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## **Transfert concomitant des bactéries et des métaux lourds dans un bassin d'infiltration : étude en colonnes en conditions hydrodynamiques réelles**

Bacteria and heavy metals concomitant transfer in an infiltration basin : columns study under realistic hydrodynamical conditions

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### **RESUME**

Cette étude s'intéresse aux mécanismes contrôlant la mobilité des métaux (Zn, Cu, Cd) et des bactéries dans un bassin d'infiltration, et à l'influence des facteurs hydrodynamiques sur le transport, afin d'évaluer le risque potentiel pour le sol et la nappe sous-jacents. Ces éléments s'avèrent très mobiles, mais leur migration est peu liée. La mobilisation depuis la couche de surface des bassins d'infiltration semble gouvernée par des mécanismes de dissolution du complexe organo-minéral. Les métaux dissous issus de cet horizon peuvent ensuite s'adsorber au cours de leur transfert avec des colloïdes ou des particules issus du sol sous-jacent, ce qui augmente les quantités de métaux transférées. Cette part de transfert colloïdal est sensible aux phases de séchage du sol et à la vitesse de percolation.

### **ABSTRACT**

This study aims at: (i) identifying the mechanisms involved in the metals (Zn, Cu, Cd) transfer in an infiltration basin, focusing on the role of the bacterial compartment; (ii) evaluating the influence of the hydrological conditions; (iii) estimating the "purifying" capacity of the infiltration basin, and in particular the role of the original soil constituting the vadose zone. We mainly demonstrated that bacteria and metals contained in the surface layer are mobile, but are not significantly bound. Dissolution of organo-mineral matter appears as the main mechanism controlling mobility in soil surface. Dissolved metals are then liable to sorb on colloids originating from the sandy underlying soil, that increase the quantities of eluted metals. This metallic colloidal compartment is sensitive to soil drying and flow rate.

### **MOTS CLES**

Bactéries, bassin d'infiltration, colonnes, conditions hydrodynamiques, métaux lourds.

## 1 CONTEXT AND OBJECTIVES

In the East of Lyon (France), stormwaters are discharged in infiltration basins to recharge aquifers, and the contaminants contained in the runoff water are supposed to accumulate at the soil surface. Infiltration practices increase consequently the organic matter content and heavy metals concentrations in the vadose zone of the basin, particularly in the surface layer [1]. The soil surface then contains a large variety of compounds liable to interact with metallic pollutants through a wide range of bio-physico-chemical reactions [2]. But little is known concerning the fate of pollutants through the vadose zone and the eventual risk for the groundwater.

Heavy metals may be mobilised by the rainfall events, under dissolved and particulate or colloidal form [3]. In particular, the colloidal compartment, including bacteria (abundant, reactive and mobile natural organic colloids) are liable to facilitate the transfer of heavy metals [4]. Fine particles and organic matter hydrophobicity also contribute to basins water clogging and water accumulation at the surface of the basin; drying occurring between two rainfall events will modify soil's properties and components' availability for transfer. Thus hydrodynamical conditions, i.e. frequency, intensity and duration of rain events, influence the basin's functioning and the mobility of compounds contained in the soil surface.

Therefore, the present study aims at:

- identifying the mechanisms involved in the metals transfer, focusing on the role of the bacterial compartment;
- evaluating the influence of the hydrological conditions;
- estimating the "purifying" capacity of the infiltration basin, and in particular the role of the original soil constituting the vadose zone.

For this purpose, the superposition of the polluted surface layer (A-Horizon) and the underlying quasi-unpolluted underlying soil (B-Horizon) was reproduced in repacked lysimeters. An alternation of drying/rewetting phases was simulated, based on realistic volumes generated in the infiltration by natural rainfalls. The originality of this study lays in considering a complex anthropogenic soil and its behaviour under realistic hydrological conditions.

## 2 MATERIALS AND METHODS

### 2.1 Site presentation

The infiltration basin studied is situated in Chassieu (East of Lyon, France). The watershed is mainly industrial and 75% waterproofed. The area of the watershed is 185 ha, the surface of the infiltration basin is 1 ha : that is why volumes entering into the basin during rainfalls are important.

The soil of the infiltration basin is constituted by the two well-discriminated layers : a thick polluted horizon (A-Horizon) on a quasi unpolluted underlying soil (B-Horizon), originating from a fluvio-glaciary carbonated deposit.

### 2.2 Columns experiments

The superposition of the two horizons (5 cm A-Horizon on the top of 36 cm B-Horizon) was reproduced in three 20-cm diameter columns. Total pore volume was 4.12 L (B-Horizon porosity is 29% and A-Horizon's one reaches 69%).

