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Round robin testing of a percolation column leaching procedure

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ABSTRACT

Round robin test results of a percolation column leaching procedure (CEN/TS 14405:2004), organised by the Flemish Institute for Technological Research (VITO), over a time span of 13 years with a participation of between 8 and 18 different laboratories are presented and discussed. Focus is on the leachability of heavy metals As, Cd, Cr, Cu, Hg, Ni, Pb and Zn from mineral waste materials. By performing statistical analyses on the obtained results, insight into the reproducibility and repeatability of the column leaching test is gathered. A ratio of 1:3 between intra- and inter-laboratory variability is found. The reproducibility of the eluates' element concentrations differ significantly between elements, materials and fractions (i.e. different liquid-to-solid ratios). The reproducibility is discussed in light of the application of the column leaching test for legal and environmental policy purposes. In addition, the performances of laboratories are compared.

1. Introduction

The column test (*CEN/TS 14405:2004*) is a percolation leaching test for soils and granular waste materials on a laboratory scale. It is a standard test used for the risk assessment of pollutants release from solid materials into seepage water, groundwater or surface waters. The test results can be used to predict the release of constituents from a waste in a specific scenario, as a function of time, by means of modelling under the hydraulic, geotechnical, hydrological, chemical, physical and biological conditions of the scenario. It simulates the leaching behaviour of a material in the short and medium term by relating the release of inorganic parameters to the liquid to solid ratio (L/S).

The column leaching test is used in Flanders (northern region of Belgium) as a legal instrument to acknowledge compliancy of waste materials that are to be used *as* or *in* building materials with environmental legislation. In the test, water is forced to flow upstream a column filled with sample material, allowing constituents to leach into the water. At 7 specific cumulative L/S ratios (0.1, 0.2, 0.5, 1, 2, 5, 10 l/kg), the leachates are collected – these are called *fractions* – and sent for chemical analysis. The exact procedure used in Flanders is defined in CMA/II/A.9.1 (Flemish Environmental Legislation, 2016) that, as well as the CEN/TS 14405:2004, is based on the Dutch NEN 7343. The main difference with CEN/TS 14405:2004 is the limitation of particle size (<4 mm) and a fixed column diameter (5 cm) of the column in which the material is tested. In CEN/TS 14405:2004, a column diameter of 10 cm is allowed as well, with a particle size <10 mm. Alternative standards for percolation column leaching tests exist worldwide, e.g. US EPA Method 1314, ASTM D4874, DIN 19528, which typically differ in the L/S at which fractions are collected, water flow rate and, in some of these methods, the upper limit of the grain size. The Flemish legal limit values for the cumulative release (at L/S = 10) are defined in VLAREMA.

Over the last decade, VITO (the Flemish Institute for Technological Research) as reference laboratory for the Public Waste Agency of Flanders (OVAM) has been organising round robin tests for the column leaching test of waste materials to acknowledge environmental laboratories of the Flemish region according to the legislation. Laboratories have to participate to the round robin test as a proficiency test, to monitor the quality of the laboratory results. Only certified labs are allowed to perform the column leaching test in the context of the regulatory framework.

Leachability depends on a number of physical parameters, including homogeneity, particle size, porosity, permeability of the solid phase influencing the flow rate and contact time between solution and solid, and temperature, as well as parameters of a chemical nature such as pH value, redox conditions, total organic carbon (TOC) content, chemical speciation of contaminants, complexation reactions and biological activity (Fytianos et al., 1998; Kalbe et al., 2008a; Quina et al., 2009; van der Sloot et al., 1996, 2006a,b). These factors are not fully studied yet by column leaching tests to gain a complete understanding of their effect on soil

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and waste material leachability. Furthermore, this abundance of factors complicates the reproducibility and repeatability of laboratory leaching tests and questions their analogy with *field conditions* (Beesley et al., 2010; Kalbe et al., 2007). Finally, a standardised test can only be valid on the precondition that reproducible test results can be achieved, not only within one laboratory, but also between different laboratories (Kalbe et al., 2008a). Kalbe et al. showed that the column leaching test has a good reproducibility, given that specifications for the procedure of the experiment are strictly followed by participating laboratories (Kalbe et al., 2008a, 2007).

This study presents a quantitative and statistical analysis of the reproducibility and repeatability of the column leaching test on 13 different mineral waste materials. In each of these materials the leachability of As, Cd, Cr, Cu, Hg, Pb, Ni and Zn was assessed. The obtained data shed light onto the influencing parameters in the column leaching test. They also provide a means to analyse the reproducibility and repeatability of the column leaching test for several soil and waste materials. In addition, the performance of laboratories was compared.

