Experimental percolation under intermittent conditions: Influence on pollutants emission from waste

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Abstract

As a precautionary measure, the re-use (or landfill) of waste requires an environmental assessment of its potential impact. This assessment is usually made by simulating the emission of pollutants with a predictive model based on laboratory tests (standardised batch leaching tests, up-flow percolation tests, acid neutralisation capacity tests [CEN, Characterisation of Waste – Leaching – Compliance Test for Leaching of Granular Waste Materials and Sludges, European Committee for Standardisation (ECS), Brussels, 2002 [1]; CEN, prCEN/TS 14405 Characterisation of Waste – Leaching Behaviour Tests – Up-flow Percolation Test (under specified conditions), ECS, Brussels, 2002 [2]; CEN, prCEN/TS 14429 Characterisation of Waste – Leaching Behaviour Test – Influence of pH on Leaching with Initial Acid/base Addition, ECS, Brussels, 2003 [3]]. These tests are performed with simpler conditions than those occurring in the scenario of re-use (saturated media, permanent inflow ...).

In order to evaluate the relevance of these tests to be considered as a reference for predictive model, the purpose of this work is to determine how the intermittent hydrodynamic flow influences the pollutants release of unsaturated waste. As a result, we could estimate whether this parameter should be introduced in the model.
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1. Introduction

The organisation of industrial societies binds its actors to manage wastes, from an environmental and socio-economic point of view. The assessment of different waste management strategies is necessary to minimise its potential impact on health and environment. For instance, the potential impact associated to the re-use of thermal process residues in road techniques is to be assessed in terms of emission of pollutants under various percolation conditions. The European methodology of evaluation EN 12-920, based on parametric laboratory tests and predictive model is usually used to assess this potential impact [4]. Nevertheless, the real emission of the waste in its re-use scenario is much more complex than what might result from these laboratory tests. In particular, the inflow intermittence modifies the water content, the repartition of the gas phase in the media, the chemical reactions and their kinetics ..., in time and space. That is why there remain questions regarding how the results of such laboratory tests can be extrapolated in order to obtain predictions of potential impacts at different scales and for different percolation scenarios. We should investigate on the minimum level of complexity for tests and models in order to describe and predict properly the behaviour of the waste.

In this context, a multiple-scale leaching project ("LIMULE" [5]) was initiated to examine the influence of scale and flow regime on the mobilization of soluble constituents from municipal solid waste incineration boiler ash and to compare the applicability and efficiency of various models over scales.

The present study is a part of this project and deals more particularly with the influence of intermittent inflow regime on chemical emission of pollutants. To explore this problematic, four down-flow columns are performed in the laboratory. The outflow and pollutant mobilization curves are compared with the results of the standardised up-flow percolation test and two down-flow lysimeter cells performed for the LIMULE program. For these devices, the low eluate analysis frequency does not show the influence of intermittent inflow on chemical species release.

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In Section 2 are described the experiments and Section 3 is devoted to the obtained results.

2. Materials and methods

2.1. Characterisation of the waste

The thermal process residues are boiler ash from a fluidized-bed MSW incineration plant, located in Gien, France. The sampling was conducted only once (18 tonnes).

The ash was mixed with a clean Loire sand (0.1–1 mm) in equal weight proportions because it had a certain tendency to indurate in conditions of prolonged contact with water. The sand–ash mixing was performed thanks to two conveyor belts and a melting tube. The stability of the mixing process was controlled every 15 min. Dry density was measured for 20 different samples of mixture and shows a good coherence of values (medium: 1.32 g/cm^3, maximum: 1.39 g/cm^3, minimun: 1.27 g/cm^3) so that the mixture could be considered as homogeneous.

The chemical composition and grain size distribution of a representative sample of mixture are given in Tables 1 and 2, respectively.