Columns were alimented with a NaBr  $10^{-3}$  M solution ( $120 \mu\text{S}\cdot\text{cm}^{-1}$ ) with an up-to-down flow, homogeneously distributed on the soil surface. Solution exceeding the infiltrating capacity of the soil was let accumulating at the soil surface. Leachates were collected at the bottom of the columns, let free to enable the leaching of colloidal

and particulate matter (the soil was supported on a drilled plaque with 1-mm diameter holes). Boundary conditions were realistic to represent drainage by macropores [5]. At three different depths, B-Horizon layer was manned with (Campbell) CS 616 probes and (SDEC) tensiometers (SKT 850T sensors on SMS(S) tubes) to respectively monitor soil water content and suction. Columns' design is represented on Figure 1 (with sizes in mm):

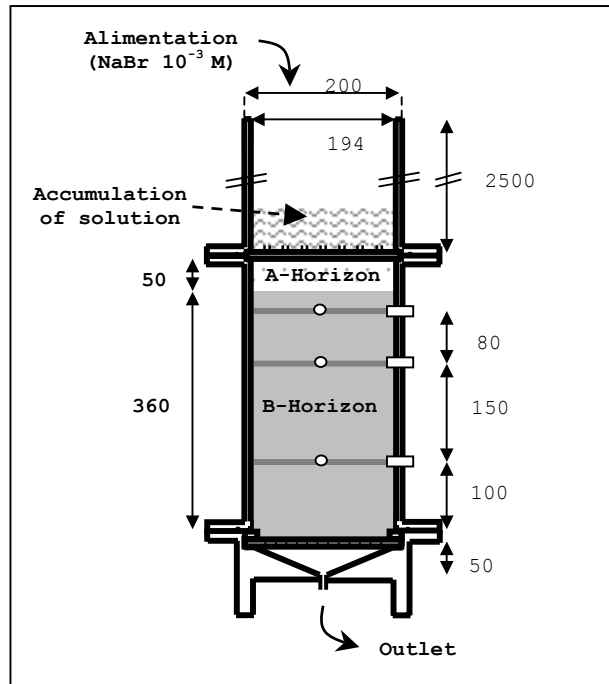


Figure 1 : Column experiments

### 2.3 Simulated events

3 kinds of rainy events were simulated. A Mean Event (ME) was first assumed to last 4 hours at the rate of 1.5 mm/h. Then intensity and duration of two extreme events were statistically (with inverse Student test at 95%) determined from one-year (2004) data on two pluviometers situated close the study area. An Autumnal Event (AE) was defined as an event whose duration was longer than the one of 95% of the events; a Thunderstorm (T) had an intensity higher than the one of 95% of the events. Total height of water was 20 mm for both AE and T events.

The volumes entering into the infiltration basins during these 3 events were then calculated, from the ratio of the area of the watershed and the area of the infiltration basin. This calculation was applied to the surface of the columns: columns were supplied with 0.73 m of solution within 4 hours for a Mean Event, and 2.42 m within 100 min and 1100 min for respectively Thunderstorm and Autumnal Event.

The three columns were simultaneously supplied with an alternation of rainfall events separated by 3-days or 2-weeks drying periods. One column was periodically sacrificed to monitor eventual metal and bacteria deposition (Fig 2):

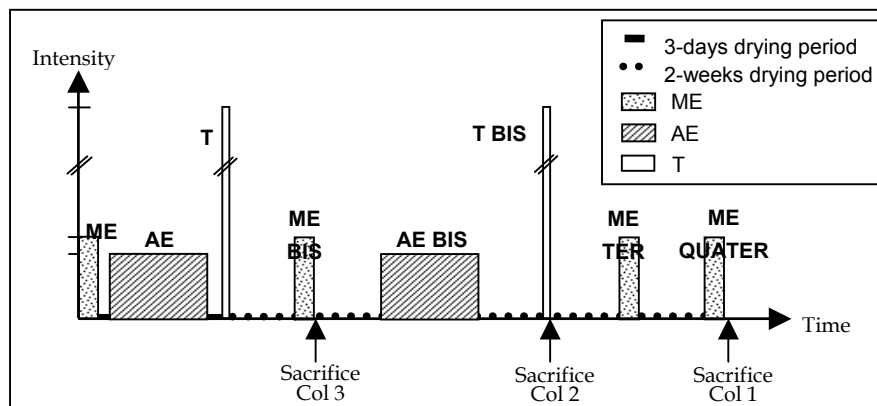


Figure 2 : Alternation of rainfall events simulated

## 2.4 Biological and physico-chemical analyses

Metals (Zn, Cu, Cd) contents, TOC, Ca, Na, NO<sub>3</sub>, Br and Cl contents were measured. pH and conductivity of the samples were also followed. Physico-chemical measurements were realized on 0.45 µm filtrated samples and on total fractions, to discriminate the pseudo-total and particulate forms of the elements.

