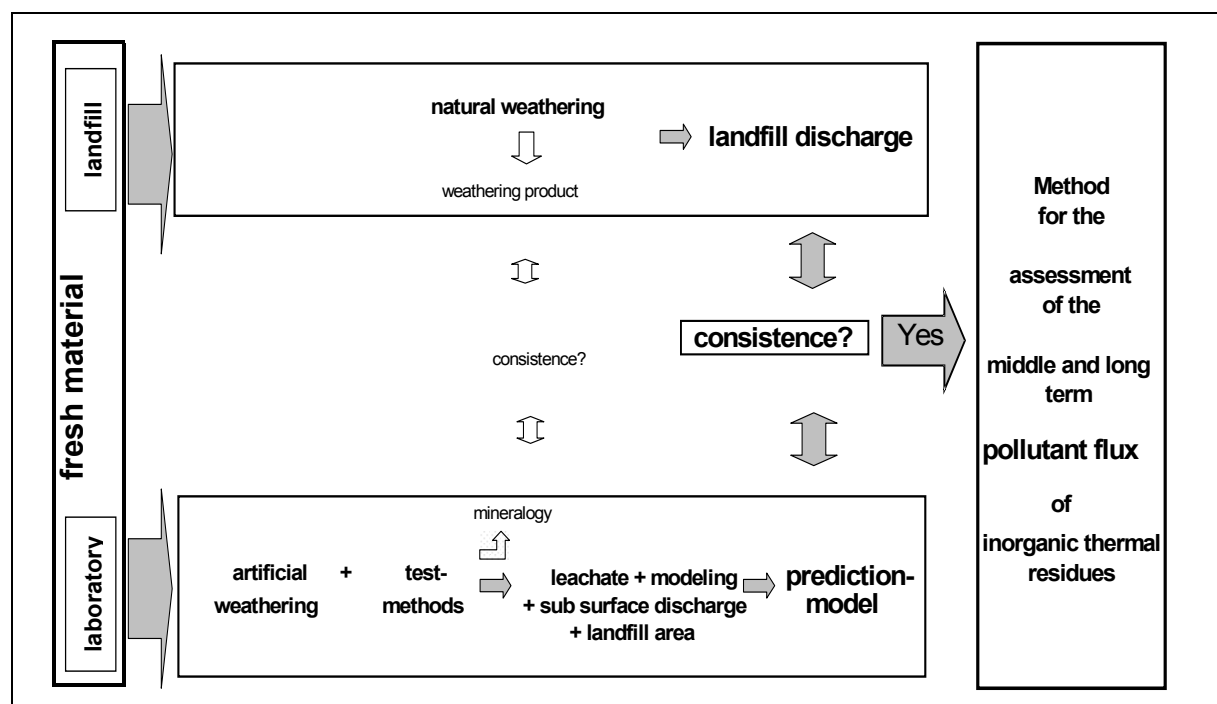


Figure 2: Process chart for the evaluation of an assessment method regarding the middle and long term pollutant flux of inorganic thermal residues.

In our project, discharge originating from landfills or 24 m³ field lysimeters containing inorganic, polluted and weathered wastes are on the one hand compared to the results of standardized tests and on the other hand to a combination of artificial weathering methods, leaching tests and discharge calculations (Figure 2). Since the „carbonate state“ is a buffered system geochemical behaviour does not change very much with the age of the „carbonate state“ material (except salts). Thus only the state of the selected material is crucial.

5 Materials and Methods

The investigated materials include bottom ashes and ashes of MSWI, rotary drum kiln slags from hazardous waste incineration, fluidized bed incineration ash, lignite fly ash, steel slags, a solidification product, galvanic sludge and Pb/Zn mineralized limestone (Table 1).

