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Accumulation de métaux lourds dans le sol superficiel d'ouvrages de gestion à la source des eaux pluviales : variabilité de la contamination et comparaison inter-sites

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ABSTRACT

In urban areas, the increasing use of infiltration devices for stormwater management raises some concerns about the potential for micropollutants' accumulation in soil and transport to groundwater. It is necessary to properly appraise the spatial variability of contamination within these systems, so as to (i) provide guidance for their selection, design, and needs for maintenance after several years of operation, (ii) optimize the representativeness of soil samplings in experimental studies, and (iii) assess the modelling framework required for long-term evaluations. In this study, we determined the spatial distribution of three trace metals (copper, lead, zinc) in the surface soil of several infiltration facilities with contrasting catchments and hydraulic configurations. On each study site, we performed a series of high-resolution samplings and physical measurements (TDR soil moisture, volatile matter, thickness of the dark upper horizon of soil). Analyses generally show a significant enrichment of Cu, Pb and Zn in the topsoil, but this accumulation is restricted to a narrow area, since concentrations markedly decrease with increasing distance to the inflow point(s) of the facilities. The inter-site comparison highlights the influence of hydraulics and heterogeneous infiltration on the contamination patterns. Moreover, the metal concentrations often appear to be correlated to the volatile matter content and thickness of the upper horizon.

RÉSUMÉ

Si les systèmes favorisant l'infiltration des eaux de ruissellement connaissent un fort développement en zones urbaines, des interrogations demeurent quant à l'accumulation de micropolluants dans le sol de ces dispositifs, et à leur éventuel transfert vers les eaux souterraines. Il est nécessaire d'appréhender convenablement la variabilité spatiale de la contamination dans ces ouvrages, afin de (i) fournir des recommandations pour orienter le choix des solutions techniques, leur conception et leur maintenance, (ii) assurer une représentativité optimale de l'échantillonnage de sol au cours des études expérimentales, et (iii) apprécier la complexité des modèles à mettre en œuvre pour des évaluations prospectives. Cette étude s'intéresse à la distribution spatiale de trois métaux traces (cuivre, plomb et zinc) dans le sol superficiel de différents ouvrages d'infiltration implantés dans des contextes urbains contrastés. Sur chaque site, on a réalisé une série d'échantillonnages à haute résolution spatiale, accompagnés de mesures physiques (teneur en eau TDR, teneur en matières volatiles, épaisseur de l'horizon superficiel sombre). Les analyses ont mis en évidence un enrichissement significatif en métaux des premiers centimètres du sol, généralement localisé autour du/des point(s) d'arrivée de l'eau dans les dispositifs. La comparaison inter-sites démontre l'influence du fonctionnement hydraulique des ouvrages, ainsi que des flux d'infiltration hétérogènes, sur la répartition des contaminants. En outre, les teneurs en métaux sont souvent corrélées à la quantité de matières volatiles et à l'épaisseur de l'horizon superficiel.

KEY WORDS

Heavy metals, Infiltration, Soil contamination, Spatial variability, SUDS

1 INTRODUCTION

Land-use changes due to urban sprawl result in rising levels of impervious cover, which increases peak flows and volumes of runoff water to be drained away. In that context, Sustainable Urban Drainage Systems (SUDS) are becoming a widespread approach for stormwater management, because of their largely recognised hydraulic and hydrologic benefits (Ferguson, 1994). Though, the implementation of infiltration facilities brings about several uncertainties about the fate of contaminants within these devices, and their potential impact on soil and groundwater (Pitt, et al., 1999).

