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Leaching behaviour of hazardous waste under the impact of different ambient conditions

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ABSTRACT

The overall objective of this study is to provide an improved basis for the assessment of the leaching behaviour of waste marked as hazardous partly stabilised (European waste catalogue code 19 03 04*). Four samples of hazardous partly stabilised waste were subjected to two leaching tests: up-flow column tests and batch equilibrium tests. The research was carried out in two directions: the first aims at comparing the results of the two experimental setups while the second aims at assessing the impact of different ambient conditions on the leaching behaviour of waste. Concerning this latter objective the effect of mesophilic temperature, mechanical constraints and acid environment were tested through column percolation tests. Results showed no significant differences between batch and column leaching test outcomes when comparing average concentrations calculated at a liquid to solid ratio of 10:1 $l\ kg^{-1}$ TS. Among the tested ambient conditions, the presence of an acid environment (pH = 4.5) accelerated the leaching process resulting in a higher cumulative released quantity measured on the majority of the investigated polluting substances. On the contrary, the effect of temperature and mechanical constraints seemed to not affect the process showing final contents even lower than values found for the standard test. This result was furthermore confirmed by the application of the principal component analysis.

1. Introduction

In the last decades, the hazardous waste management which includes collection, transportation, recycling, treatment and disposal processes has been a global major concern (Couto et al., 2013; Zhao et al., 2016). In Italy, in 2014, more than 1.2 million tons of hazardous waste were disposed in landfills while 0.9 million tons were exported in other European countries (ISPRA, 2016). In order to provide environmental protection, the disposal of hazardous waste in landfill is governed by National and European legislations (*European Council Decision 2003/33/EC of 19 December 2002*) that set strict criteria to be fulfilled. Hazardous waste can be disposed in underground storage or in landfills in accordance with the acceptance criteria or due to derogations of the limit values (e.g. inorganic elements, total organic carbon, dissolved organic carbon). In particular, hazardous waste can be disposed in non-hazardous landfills if stable and non-reactive. Considering the definition provided by European Council (2002), stable and non-reactive means that the leaching behaviour of waste do not change adversely in the long-term under landfill design conditions or foreseeable accidents as the impact of ambient conditions: e.g. temperature, mechanical constraints, etc.

Waste marked as Hazardous Partly Stabilised (WHPS, European waste catalogue code 19 03 04*) are solidified/stabilised waste that after the stabilisation process can release dangerous constituents which have not been changed completely into non-dangerous in the short, middle or long term (EPA, 2002). In Italy, the disposal of WHPS is becoming a major issue. Referring to the last recent data (ISPRA, 2016) around 671,000 tons are landfilled of which the 73% in non-hazardous landfills. Around 246,000 tons are instead exported to other European countries. This latter amount represents the 63% of the exported hazardous waste underling a critical issue concerning the management of this category of waste. The composition of WHPS is strongly heterogeneous including hazardous waste from waste processing facilities (e.g. fly and

Abbreviations: BE, batch eluates; CE, column eluates; DOC, dissolved organic carbon; L/S, liquid to solid ratio; LOD, limit of detection; PCA, principal component analysis; TOC, total organic carbon; TS, total solids; TVS, total volatile solids; WHPS, waste marked as hazardous partly stabilised.

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bottom ashes from Waste-to-Energy Plants) or wastewater treatment plants (e.g. industrial sludge). As a consequence, their chemical characteristics are also heterogeneous and their disposal scenarios concern several possibilities such as underground storage or even non-hazardous landfills in compliance with the acceptance criteria or their derogations. Nevertheless, in accordance with their characteristics, the elution process of this category of waste is not easy to predict and their acceptance in hazardous or non-hazardous landfills may be source of environmental risk due to the uncertainty of the release of polluting substances over time. As such, a deep understanding of the release of contaminants upon contact with water and under different landfill conditions (European Council, 2002) is of prime importance in order to investigate the stability of WHPS.

The release of soluble substances into water phase is the result of several phenomena that can occur simultaneously depending on leaching conditions, properties of the solid matrix and physic-chemical processes (Batchelor, 2006). Leaching tests are common tools for assessing constituent release upon contact with water (Lopez Meza et al., 2008) and according to Tiwari et al. (2015) can be divided into two general categories: static and dynamic extraction tests. In static extraction protocols (batch tests), leaching takes place with a single volume of leachant while in dynamic extraction protocols, the leaching fluid is renewed throughout the test. In particular, EN 12457, 2002 batch equilibrium test and CEN TS 14405, 2015, dynamic percolation test are acquired by the *European Council Decision of 19 December 2002* as criteria for acceptance of hazardous waste in landfill.

Batch equilibrium tests typically consists of contacting a sub-sample of material with a liquid phase in order to establish pseudo-equilibrium conditions. Once equilibrium is established, release is dependent on the geochemistry of the solid phase and on the chemistry of the liquid phase rather than on contact time (Garrabrants and Kosson, 2005).