Ca, Si, Al, Fe, K, Mg, Mn, Na, P, Si, Ti were determined by X-ray fluorescence (Philips PW2400). Ag, B, Ba, Cd, Co, Cr, Cu, Mo, Ni, Pb, Sb, Sn, Sr, V, W, Zn were determined by ICP-AES after high-sintering at 450 °C and attack with Na_2O_2/HCl. Uncertainties are about 5–10%. Loss on ignition was determined at 1000 °C. The total content of metallic aluminium was analysed by atomic absorption spectrometry after mild attack with brominated methanol. The chlorine content was determined by potentiometric analysis. Total sulfur, total organic and inorganic carbon contents were analysed with a Leco determinator. The BET surface was determined according to the French standard NF X 11-621.

Table 1
Global chemical analysis of the ash–sand mixture (from [5])

Major elements	%	Traces	mg/kg
Al_2O_3	11.5	Ag	4.3
CaO	7.6	As	46
Fe_2O_3	3.64	B	88
K_2O	3.00	Ba	955
MgO	1.3	Cd	4
MnO	0.07	Co	61
Na_2O	1.7	Cr	168
P_2O_5	1.25	Cr(VI)	<5
SiO_2	67.4	Cu	1021
TiO_2	0.69	Mo	8
Weight loss (105 °C)	0.54	Ni	63
Loss on ignition (1000 °C)	1.4	Pb	473
Cl	0.43	Sb	44
Organic carbon	<0.05	Sn	155
Total carbon	0.2	Sr	235
Total sulfur	0.26	V	24
		W	19
Al^0	1.4	Zn	2053

Major elements (total concentrations expressed as oxides in %) and trace elements in mg/kg.

Table 2
Grain size percentage distribution of boiler ash by dry screen analysis (from [5])

	Grain size (μm)					BET surface area (m^2/g)
	<63	63–125	125–250	250–500	>500	
Boiler ash	9	14	30	45	2	2.61

Chemical analysis shows that the sample is rich in Si-bearing phases and to a lesser extent in Al, Ca, Fe and K. The main traces are, in decreasing order, Zn, Cu, Ba, Pb, Sn and Sr. The ash is also rich in metallic aluminium (around 1.4 wt.%).

The granulometric distribution of the ash–sand mixture has a median of 0.4 mm with a 10% fraction lower than 80 μm.

XRD patterns were recorded on a diffractometer Bragg–Brentano (type Siemens D5000, monochromator with Co Kα1 radiation λ = 17,890 Å). X-ray diffraction analysis of the ash reveals the presence of crystalline phases corresponding to quartz, silicates – mainly microcline, plagioclase, diopside and gehlenite – anhydride sulfates ($CaSO_4$), carbonates ($CaCO_3$), iron oxides – hematite – and NaCl. An amorphous fraction is also detected.

2.2. Experimental device

Leaching of the waste in intermittent conditions is investigated using four down-flow columns. The size of the columns is chosen so as to have proportional height versus diameter ratios, in order to correlate fluid outflows and pollutants' concentrations over scales. A stirred and initially dry sand–ash mixture sample is puddled uniformly in the columns with a pestle. The material is assume to be homogeneous in each column. The average densities vary from 1.27 to 1.42 g/cm^3 for the different columns. So, even with a strict filling protocol, the similarity of percolation experiments is difficult to obtain. This observation highlights the problematics of reproducibility and extrapolation from laboratory to field scales. A peristaltic pump controls inflow water supply. Water feed rate is equal for each column at a rate of 6.5 g/min and is distributed over one aspersion event of 60 min every 24 h. Given the hydraulic conductivity of the sand–ash mixture (on the order of 10^{-5} m/s), the water feed rate theoretically resulted in unsaturated downward flow conditions. However, a noticeable water accumulation at the head of columns A, B and C suggests that a part of the media is saturated in water (the volumetric saturated water content of the ash–sand mixture is of 47%).

To investigate the role of water ionic strength on the leaching of the waste, two identical columns are used, one fed with demineralised water (column A), and the other one with tap water (column B, as for columns C and D). As the accumulation of water at the head of column A is small (maximum 3 cm in height) and rapidly percolates in the material after the end of the injection, we can still consider input water in column A as demineralised.