material		age of landfill/ heap	landfill/ heap discharge	24 m ³ field lysimeter	laboratory	discharge pH
MSWI bottom ash RAU 2	(S2)	12 a	X	X	X	8,0 – 8,5
MSWI bottom ash IGLO	(S3)	3 a		X	X	11,0 – 8,5
MSWI bottom ash + fluidized bed incineration ash	(A)	11 a		X	X	7,5 – 8,2
MSWI bottom ashes + rotary drum kiln slag + fluidized bed incineration ash	(S)	11 a		X	X	7,3 – 8,0
lignite fly ash	(Pi)	>25 a	X		X	~8,3
steel slag	(KA)	20 – 60 a	X	X	X	~8,5/~11,6
solidification product	(SB)	1 – 3 a		X	X	~8,1/~12,7
galvanic sludge (Ca(OH) ₂ neutralized)	(My)	5 – 25 a	X		X	8,2
Pb/Zn mineralized limestone	(Ga)	150 a	X		X	8,3

Table 1: Investigated materials, age, type of leachates and discharge pH.

These naturally weathered materials and their landfill or field lysimeter discharge have been collected and specified. The criteria for the selection of the materials have been a discharge pH of about 8 and a carbonate buffer in the solid. Corresponding fresh materials are artificially weathered with a water saturated air/CO₂ mixture, leached by different test methods and the results are now compared to the above mentioned discharge. At the same time fresh, artificially and naturally weathered materials were leached by standardized test methods. The test methods are shown in Table 2.

test method	naturally weathered landfill material	artificially weathered fresh material	fresh material	L/S	pH	grainsize
DEV – S4	X	X	X	10 + 5/10/15/ 20/30	own	ground, 2, 10 and 16 mm
CO ₂ test	X	X	X	10	~6,5	ground, 2, and 16 mm
modified CO ₂ test	X	X	X	10	~8,3	ground, 2, and 16 mm
availability	X	X	X	50	8,3 + 4,0	ground
pH-stat	X	X	X	5/10/50	13,0 – 4,0	ground
upflow percolation	X		X	0,05 - 10	own	4 mm
modified upflow percolation	X	X		0,05 - 10	own	2 mm

Table 2: Performed test methods, weathering stages and parameter variations.

34 components were analyzed in the leachates and CO₂ and O₂ in the pore gas. Weathering method is in principle the same as described in Rautner (2000) but the used gas is synthetic air with 30% CO₂. This allows oxidation and carbonation at the same time and yields good weathering processes at room temperature.

Mineralogical investigations are performed by XRD, DTA, DTG, SEM-EDAX, microprobe and microscopy.

The recent version of the geochemical code PHREEQC-2 (Parkhurst & Appelo, 1999) was used for geochemical modelling. Activity coefficients of aqueous species have been calculated with the DAVIES equation. Thermodynamic data were taken from the original PHREEQC-2 database, and in some cases from other sources (Appelo & Postma, 1996, Carrol et al., 1998, Perkins & Palmer, 1998).

In most lysimeters and percolation tests pore gas (CO₂ and O₂) was measured to verify the discharge pH values. This was done by modelling the pH with PHREEQC-2 assuming calcite and gypsum are the solubility controlling phases under the specific (measured) pore pCO₂.

Regarding artificial weathering, leaching and discharge rate, sensibility analysis are or have been performed for factors and processes such as pH, redox potential, liquid/solid ratio, salinity, contact time, grain size, water content, agitation, gas composition as well as type of soil and climate (literature studies).

6 Results

In the following chapter preliminary results of some selected tests and a lysimeter discharge are presented to illustrate if there is a conformity for specific ions in between these tests. In addition the measured pollutant flux of the big landfill lysimeter is compared to the calculated pollutant flux by using the upflow percolation data.

The presented material (S) is a 11 year old landfill material. It is a mixture of MSWI bottom ash (25%), rotary drum kiln slag (from hazardous waste incineration - 60%) and fluidized bed incineration ash (sewage sludge incineration – 15%) and is built into a 24 m³ field lysimeter. The landfill is situated east of Vienna where precipitation is about 600 mm per year. Average discharge rate for the first year was about 10 % of precipitation whereas about 90 % of this discharge was collected in winter 1999/2000. L/S ratio for the presented period (9 month) is about 0,33 to 0,36. The discharge pH (7,3 - 8,0) and the conductivity (about 11 mS/cm) are quite constant and show no dependence on the rain pattern. In combination with the discharge pattern this indicates that preferential flow paths do not exist.