Eluted bacteria (organic colloids) were enumerated thanks to DAPI staining, coupled to epifluorescence microscopy. TEM (Transmission Electronic Microscopy) observations aimed at identifying eluted colloids, and their eventual bound with metals.

## 3 RESULTS

### 3.1 Soils characterization

The underlying soil originates from a fluvio glaciary carbonated deposit, mainly constituted with CaCO<sub>3</sub>: pH is high (9.65) and organic content is very low (0.37%). In particular, 1 g of dry soil contains 1,83.10<sup>7</sup> bacteria. This horizon is unpolluted (40 mg.kg<sup>-1</sup> of dry weight Zn, 7.93 mg.kg<sup>-1</sup> DW Cu, 0.23 mg.kg<sup>-1</sup> DW Cd).

In the surface soil, pH is lower (7.23) and conductivity higher (219 µS.cm<sup>-1</sup>). It is characterized by high contents of heavy metals (2 g.kg<sup>-1</sup> DW Zn, 182 mg.kg<sup>-1</sup> DW Cu, 5 mg.kg<sup>-1</sup> DW Cd), bacteria (1,23.10<sup>10</sup> bacteria.g<sup>-1</sup> DW) and organic matter (13.22 % DW).

The granulometric distribution (determined by laser granulometry) demonstrates a strong enrichment of the A-Horizon compared to the sandy B-horizon on fines particles (0.3-1 µm).

This characterization underlines the evolution of B-Horizon toward A-Horizon due to infiltration practices: A-Horizon became a polluted and very reactive compartment liable to interact with metals by a wide range of mechanisms, including precipitation/dissolution, physical and chemical sorption/desorption (e.g. ion exchange, complexation).

### 3.2 Mobility and mobilisation

The three columns are greatly reproducible.

During the whole set of experiments, conductivity, TOC, Ca, NO<sub>3</sub> and Cl concentrations increase strongly with time of elution, until less than one pore volume has percolated (because of unsaturated conditions), then decrease to a background value. Figure 3 shows the different concentrations obtained with the first event (ME) on Column 1.

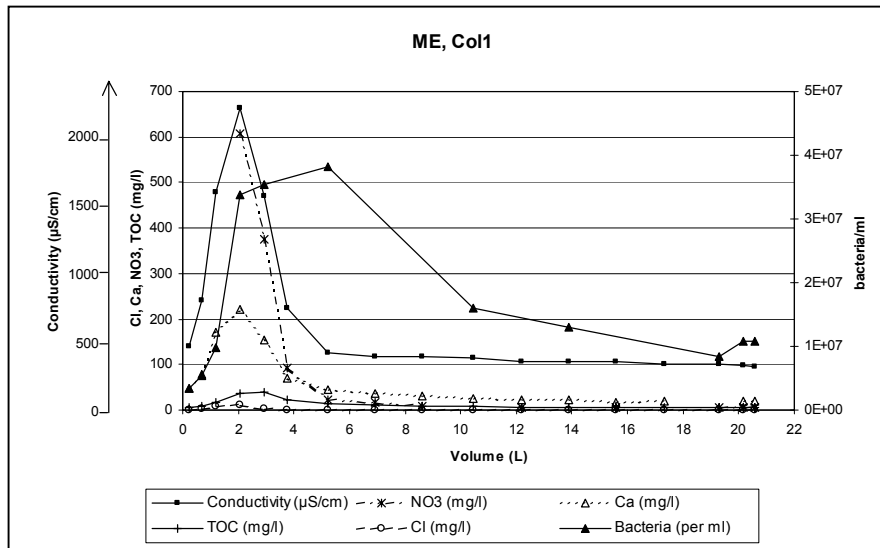


Figure 3 : Evolution of conductivity, NO<sub>3</sub>, Cl, Ca, TOC and bacteria contents during ME experiment (Column 1)

Conductivity, Ca, NO<sub>3</sub> and Cl reach their maximum concentrations value simultaneously: they are mainly released from A-Horizon, and may be considered as tracers-like parameters. During the two first experiments (ME and AE), TOC peak occurs a few time later, suggesting additional interactions in B-Horizon or stronger retention. Then it is simultaneous with conductivity, Ca, NO<sub>3</sub> and Cl maximum values. The peak attests that these elements principally originate from the surface A-Horizon, with few interactions with the B-Horizon.