2. Materials

Prior to each round robin test, several mineral waste materials are carefully analysed before being selected as round robin test material. The material has to leach sufficient amounts of the elements that are selected to be validated during the round robin test. Furthermore, a series of three to five column leaching tests have to produce sufficiently coherent results before the material is considered adequate as round robin test material. The materials are dried and processed to a grain size smaller than 4 mm. Homogeneous sub-samples (2.5–3.5 kg) are gathered by a rotating sample splitter. No further pre-treatment by the labs was necessary. Over the years, several categories of mineral waste material have been used as round robin test material, ranging from dredged material, roofing material, construction and demolition waste to mineral waste produced in industrial processes. The materials are listed in Table 1, together with the number of laboratories that participated each year in the round robin test and the average cumulative emission of the elements. Total concentrations, grain size distributions, pH, conductivity data on the samples are provided as supporting information. It can be observed that the cumulative emission, i.e. the total amount of an element leached out up to L/S 10 l/kg varies strongly for each round robin test material. Also, the number of participating labs decreased over the years. This is mainly due to the fact that labs occasionally merge or cease their activity.

3. Methods

3.1. Organisation of the round robin program

The round robin data were gathered in the framework of the Flemish certification program to acknowledge labs to perform environmental tests such as the column leaching test. Yearly, a mineral waste material was selected by VITO for the column leaching round robin test, which was sent to the participating laboratories. Each participating laboratory received identical material to analyse. No further physical treatment, such as drying, milling or sieving, was necessary. The precise instructions for the set-up of the experiment are defined in the Flemish standard CMA/II/A.9.1 (Flemish Environmental Legislation, 2016). Each lab is instructed to perform several repetitions of the test: till 2009 each lab generated three replica measurements, from 2010 on this was reduced to two.

All the laboratories performed the analyses on the eluates independently. Since different analysis techniques are allowed in the standard (e.g. ICP-OES and ICP-MS), this introduces an additional variability. To be able to estimate the contribution of the analytical uncertainty, each of the laboratories also received two reference eluates which were created using leachates from pre-testing of the round robin materials, but were spiked with additional metals, one with a low and one with a high level of spiking.

Every year, all obtained column leaching test results were bundled and statistically analysed in order to assign a z-score to each laboratory for each leached elemental concentration. The bundled dataset were firstly analysed for outliers. Subsequently, a z-score was calculated for each measured value according to

$$z = \frac{x - x_c}{s}$$

Table 1
Materials of the column leaching round robin test over the years 2002–2014. Number of participating labs each year and average cumulative release (L/S = 10) per year and element in mg/kg d.m. The cell shading (white to dark grey) represents the cumulative release relative to each column (element) (from low to high). The Flemish legal limit values for each element are given in the bottom rows, together with the minimum reporting limits labs have to reach (Flemish Environmental Legislation, 2016).

		#Labs	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn
2002	Sediment	17	0.034		0.37	0.66			5.1	0.64
2003	Bottom Ash Sand	18				9.7		0.16		0.12
2004	Surface Blast Waste	16			0.36	0.41			0.28	0.68
2005	Waste Incinerator BA	17	0.011	0.005	0.063	10		0.14	0.00003	0.18
2006	Sediment (natural stream)	15	2.8	0.17	0.37	3.7	0.047	3.6	0.33	3.1
2007	Bituminous roofing material	15	0.11	0.010	0.072	0.16		0.15	0.087	2.5
2008	Contaminated soil	15	2.0	0.035	0.10	1.7	0.001	2.0	0.082	5.8
2009	Sediment (harbour)	11	868	18	94	108		208	36	271
2010	C&D aggregates (concrete)	12	0.011	0.003	0.11	0.067		0.031	0.69	0.13
2011	Inorganic filter cake	9	0.051	0.002	0.061	0.10		0.044	0.10	0.30
2012	Sediment	8	0.25	0.044	0.65	0.71	0.002	1.4	0.10	30
2013	Sieving Sand	10	0.089	0.007	1.0	1.2	0.0005	0.16	9.8	3.0
2014	Bottom Ash	9	0.068	0.002	0.047	0.82	0.0006	0.071	0.062	0.10
-	Limit Value (Flanders)	-	0.8	0.03	0.5	0.5	0.02	0.75	1.3	2.8
-	Reporting Limit	-	0.15	0.015	0.1	0.1	0.005	0.1	0.2	0.5