In Figure 3 results (old slag S) of upflow percolation tests (median of quadruplicate repetitions; tests will be continued to L/S 10) are compared to the lysimeter discharge, DEV-S4 tests of fresh, artificial weathered fresh material and old (landfill) material as well as pH-stat (8,3) test of fresh material at specific L/S ratios. pH values are about 8 except for the fresh material (pH 10,7). Results of geochemical modelling by PHREEQC-2 are not shown here (see RAUTNER, 2000).

Three types of component behaviour could be identified. Major components like SO₄, Cl, Na, K, Ca and Mg as well as the conductivity and the pH show little discharge variations and fit the upflow percolation data very well. Extrapolation up to an L/S of 10 also indicates good conformity for major salt ions, the shown tests and this material. Na, K, Mg and Cl concentrations are linked to easy soluble salts. Calcite and Gypsum are the solubility controlling phases for Ca and SO₄ in the old respectively artificially weathered materials, whereas Ettringite is the solubility controlling phase for Ca and SO₄ in the relatively fresh material (pH 10,7). This explains the higher Ca concentrations in the pH-stat test (at pH 8,3). Geochemical modelling for Ca and SO₄ for similar materials see Figure 2 and 3 in RAUTNER (2000).

Most discharge trace element concentrations also lie in the range of the upflow percolation test (valide for Me- cations as well as for Me- anions) but show a much higher variability. One factor could be the low concentration range which makes the metal concentrations very sensitive to differing local equilibria. On the other hand a higher variability during dry periods indicates that precipitation/dissolution reactions (GERVAIS,1999) could play an important role. Clarification will be yielded by the next seasons (hopefully).