At least TOC and Ca are under dissolved form (since no difference were measured between concentrations in 0.45-µm-filtered and no-filtered samples), and elements are continuously released at a constant background concentration as soon as a "permanent" regime is reached. This would suggest that dissolution mechanisms, rather than sorption, control the mobilisation of major ions and organic matter: the pool of mobile matter seems not limited, and no diffusion stage limit the mobilisation. The preponderant release mechanism is thus supposed to be a concomitant dissolution of organic and mineral compounds from the organo-mineral complex of A-Horizon. Decrease of Na content (contained in alimentation solution) at the beginning of ME experiment, suggests cation exchange reactions, probably with Ca, but this mechanism is insignificant compared to Ca release due to dissolution.

### 3.2.1 Bacteria

During the first event (ME), bacteria content reaches a plateau rather than an peak (Fig 3). Moreover, for both first (ME) and second (AE) experiments, bacteria maximum concentration is also reached after maximum values of conductivity, Ca, Cl, NO<sub>3</sub> contents, while the peaks are simultaneous for the following experiments.

Semi-qualitative observations with DAPI staining also showed that bacterial populations eluted at the beginning of ME elution are smaller than afterwards. This gives evidence of a slower mobility of bacteria through B-Horizon, while the soil is getting structured under the water pressure. This hypothesis is supported by tensiometers and TDR measurements: for the first experiment (ME) percolation is slightly slower in the upper part of the columns, with lower water content, while during following experiments the solution percolate more homogeneously through the columns. During ME, only dissolved matter and the smallest bacteria can migrate through the column, until structural macropores get wider under water pressure, while greater cells remain sorbed or physically entrapped in the soil matrix.

No delay between dissolved matter and bacteria breakthrough is observed for following experiments. But unlike dissolved matter, bacteria contents go on decreasing without reaching any background value. Moreover, sacrifices demonstrated an enrichment on bacteria of B-Horizon : it contains at the end of the cycles 3 to 10-fold more bacteria than initially. This would suggest that bacteria mobility is limited either by their size (in B-Horizon) or by any diffusion or/and detachment step (in A-Horizon). For instance, similar kinetic patterns between TOC and bacteria contents suggests organic matter specific sorption mechanisms in the sand.

### 3.2.2 Metals

At least Zn and Cu are eluted under both dissolved and colloidal or particulate form (see Fig 4 for ME experiment). However, previous studies implementing sole A-Horizon under the same hydrodynamical conditions showed that metals originating from this polluted layer were entirely soluble. Thus B-Horizon generates a pool of particulate and colloidal matter liable to interact with metals. This hypothesis is supported by TEM observations on leachates stemming from B-Horizon, where oxyhydroxides and clay particles were identified. Metals would thus be mobilized under dissolved form from the A-Horizon and be afterwards sorbed on colloids in B-horizon.

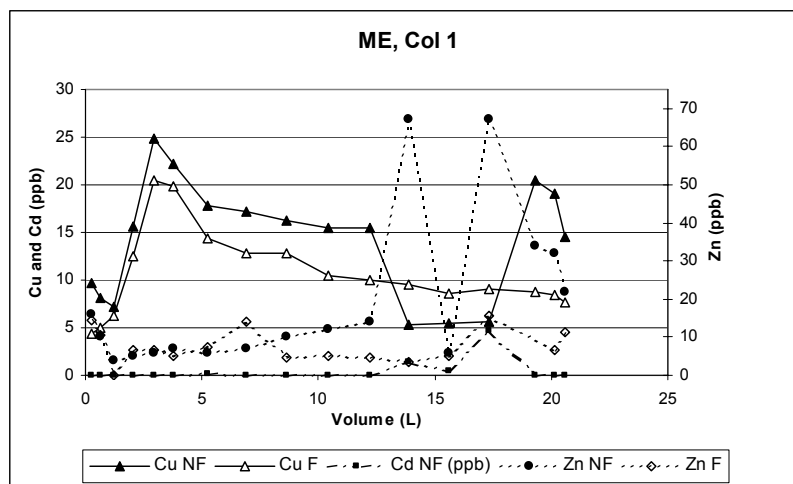


Figure 4 : Evolution of dissolved (F) and pseudo-total (NF) metals contents during ME experiment (Column 1)

Moreover, Cd and Zn have a specific behaviour, compared to Cu. This is first illustrated during the first event simulated (Fig 4). Dissolved (0.45 µm filtered subsample, F) and pseudo-total (non-filtered sample, NF) Cu concentrations reach a maximum value a few time later than conductivity, Ca, TOC, NO<sub>3</sub> and Cl contents. Zn and Cd concentrations are maximum 10 pore volumes later, while pseudo-total Cu content drastically decreases, suggesting either competition or opposite mobility mechanisms for Cu on the one hand, and Zn and Cd on the other hand (this particularity was observed for the 3 columns).

