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# As transport characterization from the soil to the groundwater: an original laboratory study

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## Abstract

Heavy metals are major soil pollutants since a lot of former industrial soils are polluted by these contaminants. In the context of risk assessment of contaminated sites, they are of particular concern because of their toxicity toward human beings. Nevertheless, the vadose zone of the soil is not taken into consideration in this kind of study, although this is where the pollution enters the soil. That is why mechanisms responsible for trace element release in the unsaturated zone of the soil have to be understood. In addition, tools and methods to measure and put forward interactions between contaminants and the solid matrix have to be provided. Therefore, we selected a soil from a wood preserving facility site (contaminated by As) and decided to follow As release as it was the only trace element measured on site at a concentration in groundwater above the EU guidelines for drinking water quality. We designed an original laboratory set up: two columns were filled with 2 sub-samples of the As contaminated soil (202 and 253 mg/kg) and As release was studied as it could occur on site. Two main phenomena were simulated during these experiments: rain water infiltration and an increase in the water table level. After the results of the two experiments, it was shown that As concentrations at the outlet of the two columns were constant over time. Given that and the results of the sequential extraction carried out on this soil, As release could mainly occur from the soluble and exchangeable parts. This will be modeled in a future work in order to confirm this hypothesis.

## Introduction

In industrialized countries, a lot of brownfields polluted with trace metals have been identified. In France, site management strategies are based on a site specific approach. This corresponds to a combined approach between field investigations and modeling to understand and predict pollutant migration through the soil. In these studies the chemistry and hydrology of the vadose zone of the soil are not considered although the pollutants are entering the soil from the unsaturated zone. That is why it seems necessary to have a good knowledge of the mechanisms responsible for heavy metal retention and transfer from the vadose zone of the soil to the water table, in order to put forward potential risks for groundwater. In the case of trace metals, mechanisms controlling their mobility in soils are relatively well known but the predominance of one over another is still unclear. Most of the time, pollutant transport is evaluated at the laboratory scale with batch or column experiments. Nevertheless, these systems are of small dimensions, the soil used is generally sieved, and these studies are mainly carried out under saturated flow conditions (Michel, 2009). We therefore may wonder whether these conditions are representative of what occurs in the field. That is why experiments at a pilot scale are necessary to assess heavy metal transport at the field scale. But these kinds of experiments are rather time consuming and quite expensive, and as a consequence cannot be used when doing risk assessment studies.

Therefore, the aim of this work was to design an original laboratory experiment able to represent trace metal transport as it occurred on site. This study focused on As release as it was the trace element released on site at concentrations above the EU guidelines for drinking water quality. This will help

us in putting forward mechanisms responsible for As migration in this soil.

## Materials and methods

### *Polluted soil sample*

This work was carried out in the frame of the CAPHEINE project (Characterization of trace element transfer in the vadose zone of the soil), funded by ADEME (French Environment and Energy Management Agency).

The polluted soil was sampled on an in-service wood preserving facility site, located in the northeast of France. The wood preserving activity for phone and power line poles started in 1929. Different preserving techniques were used so far, meaning that different kinds of chemicals were applied and may have contaminated soil and groundwater. The main chemicals used were chromated copper arsenate compounds (CCA) and creosote distillates. Soil samples were taken between 0 and 6 m depth, where the main pollution source was supposed to be localized. The surface of this area was 100 m<sup>2</sup> (8 m x 12 m) and soil was sampled with a helical auger. The soil was analyzed with a portable X-ray fluorescence analyzer. Arsenic was detected between 2.5 and 3 m depth, that is why 21 kg of soils were collected at this depth with a helical auger, placed in plastic bags and stored in a cooler before being analyzed in the laboratory. These samples were homogenized by quartering and separated in 2 sub-samples (S1 and S2). A small quantity of each sub-sample was dried, sieved, grinded and submitted to a sequential and a total extraction. As distribution in these soils was assessed using a slightly modified sequential extraction according to the Tessier protocol (Tessier et al., 1979). This modified protocol was published by Denys (Denys et al., 2008). These chemical

extractions were done on three replicates. Total concentrations for As were 202 and 253 mg/kg for S1 and S2, respectively. As was found in soluble and exchangeable form (about 8 %) and mainly bound to manganese and iron oxides (more than 50 %). In addition, soil physico-chemical parameters such as grain size distribution, pH, TOC, CEC, and total trace element content were determined on each fraction.

#### Experimental set up

Two laboratory columns were used in this study. They were all the same size and were made of transparent PVC and composed of 3 parts (Figure 1). The middle part was filled with 5.4 kg of As polluted soil. This part was 30 cm high and 12 cm in diameter. The lower part of the set up was composed of a funnel.

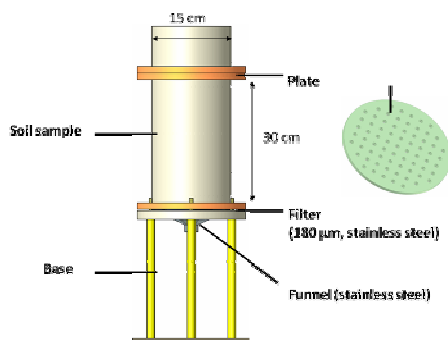


Figure 1. Laboratory column.

#### Experimental strategy

Two kinds of experiments were conducted with two columns. The first one was an infiltration column (C1) and was filled with the sub-sample S1. Cycles of rain water infiltration and drying periods were simulated: DI water was applied at the top of the column thanks to a peristaltic pump, and samples were collected at the bottom. Note that between the upper and middle parts, a plate was sealed; it was composed of holes, and pipes were set in these holes, so that the soil in the middle part was homogeneously fed. The water fluxes were controlled and adjusted to the fluxes recorded on site. The influence of the flowrate (between 0.04 and 0.25 mL/min), the feed duration (between 2 and 10 days), and the drying period (between 6 and 90 days) was studied. As the flowrate was very low, the column ran under unsaturated conditions.

The second column (C2) was filled with sub-sample S2 and saturated upward with calcium nitrate ( $2.5 \cdot 10^{-3}$  M) thanks to a peristaltic pump. The flowrate varied between 0.02 and 0.1 mL/min and was adjusted to the water table level increase speed measured on site. When the soil column was fully saturated, the flow was stopped, the pump was disconnected and water was collected at the bottom of the column. Several cycles were applied to the column and the flowrate influence on As release was studied.

pH, conductivity, redox potential, TOC and As content were measured at the outlet of both columns.

#### Results and discussion

At the outlet of C1, As concentrations were constant over time and were mainly around  $3\,000 \mu\text{g/L}$ . No influence of the flowrate, the feed duration and the drying period on As release was noticed. The same trend was observed for C2: As concentrations at the outlet of the column did not show any particular evolution over time and were mainly around  $2\,000 \mu\text{g/L}$ , with no influence of the flowrate. These results indicated that As release in this soil was not rate limited. Therefore, we could hypothesize that it resulted from a thermodynamic equilibrium and that As could mainly come from the soluble and exchangeable parts.

#### Conclusions

In this work, As release was studied at the laboratory scale. An original laboratory column helps us to identify phenomenon responsible for As release in the unsaturated zone of the soil. In a future work As release in these soil columns will be modelled in order to confirm the hypothesis of As coming from soluble and exchangeable parts. This study paves the way for a better understanding of trace element behavior in the vadose zone of the soil during risk assessment studies and gives a new approach to study this part of the soil.

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