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Leaching of chloride, sulphate, heavy metals, dissolved organic carbon and phenolic organic pesticides from contaminated concrete

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ABSTRACT

Concrete samples from demolition waste of a former pesticide plant in Sweden were analysed for total contents and leachate concentrations of potentially hazardous inorganic substances, TOC, phenols, as well as for pesticide compounds such as phenoxy acids, chlorophenols and chlorocresols. Leachates were produced by means of modified standard column leaching tests and pH-stat batch tests. Due to elevated contents of chromium and lead, as well as due to high chloride concentrations in the first leachate from column tests at L/S 0.1, recycling of the concrete as a construction material in groundworks is likely to be restricted according to Swedish guidelines. The studied pesticide compounds appear to be relatively mobile at the materials own pH > 12, 12, 9 and 7. Potential leaching of pesticide residues from recycled concrete to ground water and surface water might exceed water quality guidelines for the remediation site and the EU Water Framework Directive. Results of this study stress the necessity to systematically study the mechanism behind mobility of organic contaminants from alkaline construction and demolition wastes rather than rely on total content limit values.

1. Introduction

Construction and Demolition (C&D) waste makes up for a significant part of solid waste streams. The total production of C&D waste in, for example, Europe was estimated to 970 Mtonnes in 2006 (EC, 2011, for all countries reporting to Eurostat). 6–10 million tons of C&D wastes are produced annually in Sweden (SMED, 2009), which has a population of 9.6 million. A part of this C&D waste is made up by residues from remediation of contaminated sites: In Sweden alone there are at least 80,000 known or suspected contaminated areas (Swedish EPA, 2013). Often, industrial sites comprise of both contaminated soil and buildings or constructions affected by industrial activities, spill, accidents etc.

Data on the amounts or properties of contaminated C&D waste in Sweden are, however, scarce, except for a few publicly financed remediation projects with buildings or C&D wastes contaminated with petroleum products (gasoline, oils), plasticizers (phthalates) and PAH from tar (Svensson, 2009; Landström and Östlund, 2011; Niklasson and Söderström, 2005). Comprehensive national data on properties and the extent of contaminated buildings or C&D wastes in general are only gathered for polychlorinated biphenyls (PCBs, a group of prioritized persistent organic pollutants) in concentrations above 500 ppm (parts per million). These have been screened or remediated in recent years (Swedish EPA, 2002; Lilliehorn and Bernevi-Rex, 2010).

Reclaimed, crushed concrete and bricks have a large potential for recycling (Behera et al., 2014; Khalaf and DeVenny, 2004). The coarse aggregates from recycled C&D wastes exhibit properties which make them potentially suitable for road surface, base or sub-base applications, as well as for precast concrete production or simple backfilling (Herrador et al., 2012; Arulrajah et al., 2013; Leite et al., 2011; Soutsos et al., 2012). To a certain extent, aggregates from reclaimed concrete can also be recycled in the production of new concrete, although certain national standard quality requirements might be difficult to achieve (Rao et al., 2007; Rodriguez-Robles et al., 2014). Nevertheless, source separated and recovered C&D wastes such as aggregates from reclaimed concrete might qualify for being considered as a product or secondary raw material, and, in the future, cease to be classified as waste (Villanueva et al., 2010; Delgado et al., 2009). Comprehensive studies on the mobility of organic substances from concrete C&D wastes are, with the exception of PAH and concrete additives, however, scarce (Butera et al., 2014, 2015; Roskam and Comans, 2009; Brantley and Townsend, 1999; Krueger et al., 2012; Togerö, 2006).

One of the most prominent cases of soil contamination and remediation in Sweden is the remediation project *BT Kemi*

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efterbehandling (Englöv and Vanek, 2010; van Praagh et al., 2014a, b). During the 1960s and 70s, *BT Kemi* (short for Bönnelyche & Thuröe) was one of few corporations producing pesticides in Sweden. At a former sugar syrup factory in Teckomatorp, a small village in Southwestern Sweden, *BT Kemi* produced phenoxy acids, chlorophenols and chlorocresols, as well as dinoseb. Several buildings and subground constructions of the former sugar syrup factory were kept and used for pesticide production and disposal. Soil, ground water and surface water, as well as buildings and constructions, were or still are polluted with residues from former pesticide production.