Fluid outflow is monitored automatically by a fluid sampler. The eluates are then collected and conditioned for chemical analysis after a 0.45 μm filtration. Al, Ca, Cr, K, Na and Zn

Table 3
Summary of experimental conditions

	Dimensions	Density	Inflow rate (g/min)	Quality of water
A	$D = 10$ cm, $h = 37$ cm	1.31	6.5	Demineralised
B	$D = 10$ cm, $h = 37$ cm	1.31	6.5	Tap water
C	$D = 10$ cm, $h = 67.5$ cm	1.27	6.5	Tap water
D	$D = 20$ cm, $h = 51.5$ cm	1.42	6.5	Tap water
Up-flow percolation test	$D = 10$ cm, $h = 30$ cm	1.3	0.8	Demineralised
Indoor lysimeter	$L = 2.5$ m, $l = 1.75$ m, $h = 0.9$ m	–	16	Tap water
Outdoor lysimeter	$L = 3$ m, $l = 2$ m, $h = 2$ m	–	Rainfall	Tap water

were analysed by ICP-AES (Ultima 2 Jobin Yvon) with the standard NF EN ISO 11885. Cl^- and SO_4^{2-} were analysed by ionic chromatography (IC25 Dionex) with the standrad NF EN ISO 10304-2. Uncertainties are about 5%. Detection limit is 0.001 µg/L for Al, Ca, Cr and Zn; 0.1 µg/L for K, sulfate and chlorine; 0.008 µg/L for Na.

Waste has been leached for 18 days with a stop of 10 days between the 13th and the 14th injection to reveal the influence of long residence time of water inside the media on outflow concentrations. Experiment on large column D has been carried out for 26 days to provide a significant L/S ratio (outflow water volume per kg of dry waste) over the duration of the experiment.

In order to compare our experimental results with larger scale experiments under different percolating conditions, we used available data from lysimeter cells leached during 24 months. One lysimeter is located indoor (4 m³, 2.5 m × 1.75 m × 0.9 m) with controlled water supply (cyclic aspersion of the surface with an inflow rate of 24 L/day) and the other one is open to rainfall (12 m³, 3 m × 2 m × 2 m). The flow throught lysimeters resulted inhomogeneous. This is due to the material swelling caused by the oxydation of metallic aluminium (see Section 3.2).

The standardised up-flow percolation test [2], considered as the reference of permanent hydraulic regime, is also used to compare the results.

Finally, each device is leached with different hydraulic regime under atmospheric conditions. Table 3 summarises the experimental conditions.

3. Results

3.1. Hydraulic data

Outflow for columns A and B started after three injections, and after four injections for column C, while the larger column (D) started to flow off after 12 injections. For a same inflow rate, we can note a direct relation between the volume of dry waste and the time for the water to flow off. This indicates that the volume of water necessary to obtain an outflow depends on the initial mass of waste and is independent from the water velocity. Water transfer is balanced considering a residual volumetric water content of 27% and an average evaporation rate around 4%. Fig. 1 shows the evolution of outflow rate as a function of time and L/S ratio for column A.

The hydraulic regime reaches steady state a few days after the beginning of outflow. However, this steady state regime can slightly vary from day to day into a spindle of curves without any logical evolution, as shown in Fig. 2 for the four columns.

This may indicate a macroscopic permeability variation due to the reactivity of the waste, observed in the form of swelling and temperature variations (Section 3.2; [5,9]) and the inherent variability of water path in the media. We can notice that the pause of 10 days without injection did not influence the system (see Fig. 1).