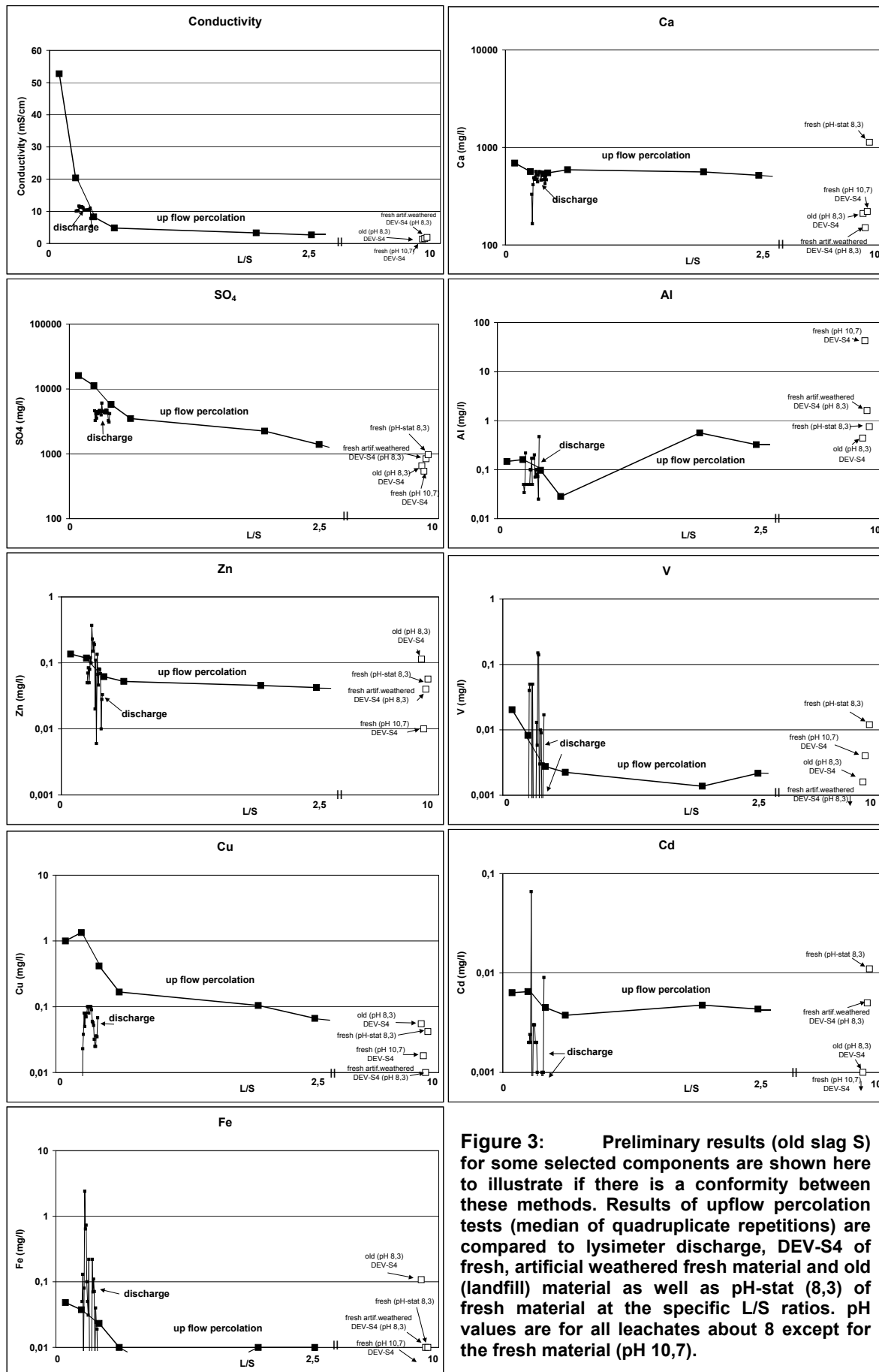


Figure 3: Preliminary results (old slag S) for some selected components are shown here to illustrate if there is a conformity between these methods. Results of upflow percolation tests (median of quadruplicate repetitions) are compared to lysimeter discharge, DEV-S4 of fresh, artificial weathered fresh material and old (landfill) material as well as pH-stat (8,3) of fresh material at the specific L/S ratios. pH values are for all leachates about 8 except for the fresh material (pH 10,7).

For the discharge and the upflow percolation test willemite (Zn_2SiO_4) seems to be the solubility controlling phase for Zn (see RAUTNER, 2000). The pH-stat (pH 8,3) and the batch tests at L/S 10 yield concentrations which vary up to two orders of magnitudes.

Discharge concentrations of Cd, Pb, Cu and Fe (the third identified group) also show a high variability. Pb concentrations are most the time below detection limit for the lysimeter discharge as well as for the upflow percolation test. Compared to the upflow percolation test Cd and Cu show overall lower discharge concentrations whereas Fe values from winter discharge throughout outrange the percolation test. One reason could be surface complexation and/or surface precipitation processes of Cd and Pb to/with Fe- and/or Al-hydroxides under landfill conditions (MEIMA, 1997) which eventually cannot be reproduced quite well with this type of upflow percolation test. On the other hand the redox state may have changed in the lysimeter. Relatively high Fe values in winter (water saturated material) indicate that Fe^{2+} is dissolved whereas decreasing Fe concentrations (below quantification limit) in the summer discharge (dry material) indicate that oxidized Fe^{3+} phases are the solubility controlling phases (see also the paragraph below). Investigations are carried out right now. Regarding Cu the following scenario is possible: the Cu concentrations in the field lysimeter are determined by the solubility of tenorite (CuO). It seems that brochantite ($Cu_4SO_4(OH)_6 \cdot 1,3H_2O$) has already been discharged in the landfill within the first 10 years (before sampling) whereas in the upflow percolation leachate, brochantite is the solubility controlling phase up to an L/S of about 0,5. This can be explained by the neoformation of brochantite due to sample preparation (creation of fresh surfaces). At higher L/S ratios in the upflow percolation test this phase is depleted and tenorite controls the Cu concentrations.