During next experiments, Cd concentrations are under detection limits. Both Zn and Cu pseudo-total contents are very high at the beginning of elution, unlike dissolved fraction. Non-dissolved metallic fractions remain significant: until 40 % of Cu (during T) and 70% of Zn (during ME) collected are under particulate form).

Analysis of the layers collected after sacrifices showed no significant enrichment in metals of B-Horizon. But it does not mean that there is no metallic migration [6]. Moreover, we simulated the equivalent of 104 mm of pluviometry for whole cycle on Column 1; this value is well under one-year pluviometry in the East of Lyon (≈650 mm) and not enough important to assess a long-term risk. Much more Zn and Cu are however eluted from columns than predicted from small columns with sole A-Horizon: presence of B-Horizon increases metal transfer. *In situ* measurements also showed bigger concentrations of Zn in the groundwater than in surface runoff.

### 3.3 Influence of the hydrological conditions

#### 3.3.1 Specificity of the first event simulated (ME)

ME hydrological and structural specificity was mentioned above. Moreover, ME mobilises greater quantities of Cu (the peak is 2 times greater than during the second event), Zn (3 times), COT (3,5 to 4 times), NO<sub>3</sub> (7 times), Ca (3 times) and Cl (33 times). Oppositely, maximum value of bacteria content is only twice more important during ME than AE (negligible differences for biological parameter).

#### 3.3.2 Influence of drying phases

Drying phases seems to enhance the mobilisation of dissolved mineral and organic matter (at least conductivity and Ca, TOC contents).

Oppositely, no significant influence of drying phases on metals mobility were observed: neither the quantities nor the repartition between the dissolved and particulate phases were modified.

From the T event, bacteria release is very similar from one infiltration to another: drying periods have no consequence on their mobility.

#### 3.3.3 Influence of alimentation flow rate

Tensiometers and TDR measurements corroborate that water content was higher in the columns during T event than during ME and AE. A higher flow velocity is supposed to reduce bacterial adhesion [7] and increase bacteria dispersivity [8]. Yet T and T BIS events mobilize less bacteria all along infiltration, and the maximum peak is not wider. Thus cells retention seems to be due to entrapment into inner aggregates or sorption onto soil surface, rather than onto air-water interfaces. The main mechanism controlling bacteria mobilization seems to be the dissolution of the organo-mineral complex, to whose stability microorganisms contribute. Their mobility is then governed by size-depending processes.

Similarly, conductivity, TOC or Ca contents are not influenced by the flow velocity. This supports the hypothesis that dissolution mainly govern their release from an organo-mineral gangue.

Zn and Cu pseudo total contents increase at the beginning of T infiltration compared to AE infiltration, while dissolved content seems not to be influenced. Thus bound metals and colloids are more mobile or more detached with higher water content and higher flow rate. Moreover, as already seen in small columns with A-Horizon, quantities of Zn eluted during AE and T are similar (it depend mainly on height of solution alimanted) while quantities of Cu eluted during AE is much greater than with T, because a significant concentration of Cu is continuously released.

#### 4 DISCUSSION AND CONCLUSION

- Notable differences were observed between Cu on one hand and Zn and Cd on the other hand; such differences had already been reported, e.g. by [6]. In particular, they do not breakthrough simultaneously and even seems to be negatively correlated. Moreover, quantities of mobile Zn depend on total height of water infiltrated, while Cu is correlated to duration of events.
- Cu affinity for organic matter is usually evoked to explain these differences. But here TOC and bacteria seems to be sensitive to different mechanisms. Bacteria are abundant, but their transfer is limited by their size, and eventual diffusion or detachment steps. If they participate to metallic transfer facilitation, they do no accelerate the transfer and are not preponderant. Great quantities of (eventual pathogen) microorganisms are however released.
- Non-bacterial colloidal or particulate parts of mobile metals are however significant, as demonstrated with TEM observations and comparison of filtered and unfiltered samples: dissolved metals from the polluted horizon may recombine with particles emanating from B-Horizon, in particular clays and oxyhydroxides are suspected to participate to the metal transfer.
- The first event simulated determine the soil structuration during infiltration. This point underlines the bias introduced in column experiments as soon as one single event is simulated. Drying phases influence the mobility of dissolved mineral and organic matter, but neither metals nor bacteria. An higher infiltration rate has no influence on dissolved matter and bacteria, but the metallic colloidal fraction is sensitive to high flow.

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