Outflows curves of the columns A and B, whose difference is inflow water quality, are close in terms of maximum rates and evolution. However, column A have a longer tailing, which suggests inherent microscopic water path differences or existence of stagnant zones. Outflow curves of column C show a noticeable difference in hydraulic behaviour with a preferential path, which could not be predictable. As a result, the spindle of curves for
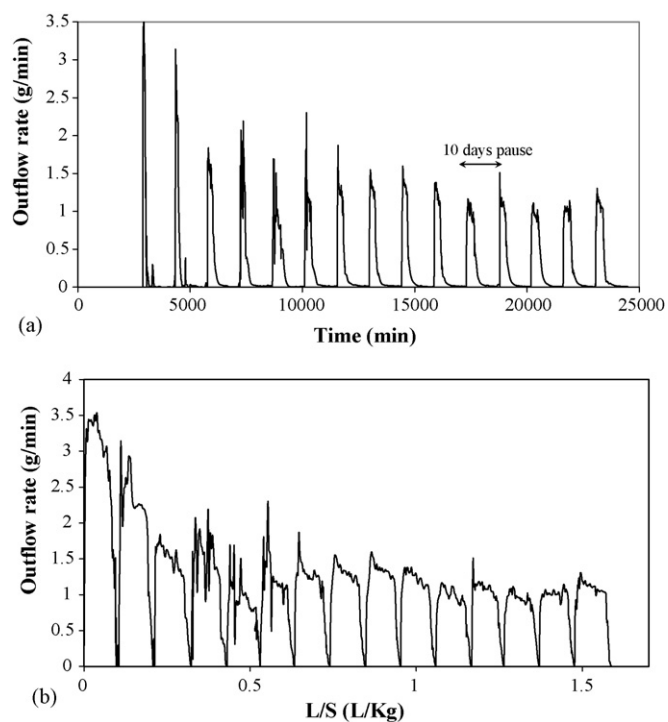

Fig. 1. Outflow rate for column A as a function of time (a) and as a function of liquid vs. solid ratio (b) for a daily injection at an inflow rate of 6.5 g/min during 60 min.
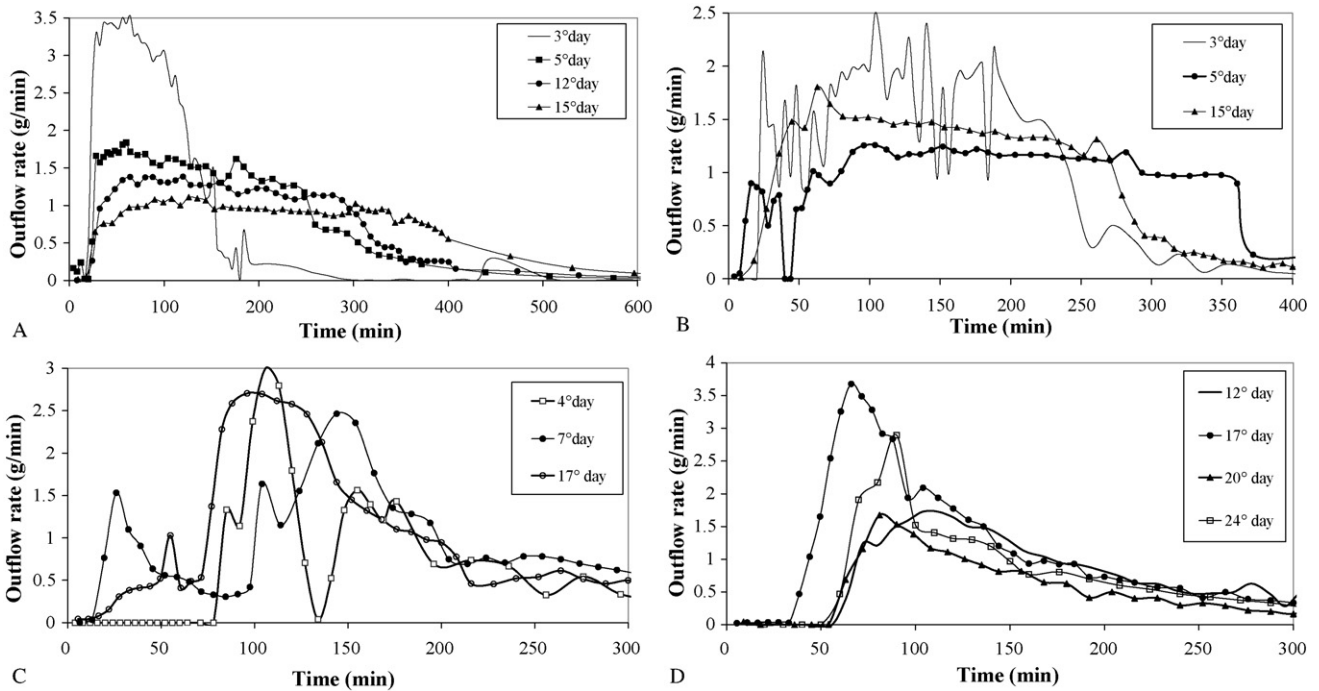
Fig. 2. Comparison of selected outflow curves as a function of time for columns A, B, C and D (zooms) with a daily injection at an inflow rate of 6.5 g/min during 60 min.