Dynamic extraction tests provide information about the kinetics of contaminant mobilisation and results are presented as a function of time. Among dynamic extraction tests, flow-through tests (e.g. column tests) generate results that reflect real systems subject to fluid flow and solute transport and they are used to obtain information on short and long term leaching behaviour (Tiwari et al., 2015). Water is percolated through a column of material and collected as a function of liquid to solid ratio (L/S), which is used to represent leaching time. Flow-through leaching test methods can be used to simulate the leaching process of wastes disposed under particular landfill conditions. For example when waste is more permeable than its surrounding materials or when it has degraded under various environmental stresses to a state that ground water can flow-through the waste via the porosity system of the waste matrix (Poon and Chen, 1999). In this case, when the leachant flows through the waste, it carries away the mobile fraction of the contaminants. At the same time the immobile fraction is continuously solubilized to re-establish the equilibrium. The leachant flowing via the porosity system of the solid waste matrix carries away the mobile fraction and causes a contaminant concentration gradient which accelerates the leaching process. Because of its ability to accelerate the leaching process the flow-through leaching test methods can be used to study the long term leaching performance of waste (Poon and Chen, 1999). Analysing the leaching behaviour over time, different concentration-time patterns can come to light depending on dissolution/precipitation of minerals as well as sorption desorption processes. Under this perspective, Dijkstra et al. (2006a) studied the leaching behaviour of municipal solid waste incinerator bottom ash identifying five patterns: (1) rapid dissolution phase until exhaustion; (2) dissolution until equilibrium; (3) dissolution of a phase that does not reach equilibrium during the experiment; (4) rapid dissolution followed by a slower dissolution of a less soluble phase; (5) concentration decrease.

When comparing batch and column tests, batch experiments offer the advantage of easier design, while column testing provides an optimum approximation to leaching processes that occur under field conditions without compromising reproducibility of experiments (Butera et al., 2015; Delay et al., 2007; Dijkstra et al., 2006b). Column tests are more suitable for prediction purposes, but they are often time-consuming, reaching duration of several weeks. Alternatively, batch tests can be carried out in shorter periods of time, varying from several hours to few days. In the light of the practical advantages and disadvantages, it is important to understand the similarities and differences between constituent leaching under batch and column tests with the aim to provide effective tools for environmental decision-making (Lopez Meza et al., 2008).

Several researches have been performed on various waste categories aim to study the leaching behaviour by means of batch and column tests: stabilised/solidified waste (Barna et al., 1997; De Windt et al., 2007; Liu et al., 2013; Malviya and Chaudhary, 2006; Poon et al., 2001), mining waste (Al-Abed et al., 2008; Turner et al., 2009), construction and demolition waste (Butera et al., 2014, 2015; Delay et al., 2007; Lopez Meza et al., 2008; Nielsen et al., 2006; Roussat et al., 2008), contaminated soils (Cruz Payán et al., 2012; Gardner et al., 2007; Hartley et al., 2004), fly-ash stabilised soils (Bin-Shafique et al., 2006), soils used in construction works (Quaghebeur et al., 2006). To our knowledge, the leaching behaviour of WHPS and its alteration to foreseeable landfill conditions has not yet been studied.

The overall objective of this study is therefore to provide an improved basis for the assessment of constituent release from WHPS. Four WHPS samples obtained from different waste treatment facilities in Tuscany (Italy) were subjected to both batch equilibrium and up-flow column tests and evaluated in relation to: differences between column and leaching test in the release of polluting substances and the impact of different ambient conditions on the leaching behaviour (temperature, mechanical constraints and acid environment).

2. Materials and methods

2.1. Materials

Four WHPS samples (W1, W2, W3 and W4 – European Waste Code 19 03 04*) were obtained from four different treatment facilities in Tuscany (Italy). WHPS samples were selected based on easy procurement of the waste and according to chemical analysis with the intent to study a significant range of cases. According to the description of the production process provided by the facility operators, W1-W4 were obtained after a solidification/stabilization treatment of hazardous and non-hazardous waste. In particular W1-W4 were composed by: fly ashes containing dangerous substances (19 01 13*), bottom ashes and slags containing dangerous substances (19 01 11*), filter cakes from gas treatment (19 01 05*), soil and stones containing dangerous substances (17 05 03*), solid wastes from gas treatment containing dangerous substances (10 02 07*), sludge from treatment of urban waste water (19 08 05), sludge from biological treatment of industrial waste (19 08 12) and sludge containing dangerous substances from biological treatment of industrial waste water (19 08 11*). The hydraulic binders used for the treatment were lime and Portland cement. The sampling was done in compliance with the standard procedure EN 932-1 (1996): sample increments were collected from different positions in the stockpiles by means of a shovel and combined into primary samples (approximately 20 kg) which were transported to