The site has been target of several remediation projects since production seized. The amount of contaminated concrete on site is estimated to approximately 5000 m^3, part of which has been dug up, cut and stored on site. Until lately, contaminated construction materials occurring at remediation projects in Sweden have been regarded as though they were contaminated soil: Guidelines and generic or site specific environmental limit values according to the Swedish Environmental Protection Agency (Swedish EPA, 2009) have been applied, despite the obvious differences in physical-chemical properties of soils and, for example, concrete or bricks. Alternatively, the Swedish ordinance on waste (“Avfalls förordningen”, SFS 2011:927) or guidelines issued by the Swedish Waste Association on contaminated soils and aggregates (Swedish Waste Association, 2007) have been used to assess whether soil or construction and demolition wastes have to be regarded as hazardous wastes, and, thus, should not be readily recycled or reused on site unless treated.

The Swedish EPA has published guidelines for the recycling of granular waste materials in groundwork constructions in non-bound form (Swedish EPA, 2010, see Saveyn et al., 2014, for a summary in English). Two different types of recycling are defined by the guidelines and subject to recommended “maximum contamination levels”: (1) “Free” use without regulatory restrictions outside environmentally sensitive areas corresponding to a risk level considered “negligible”, and (2), use for landfill capping above liner. These recommended maximum levels include certain heavy metals, both total contents and leached amounts/concentrations at L/S 10/0.1, as well as total contents of polyaromatic hydrocarbons (PAH). The maximum levels are consistently lower than acceptance criteria for inert waste to landfill in the EU, as of council decision 2003/33/EC. Despite the limitations of using total concentrations for evaluating potential environmental risks to, for example, ground water resources (see for example Roskam and Comans, 2009), the Swedish guidelines rely on total content limits for PAH. How risks from other organic contaminants, such as found at the *BT Kemi* site, or their potential leaching have to be considered is addressed only in a principle way in these guidelines. Recycling the contaminated concrete on site rather than disposing it of at an off-site landfill would limit transportation considerably and safe landfill resources. The question is, however, whether recycling the concrete as a construction material on-site or near the site would lead to unacceptable risks for ground and surface water.

As part of the ongoing remediation project at *BT Kemi*, concrete samples have been the target of previous investigations. The scope of these was as follows:

- Investigate whether total contents of inorganic compounds might restrict recycling of crushed, contaminated concrete at the *BT Kemi* site in accordance with national Swedish guidelines.
- Study the potential mobility of contaminants by means of leaching tests and pH-stat tests.
- Determine whether recycling of crushed concrete on site states a potential risk to ground- or surface water resources.

Preliminary results with regard to those three parts are presented in this article.

2. Methods and material

2.1. Remediation site and solid samples

For this study, subsamples were taken from pre-sorted stockpiles from demolition campaigns. These originated from underground basins. These basins measure ca 300 by 8 m, constructed from steel reinforced concrete with a wall thickness of 15–25 cm. They were originally intended for washing sugar beets prior to entering the sugar syrup factory. Allegedly, they were subsequently used for pesticide storage during the 1960s and 70s. Four subsamples from different locations (D4, D5, D6, and D13) in the northern area of the remediation site were analysed. The grab samples were treated as if they belonged to the same population (single construction). For the naked eye visible parts of the steel reinforcement were removed from the concrete stockpiles after cutting.

Grab samples were put in airtight bags, sealed and stored dry and dark at a maximum of 8–15°C. Subsamples were packed dry and dark prior to transport to analytical laboratories. For total content analyses, samples were both crushed and grinded.

2.2. Leaching tests and analytical techniques

Tables 1a and 1b contain information on employed tests and analytical techniques.

Subsamples were crushed and mixed proportionally to a collective sample for the leaching tests (maximum particle size <4 and <10 mm for the batch and column tests, respectively). The leachat was supplied from a PEHD (low density polypropylene) tank through Tygon-tubing (*Saint-Gobain*). Leaching tests were carried out at a commercial laboratory (*Eurofins Environment*). Leachate samples were left for sediment to settle and subsequently decanted. Leachate samples were neither filtrated nor conserved by additives. Analyses were carried out as soon as possible by a commercial laboratory (*ALS Scandinavia or Eurofins Environment*). Samples were transported dark and at a maximum temperature of 4°C. The leachates were collected in a PEHD (polyethylene high density) container.

Leaching tests employed in this study deviated from standard leaching tests (see Table 1a). The reasons for these deviations were twofold: Time- and budget constraints (fewer pH and L/S sampled in the pH-stat and column tests, respectively) and lack of standardized procedures available at commercial labs in Sweden at the time (to determine leaching of organic substances). The implications of these deviations are discussed in the results and discussion section below.

All leaching tests were carried out in duplicates. Subsamples from two different sampling locations (D4 and D6) were used for pH-stat testing.

3. Results and discussion

3.1. Total contents

3.1.1. Inorganic parameters

Table 2 summarizes results from the analyses of total inorganic content in concrete samples.

Results in Table 2 indicate the following.

- Metal contents deviate from sample to sample, especially Ba, Cr, Fe, Ni and Pb.