column C is wider. Column D is to be considered with a different point of view as the water velocity is lower, no accumulation on top-column is observed and so the waste is entirely unsaturated with an important dispersion.

In conclusion, we can see that, even with an homogeneous material and small columns, there is a time variability of the out-flow and a real scale effect. In real conditions, the intermittence of inflow may create a huge heterogeneity of the flow through the porous media, which can be difficult to model. We must now determine if this variability of flow have an influence on the leaching, and so, if we have to take it into account in the model.
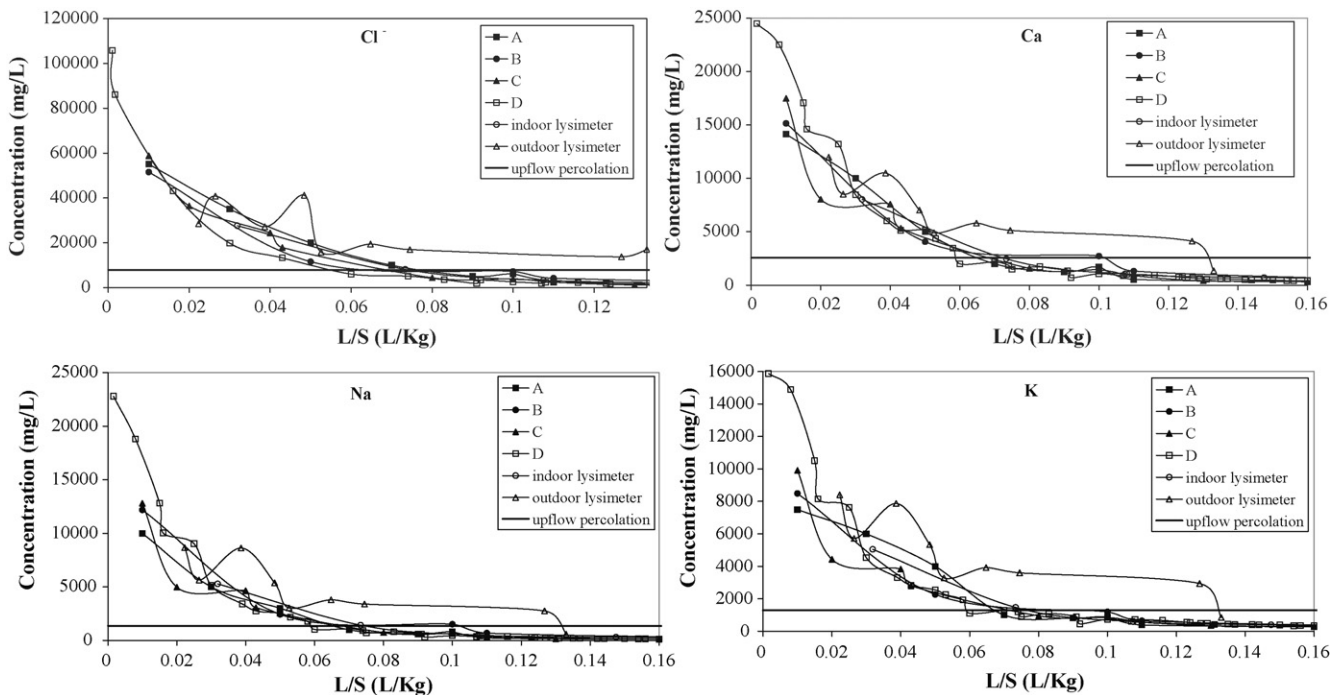

Fig. 3. Evolution of the concentration of soluble salts (Cl^-, Ca, Na, K) in the leachate as a function of L/S ratio for the colums A, B, C and D, the indoor and outdoor lysimeters and the standardised up-flow percolation test (zooms).

