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An ecotoxicological evaluation of aged bottom ash for use in constructions

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ABSTRACT

Municipal and Industrial Solid Waste Incineration (MISWI) bottom ash is mainly deposited in landfills, but natural resources and energy could be saved if these ash materials would be used in geotechnical constructions. To enable such usage, knowledge is needed on their potential environmental impact. The aim of this study was to evaluate the ecotoxicity of leachates from MISWI bottom ash, aged for five years, in an environmental relevant way using a sequential batch leaching method at the Liquid/Solid-ratio interval 1–3, and to test the leachates in a (sub)chronic ecotoxicity test. Also, the leachates were characterized chemically and with the technique of diffusive gradients in thin films (DGTs). By comparing established ecotoxicity data for each element with chemically analysed and labile concentrations in the leachates, potentially problematic elements were identified by calculating Hazard Quotients (HQ). Overall, our results show that the ecotoxicity was in general low and decreased with increased leaching. A strong correspondence between calculated HQs and observed toxicity over the full L/S range was observed for K. However, K will likely not be problematic from a long-term environmental perspective when using the ash, since it is a naturally occurring essential macro element which is not classified as ecotoxic in the chemical legislation. Although Cu was measured in total concentrations close to where a toxic response is expected, even at L/S 3, the DGT-analysis showed that less than 50% was present in a labile fraction, indicating that Cu is complexed by organic ligands which reduce its bioavailability.

1. Introduction

A question of rising concern is strategies for sustainable ash management when incinerated ash residues are increasing in the European Union (Blumenthal, 2011). At present, Municipal and Industrial Solid Waste Incineration (MISWI) bottom ash is mainly deposited in landfills, but natural resources and energy could be saved if these ash materials would be used in geotechnical constructions, e.g. in embankments and roads (e.g. Chandler et al., 1997; Toller et al., 2009). Such usage is already common in some countries (e.g. The Netherlands, Van Gerven et al., 2005) and emerging in others (e.g. Sweden). However, there are potential conflicts to the goal of reducing toxic pollution and recycling of ash materials, triggered by energy and material efficiency targets, and before use in constructions it is necessary to carefully assess the risks associated with the contaminants present in the ash.

Ash consists of a complex mixture of hazardous and non-hazardous elements, mainly metal elements and other inorganic substances (Donatello et al., 2010). In the simplest way, ecotoxicological properties of ash may be predicted based on chemical analysis of known hazardous elements in the solid material, which then are compared to literature toxicity data for the same elements (Stiernström et al., 2011). However, such an approach may be questionable since aspects of e.g. bioavailability, leaching capacity, speciation and mixture toxicity is not included. For instance, depending on parameters such as pH, level and quality of dissolved organic material and particle size, elements will prevail as different species with varying leaching capacity, bioavailability and toxicity (van der Sloot et al., 2009; Postman et al., 2009). Many contaminants may also be strongly sequestered by the solid matrix, and are hardly released to other compartments (Escher and Hermens, 2004; Enell et al., 2008). As a consequence, there is often a poor relationship between the theoretical ecotoxicity of analysed individual elements and their contribution to the measured total toxicity (Bihari et al., 2005; Sundberg et al., 2005), which may lead to either over- or underestimations of risk potential, and ultimately incorrect risk management decisions (Stiernström et al., 2013). To increase the relevance of hazard and risk assessment related to ash and other waste streams, it is becoming increasingly common to study the ecotoxicity of the solid material or leachates generated from the solid material using bioassays (e.g. Ferrari et al., 1999; Lapa et al., 2002; Pandard et al., 2006; Wilke et al., 2008; Römbke et al., 2009; Moser and Römbke, 2009; Stiernström et al., 2011).

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These approaches certainly have the potential to improve both hazard classification and risk assessment of e.g. ash materials, however, there are still problematic aspects that need to be considered. Of relevance for this study, essential macro elements (e. g. K and Ca ions) may be present in such high concentrations in ash leachates that they are the major cause to the observed effects in the ecotoxicity tests (Stiernström et al., 2011). These macro elements may also have an antagonistic effect on the toxicity of heavy metals released from ash (Di Toro et al., 2001). Thus, for both hazard classification and risk assessment of ash materials, it is crucial to understand the mechanisms governing toxicity of the studied material, i.e. speciation, leaching capacity, mobility, bioavailability and synergistic/(ant)agonistic effects. The overall aim of this study was to evaluate the long-term ecotoxicity of leachates from aged MISWI bottom ash, for potential utilization as filling material or for construction purposes. Aging, as a treatment form, has the potential to reduce the inherent hazard property of bottom ash (Gori et al., 2013; Stiernström et al., 2011). Based on the hypothesis that the toxicity of the leachates will decrease with increased leaching, a sequential batch leaching method for simulation of increased leaching was performed at a Liquid/Solid (L/S)-ratio interval of 1–3 L/kg. This L/S-ratio is relevant for risk assessment of recycling of mineral materials in geotechnical constructions (Susset and Grathwohl, 2011). In a first *data generation step*, the produced leachates were (i) tested with a (sub)chronic ecotoxicity test (a partial life cycle test investigating Larval Development Ratio, LDR) with the crustacean *Nitocra spinipes*, (ii) characterized chemically using conventional element analyses, focusing on key elements identified as problematic in previous studies of ash materials (Stiernström et al., 2011, in press) and (iii) analysed with the technique of diffusive gradients in thin films (DGTs) to obtain the labile fractions of selected elements. By using this technique it is possible to assess the amount of labile cations, defined as the sum of free ions and weakly bound complexes able to migrate into a resin layer (Zhang and Davison, 1995). The technique is analogous to migration across a cell membrane and thus DGT-labile fractions are assumed to represent the bioavailable fraction more closely than fractions assessed by conventional analysis of filtrated or unfiltrated leachates. This information was considered crucial for interpreting the results of ecotoxicity tests according to the results from other studies on ash materials (Stiernström et al., 2011, 2013), since metals associated with colloidal or particle matter are expected to be markedly less bioavailable to the test organisms. In a second *evaluation step*, potentially problematic elements were identified by calculating Hazard Quotients (HQ), taking into consideration the toxicity and concentration of each element.