Component	measured flux (mg)	calculated flux (mg)	variation factor	landfill flux (g) / year
	24m ³ field lysimeter - first period	upflow percolation – first period	lysimeter / upflow percolation	at L/S 2,5 (in about 2000 years)
SO₄²⁻	1888190,0	3119850,0	-1,7	7515000
Na	587358,0	561600,0	+ 1,1	136800
Ca	204332,0	295785,0	- 1,4	2795400
Fe	223,0	12,6	+ 17,8	54
Al	49,0	52,3	- 1,1	1746
Zn	43,0	33,5	+ 1,3	229
Cu	25,1	224,6	- 9,0	360
V	11,7	1,5	+ 7,9	12
Cd	5,0	2,4	+ 2,1	23

Table 3: Measured pollutant flux of the big landfill lysimeter compared to the calculated pollutant flux. Positive variation factors indicate that flux under natural conditions is underestimated by this type of laboratory test whereas negative factors indicate overestimation. Calculated pollutant flux for the below described landfill in about 2000 years is presented in the right column.

For the evaluation of the pollutant flux model in a first step the measured pollutant flux of the big landfill lysimeter is compared to the calculated pollutant flux by using Equation 2 ($PF_p = C_p \times SSD \times A = [mg/a]$). The upflow percolation data yield the concentration (C_p). For SSD the lysimeter sub surface discharge rate is used whereas the area is defined by the lysimeter area. Results are presented for the naturally weathered 11 year old material (Table 3), the first period and the above presented components. Except for Fe, Cu and V which show a variation about one order of magnitude, calculated data fit the measured data very well. It is noticeable that the higher variations occur with redox sensitive elements (see also the paragraph above). On the other hand this low (0,33-0,36) L/S ratio (salt) concentrations show high decrease rates with little L/S variation (see Figures 3). This makes the calculated flux very sensitive to the selected L/S ratio of the upflow percolation test at low L/S ratios. Sensitivity will therefore be smaller when L/S ratio is above 0,6. Calculations for an L/S ratio of 2,5 are also shown in Table 3 by assuming the same landfill location (\rightarrow SSD), an landfill area of 300 m x 300 m and a height of 30 m. Under the same climatic conditions and for this material the L/S of 2,5 will be reached in about 2000 years.

7 Conclusions

For the development of a prediction model to assess the middle and long term emission behaviour of inorganic wastes, standardized and modified test methods and a pollutant flux model are evaluated. Preliminary results indicate that a prediction of the middle and long term pollutant flux of a landfill with uncertainties less than one order of magnitude is possible for most components.

Concentrations (C), derived by upflow percolation tests using weathered wastes, the sub surface discharge (SSD) and the landfill area (A) are the input parameters for the pollutant flux equation ($PF_p = C_p \times SSD \times A = [mg/l] \times [l/m^2/a] \times [m^2] = [mg/a]$). Major salt ion concentrations and most trace element concentrations can be derived very well from laboratory methods. These test methods can be a combination of artificial weathering and upflow percolation tests. The calculated flux, using laboratory data, fit the measured landfill lysimeter flux very well. For most components the variation factor is lower than two. Only the redox sensitive elements Fe, Cu, and V show a variation factor of about one order of magnitude. It seems that some landfill processes like redox processes or surface complexation and/or surface precipitation of Cd and Pb eventually cannot be reproduced quite well with this type of upflow percolation test.

To build a broad basis for our pollutant flux model eight weathered materials are investigated right now by studying their landfill discharge and performing a variety of laboratory weathering and leaching tests as well as simultaneous geochemical modelling.

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Table 1: Investigated materials, age, type of leachates and discharge pH.

Table 2: Performed test methods, weathering stages and parameter variations.

Table 3: Measured pollutant flux of the big landfill lysimeter compared to the calculated pollutant flux. Positive variation factors indicate that flux under natural conditions is underestimated by this type of laboratory test whereas negative factors indicate overestimation. Calculated pollutant flux for the above described landfill in about 2000 years are presented in the right column.

Figure 1: Processes, factors and time influencing the long term pollutant flux of a landfill / heap.

Figure 2: Process chart for the evaluation of an assessment method regarding the middle and long term pollutant flux of inorganic thermal residues.

Figure 3: Preliminary results (old slag S) for some selected components are shown here to illustrate if there is a conformity between these methods. Results of upflow percolation tests (median of quadruplicate repetitions) are compared to lysimeter discharge, DEV-S4 of fresh, artificial weathered fresh material and old (landfill) material as well as pH-stat (8,3) of fresh material at the specific L/S ratios. pH values are for all leachates about 8 except for the fresh material (pH 10,7).