3.2. Leachates analysis

The evolution of soluble salts as a function of L/S ratio for each column, up-flow percolation test, indoor and outdoor lysimeters is shown in Fig. 3. Note that up-flow percolation results represent mean concentrations (rollup of water which is then sampled for the maximum L/S ratio reached), while the other results are punctual withdrawal data.

As seen in this figure, soluble salt concentrations show a rather typical exponential-type decrease with increasing L/S ratio. Intermittent inflow regime and scales do not seem to influence salts mobilization from the waste to the percolating water. The comparison of the soluble salts data collected from the different scales of percolation is seen to be very good. However, we can notice that the outdoor lysimeter salt concentrations are more variable, which correspond to a higher level of intermittence in the water feed. It should be noted that standardised up-flow percolation test underestimate the initial emission of salts, and so predictive model might not represent a punctual contamination which could cause the undrinkability of aquifer over a short period of time (1 or 2 years).

With respect to reactive constituents, Fig. 4 shows that there is an influence of hydraulic intermittence on pollutants emission. L/S ratio might not be the more adapted parameter to describe the breakthrough curves and to extrapolate the results over scales.

The sulfates are more mobilized for high water velocity through the waste in opposition to the aluminates behaviour. Moreover, this figure reveals a clear relationship between the observed evolutions of Al and Cr (note that Al and Cr releases for the outdoor lysimeter are not mentionned in these graphs

for a better legibility, and are similar to column D evolution). When there is a long time of residence of water in the porous media, there is no release of Cr and an important release of Al. In opposition, a short time of residence of water leads to a release of Cr without Al. This phenomenon is interpreted as a result of a redox control on Cr solubility in presence of metallic Al [5,9]. In alkaline conditions (pH > 10), metallic aluminium can be oxidised by OH^-, dissolved oxygen or Cr(VI). In this last redox reaction, Cr(VI) is reduced in Cr(III) which remains insoluble. To sum up, the Al/Cr correlated emission is dependent to experimental leaching conditions (variation of pH, dissolved oxygen content, flow velocity, presence of unsaturated zones, disponibility of metallic aluminium grains . . .). In consequence, the confrontation between the dynamics of flow and the kinetics of chemical reactions influences the pollutants emission. The variation of inflow intermittence and the scale of experiment modify this confrontation and so play a role in the Cr and Al releases. That is why it seems important to study the release of contaminant under variables conditions of percolation in term of intermittence, water flow velocity and scale.