2. Materials and methods

2.1. Experimental design

The rationale for starting the testing at L/S 1 was due to practical requirements; it was not possible to generate sufficient leachate quantities for both chemical characterization and ecotoxicity tests at lower L/S-ratios. Column leaching tests, which provide a flow-through pattern similar to that found in field conditions, are the common method to apply in order to obtain leaching characteristics for accumulated L/S-ratio. However, the experimental design of the column test is not suitable for subsequent ecotoxicity testing (due to e.g. long duration time and risk of degradation of organic matter). Hence, we used a batch test designed for characterization of waste (EN 12457-3) and modified it to include three subsequent steps of leaching at cumulative L/S ratios of 1, 2 and 3 L/kg (see Section 2.3 below).

The DGT technique used in this study was originally developed for the measurement of trace elements in natural water (Davison and Zhang, 1994; Zhang and Davison, 1995; Denney et al., 1999) but has been further developed to be applicable also for studying macro elements, e.g. Ca and Mg (Dahlqvist et al., 2002). Today, it has become a common *in situ* sampling technique for measuring labile concentrations in natural waters, soil and sediments (e.g. Balistrieri and Blank, 2008; Oporto et al., 2009; Sherwood et al., 2009). It has also been extensively tested in brackish waters from the Baltic sea, used as dilution media here, in the (sub)chronic tests (Österlund, 2010; Forsberg, 2005). However, it has, to our knowledge, not previously been used to determine labile concentrations in leachates from ash materials. Compared to natural waters, ash leachates contain highly elevated levels of some elements. In addition, the available test volume received from a leaching test is limited compared to sampling natural waters, and thus depletion of elements at low concentrations may occur. Thus, special care was here taken to calculate appropriate deployment times of the DGT experiments (see Section 2.3.3 below).

Although it is preferred to apply ecotoxicological test methods with species representatives from different trophic levels in hazard classification and risk assessment of complex pollution matrices (e.g. Stiernström et al., 2011), in this study we chose to focus on performing repeated studies using a (sub)chronic ecotoxicity test with the species that has been the most robust and sensitive in our previous studies on ash materials, i.e. the harpacticoid copepod *N. spinipes* (Stiernström et al., 2011, 2013, see Section 2.3.4 below).

2.2. Selection and preparation of material

The selected material was a bottom ash with origin from a modern incineration plant in Sweden, incinerating mainly municipal solid waste and additionally fractionated organic industrial waste on occasions. (Tekniska Verken, Linköping). The ash was aged for five years during outdoors storing in an open pile until the sampling occasion. Due to the aging of the material, it produced leachates of acceptable pH levels for the test organisms and thus found suitable for the study (no need to adjust pH before ecotoxicity testing). The material was stored in sealed containers in cold storage (4 ± 2 °C) until the time of the study.

A chemical characterisation showed an element composition and a content of potentially toxic metals at levels that can be considered representative for modern Swedish MISWI-bottom ash materials (data shown in Table 1S in the Supplementary information). The elemental composition of the material was investigated using microwave assisted digestion with concentrated nitric acid (HNO_3) and Inductively Coupled Plasma Atom Emission Spectrometry (ICP-AES) for determination of Ca, Cu, K, Mg, P, S and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for Zn, As, Cd, Cr, Mo, Ni, Pb, and Sb.

In order to prepare representative test portions, the material was homogenised and divided into subsamples according to EN 15002:2006 applying coning and quartering to split the sample. Instead of crushing the oversized material particles >2 mm, they were sorted out by sieving the samples (without prior drying) following ISO 11464. This sample preparation is a deviation from EN 12457-3 but is assumed by the authors to provide test samples that better resembles actual conditions of the material when used in construction application.

2.3. Data generation

2.3.1. Leaching test

The leaching followed the procedure of EN 12457-3 (two stage batch test), but was modified to include three subsequent steps of leaching at cumulative L/S ratios of 1, 2 and 3 L/kg. In each leaching