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Effects of colloidal particles on the results and reproducibility of batch leaching tests for heavy metal-contaminated soil

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Abstract

A simple batch test leaching procedure is used in many countries, as a compliance test, to evaluate the leaching of mainly inorganic substances from soil. However, agitating certain types of soil and then passing the solution through a membrane filter with 0.45-µm pores yields filtrates that have been colored by the colloidal particles. These colloids might affect the results of the inorganic substances obtained in the batch tests. In this study, we evaluated the effects of colloidal particles on the results and the reproducibility of batch tests on contaminated soil using different agitation methods and membrane filters with different pore sizes. The leaching behaviors of As, Pb, Se, F and Cl from three types of soil were studied. The As and Pb concentrations in the leachates of some types of soil were clearly affected by the amount of colloidal particles with a diameter of 0.10–0.45 µm and by the agitation method used. This was probably because As and Pb were present mainly in the particulate form in the leachate that had been passed through a membrane filter with 0.45-µm pores. This is not the case for every type of soil. The results of batch leaching tests showed that not only dissolved but also colloidal forms with a diameter of 0.10–0.45 µm might be included and that the existence of colloidal particles in the leachate decreases the batch leaching test reproducibility.

1. Introduction

To evaluate the risk of contaminated soil for groundwater contamination and human health, leaching tests are commonly conducted in many countries. Several leaching test methods have been developed including batch tests (International Standardization Organization, 2007a,b; German Standardization Organization, Deutsches Institut für Normung, 2015; European Committee for Standardisation, CEN Comité Européen de Normalisation, 2002; United States Environmental Protection Agency, 2013b), up-flow column percolation tests (United States Environmental Protection Agency, 2013a; International Standardization Organization, 2007c; Naka et al., 2016) and sequential extraction methods (Arain et al., 2008; Kazi et al., 2005).

Batch tests are used as compliance tests for contaminated soil and waste in many countries, e.g., Japan (Ministry of Environment, 1991), Germany (German Standardization Organization, Deutsches Institut für Normung, 2015) and the United States (United States Environmental Protection Agency, 2013b), and in international standards (International Standardization Organization, 2007a,b) because such tests are simpler than

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other leaching tests, such as up-flow column percolation tests, and they can be performed quickly. Therefore, batch test results need to be reproducible and reliable.

The basic procedure for a batch test performed on inorganic substances is quite simple. Firstly, a solid sample is placed in a suitable bottle and a leachant is added at a specified liquid-to-solid ratio. Secondly, the mixture is agitated for a specified period. Next, the solid material is allowed to settle and the mixture is centrifuged. Then, the liquid is decanted and passed through a membrane filter (MF) to obtain the leachate solution. Finally, the contaminants in the leachate are determined. Table 1 shows batch test procedures used by ISO that are widely applied in Europe, Germany, the United States and Japan. The basic test procedures are similar, but there are some differences, e.g., the type of leachant (demineralized water or 1 mmol $CaCl_2$ solution), the agitation method (rotation, roller table, horizontal or vertical shaking), the agitation intensity (5–200 rpm), the agitation time (6–72 h), the centrifuge rotation speed and centrifuge time (2000–3000*g* for 20 min for inorganic contaminants and 20,000–30,000*g* for 30 min for organic contaminants) and the filter pore size and material (membrane filter with 0.45 μm pores for inorganics and glass fiber filter with ≤0.7-μm pores for organics).

There are some important requirements for successful batch test methods. The first requirement is equilibrium; allowing equilibrium or semi-equilibrium conditions to be reached is the most important way of obtaining comparable batch leaching test results (International Standardization Organization, 2007a,b). Every test requires the assumption that equilibrium or semi-equilibrium is reached at the specified contact time. However, the equilibrium status that is reached will depend on the soil type and the species being investigated. In the ISO TS21268-1 procedure, it is stated that contact for 24 h is sufficient for equilibrium to be reached and that 6 h may be sufficient in the following specific cases when "it can be demonstrated that equilibrium or semi-equilibrium is reached or, for quality control purposes, a quick turn-around time is required" (International Standardization Organization, 2007a).

The second requirement is the removal of suspended matter and colloidal particles. It is well known that organic contaminants, such as polycyclic aromatic hydrocarbons (PAHs) and PCBs, are strongly adsorbed into colloidal particles, such as organic matter and clay particles (Jacobsen et al., 1998; Prechtel et al., 2002; Massoudieh and Ginn, 2007). To avoid or reduce the effect of organic contaminants adsorbed into colloidal particles, the removal of the colloidal particles is important. In the procedures used in Germany and the ISO TS 21268-2 methods, the supernatant after agitation was centrifuged at 20,000–30,000*g* for 30 min to remove the colloidal particles without 0.45-μm MF filtration for organic substances. In the German standard, it is specified that the turbidity of the leachate must remain below 20 formazin nephelometric units (International Standardization Organization, 2007a, German Standardization Organization, Deutsches Institut für Normung, 2015).

Otherwise, little attention is generally paid to the colloidal particles for batch tests for inorganic substances, because the leachate is usually passed through a 0.45-μm MF (see Table 1). However, indirect evidence of the presence of colloids in leachates passed through 0.45-μm MFs and a correlation between colloid concentrations and the concentrations of some inorganic substances has been found in a small number of studies. Bergendahl and Grasso (1998) measured the amounts of particles with diameters less than 1 μm in leachates produced using different agitation times and found that the concentrations of colloids with diameters of 0.72 and 0.83 μm and the total surface area of colloids in the leachate increased as the agitation time increased. Dalgren et al. (2011) found higher Cu, Pb and Zn concentrations in unfiltered column leachate samples than in remediated soil samples that had been passed through a 0.45-μm MF.

The above-mentioned studies indicate that the results of batch leaching tests for inorganic substances, particularly inorganic substances that tend to be adsorbed by soil and colloidal particles with high distribution coefficients (such as Pb) by the surface charge of soil and colloidal particles, might be affected by colloidal particles with diameters less than 0.45 μm. Bergendahl and Grasso (1998) did not investigate the influence of agitation on the relationship between the amount of colloids in the leachate and the heavy metal concentrations, but they did find colloids in leachates that had been passed through 0.45-μm MFs under certain conditions.

Batch tests are performed to determine the concentrations of substances of interest that are dissolved from contaminated soil. The results of the above studies indicate that the measured concentrations of inorganic substances by batch tests might include not only dissolved inorganic substances, but also colloid-adsorbed forms of these substances if the filtered leachates include colloidal particles. Furthermore, if the total amount of colloids in a filtered leachate increases as the agitation time is increased, as described by Bergendahl and Grasso (1998), the agitation conditions might affect the reproducibility of the results of batch tests on inorganic substances.

This study was performed with the aim of evaluating the effects of colloidal particles on the results of batch leaching tests on soil contaminated with inorganic substances when different agitation methods and MFs with different pore sizes were applied. Five inorganic substances were evaluated; they were classified into three groups, namely, (1) Pb, which has high adsorption potential because of its positive charge, (2) Se, Cl and Se, which have low adsorption potential because of their negative charge and (3) As, where the adsorption potential changes according to the soil-water conditions and the form of As used. Five agitation methods and two types of MF (with 0.45- and 0.10-μm pores) were tested using three types of soil. The concentrations of five inorganic substances and the turbidity

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