As curves show poor difference between pollutants emission in columns A and B, ionic strength does not seem to influence the pollutant emission, except for Zn ions (Zn release for columns C and D, and lysimeter cells are not shown for a better legibility and are close to the up-flow percolation test value). The low ionic strength of rain water should hence affect Zn emission. The mecanism of this phenomenon could be explained by the colloidal particle mobilization due to the low ionic strength [6,7] and the preferential adsorption of Zn ions on colloids [8].
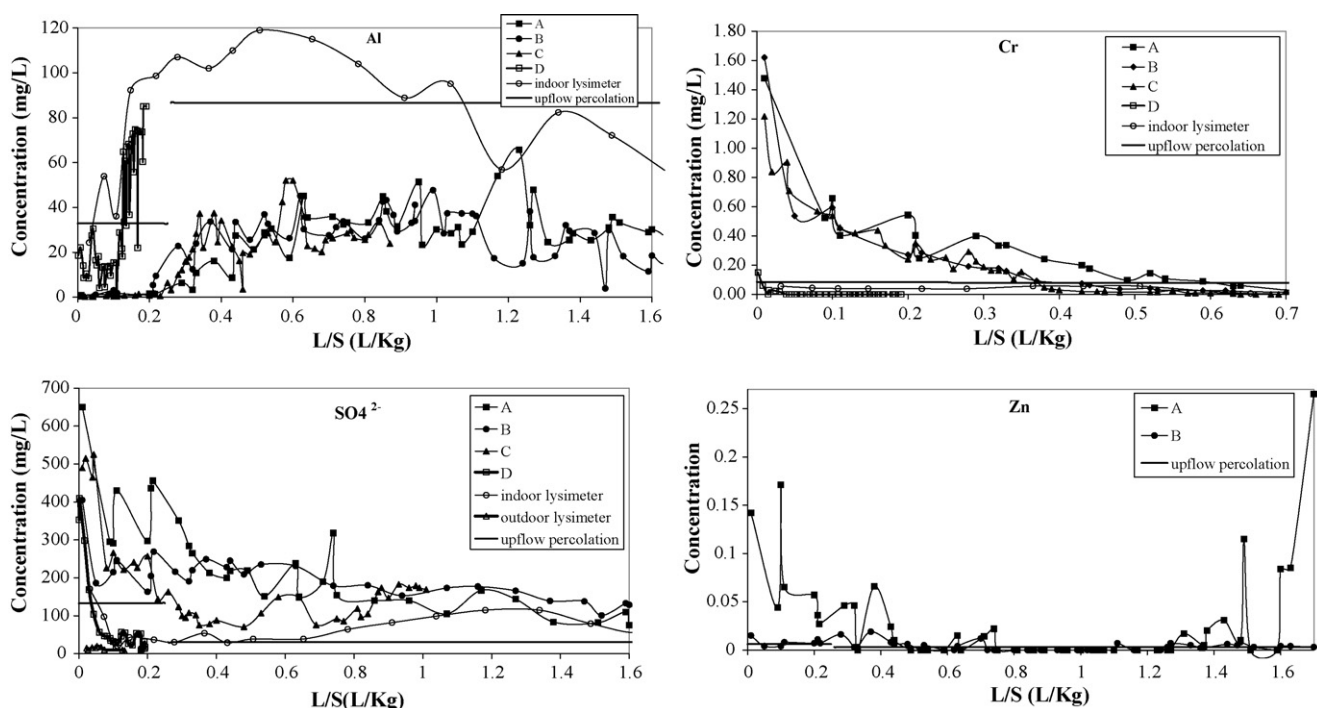

Fig. 4. Evolution of the concentration of reactive constituents (Al, Cr, SO_4^{2-}, Zn) in the leachate as a function of L/S ratio for the colums A, B, C and D, the indoor and outdoor lysimeters and the standardised up-flow percolation test (zooms).

4. Conclusion

This work shows the limits of standardised tests under permanent inflow conditions and saturated media. In the case of soluble salts, there is a good correlation between concentration evolations obtained as a function of liquid–solid ratio, at the different scales of observation and with different intermittent inflow rates. For such constituents, the data suggest that the intermittence of inflow is not an influent factor on the emission.

Meanwhile, in the case of reactive constituents, the releases result simultaneously from the inflow variability and the heterogeneity of the porous media (even more in unsaturated conditions), as well as from the complexity of mineral phases and chemical interactions. Al/Cr correlated emission is a good example of the "dynamics of flow through the waste versus kinetics of Al^0 oxidation" confrontation.

That is why it seems important to study the release of contaminant under variable conditions of percolation in term of intermittence, water flow velocity and scale.

In the other hand, mechanist or functional modellings of this multi-variable dependent system might be difficult (and inapproriated, owing to uncertainties and imprecisions on phenomenons). In this context, a fuzzy rule-based model should be more adapted. Fuzzy logic is a promising tool, which allows representing a variable and imprecise system without knowing the intern mechanisms. A fuzzy rule-based system will be soon designed to better take into account the variability and heterogeneity of the system.

Acknowledgements

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