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# As transport characterization in the vadose zone of the soil: a combined study between field and laboratory experiments

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## 1. Introduction

In industrialized countries, a lot of brownfields polluted with trace metals are identified. In France, site management strategies are based on a site specific approach. This corresponds to a combined approach between field investigations and modeling. In these studies the chemistry and hydrology of the vadose zone of the soil are not considered though 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 in soils from the vadose zone to the water table, in order to put forward potential risks for groundwater. 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. 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 characterize As transport in soil in the field and to put forward mechanisms responsible for its migration in the vadose zone of the soil by setting up an original laboratory experiment. 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.

## 2. Materials and methods

### 2.1. Site description

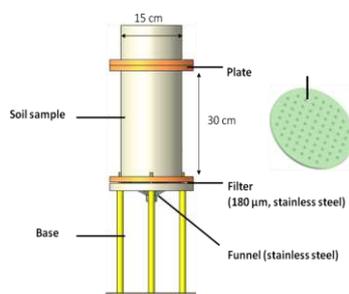
This study was carried out on an in-service wood preserving facility site, located in the north east of France. The wood preserving activity for phone and power line poles started in 1929. The main chemicals used were chromated copper arsenate compounds (CCA) and creosote distillates. On site, soils are sandy clay loam. The mean depth of the water table is 6 m from the ground surface; the water table level variation is about 1 m over a year depending on the season. A regulatory monitoring of the groundwater quality was initiated by a prefectural order in 1998 and 4 water wells are monitored: a well was installed upstream (W83), an other one downstream (W84), and 2 were placed in the centre of the site (W82 and W14). The well W14 is an industrial well: a pump is installed at about 9 m below the ground surface and water is pumped for the need of industrial activities, in order to dilute CCA compounds. Groundwater quality is measured twice a year.

### 2.2. Soil sample for laboratory experiments

Soil samples were taken between 0 and 6 m depth, right upstream the industrial well where the main pollution source was supposed to be localized after the analysis of the As groundwater concentrations over time. A quantity of 21 kg of soil was sampled 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-samples was dried, sieved, grinded and submitted to a sequential and a total extraction. 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.

### 2.3. 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**

## 2.4. 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. 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 rise 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.

## 3. Results and discussion

### 3.1. Results from field experiments

Arsenic concentrations in the wells upstream and downstream were below the EU guidelines for drinking water quality ( $10 \mu\text{g L}^{-1}$ ). Concentrations were a little bit higher in W82, showing only one value above the EU guidelines. Nevertheless, arsenic concentrations in W14 were much higher and above the EU guidelines, varying between 5 and  $100 \mu\text{g L}^{-1}$ . Given the small radius of influence of the industrial well (below 1 m), the pollution source was supposed to be located right upstream this well. In addition As release was highly correlated to the water table level: the higher the water table level, the higher the As concentration in groundwater. This indicated that when the water table level was high, water was in contact with the pollution source allowing As to migrate in solution.

### 3.2. Results from laboratory experiments

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 fractions.

## 4. Conclusions

In this work, As release was studied at the field scale and at the laboratory scale. Experiments in the field helped us to identify the main pollution source location and showed us that As release in groundwater was highly correlated to the water table level. An original laboratory column allowed 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 being released from soluble and exchangeable fractions. This study paves the way for a better understanding of trace elements behavior in the vadose zone of the soil during risk assessment studies and gives a new approach to study this part of the soil.