Experimental works led on such facilities revealed a significant accumulation of metals and PAH in the upper horizon of soil; the concentrations of contaminants tended to decrease with depth, in such a way that the contents below 50 to 70 cm were generally found to be close to the geochemical background (Mikkelsen, et al., 1996; Winiarski, et al., 2006). Most of the results obtained so far would suggest that infiltration-based SUDS exhibit a good potential for short- and mid-term pollution retention. However, in these studies, the selection of the coring points did not systematically consider the spatial distribution of contaminants in the surface soil, which was yet shown to exhibit high variability (Le Coustumer, et al., 2007; Kluge & Wessolek, 2012). Assessing this distribution may also provide valuable indications for prospective evaluations, regarding the need for models with higher complexity than the usual one-dimensional framework, in which the infiltration fluxes are assumed to spread homogeneously at the soil surface (Quinn & Dussaillant, 2014).

Although several sources of variability have been identified, among which topography, disparities in flooding areas, "historical" accumulation, or the presence of technical installations (*e.g.* street lamps or barriers), the distribution of contaminants in the topsoil does not appear to be evidently predictable. The objectives of the present study are (i) to address the accumulation, and resulting spatial distribution, of three trace metals (copper, lead and zinc) in the upper horizon of several SUDS, and (ii) to emphasize factors which may explain the different contamination patterns, through an inter-site comparison of physical parameters, morphologies and watersheds of the investigated systems.

2 METHODOLOGY

Description of the study sites. A series of 5 SUDS allowing for water infiltration, which had been in operation for more than 10 years in the Paris region, were selected for their contrasting hydraulic characteristics, morphologies and watersheds (Table 1).

Sampling and *in-situ* measurements. The field campaigns were undertaken between April and November 2015. Samplings were carried out along a rectangular grid with < 2.5 m^2 meshes. At each node: (i) the upper 2-3 cm of soil were sampled with a stainless steel trowel; (ii) soil moisture was measured with a time-domain reflectometer (*Spectrum Technologies*, FieldScout probe TDR 100), in order to visualize the flow pathways in the devices; (iii) a 30-cm-deep soil core was dug with a hand auger, so as to measure – when distinguishable – the thickness of the dark upper horizon, which was noticed to be variable in space within several study sites.

Laboratory analyses. The soil samples were oven-dried at 40°C for 7 days, gently broken and then passed through a 2-mm nylon sieve. The Cu, Pb and Zn concentrations were determined by X-ray fluorescence (*Thermo Scientific*, Niton™ analyzer XL3t). The limits of quantification are sample-dependent, as they vary according to the signal received by the analyzer, but they were in any case lower than 20, 10, and 30 mg/kg for copper, lead, and zinc, respectively. A fraction (8-10 g) of each sample was calcined at 550°C for 6 hours, so as to determine its volatile matter content from mass difference. Soil pH (Table 1) was determined in triplicates from composite surface samples, in a solution of soil and ultrapure water (volumetric ratio of 1:5) after 1h of equilibration.

Table 1. Description of the investigated infiltration facilities

Site name	Fleury	Dourdan1	Dourdan2	Vitry	Sausset
Type of device	Infiltr. basin	Infiltr. basin	Swale	Swale	Small basin
Watershed	Residential	Road + car park	Mixed	Industrial + road	Car park
Watershed area	2 ha	1.2 ha	3000 m^2	900 m^2	500 m^2
Inflow of water	Pipe (Ø300)	Pipe (Ø600)	Pipe (Ø500)	Lateral openings	Direct runoff
Operating time	> 25 years	> 20 years	11 years	10 years	14 years
Soil pH	7.8 ± 0.2	7.4 ± 0.1	6.8 ± 0.05	8.1 ± 0.05	7.9 ± 0.1

Spatial interpolation and cartography. The different scalar fields were interpolated using universal kriging with a local first-order trend model, so as to account for the deterministic component of these variables (Chauvet & Galli, 1982). Cartographies of the trace metal contents, soil moisture, volatile matter and thickness of the surface horizon were generated using a 10 cm x 10 cm interpolation grid over the whole area of the devices.

3 RESULTS AND DISCUSSIONS

The distribution of trace metals appears to be variable in space – as observed in previous studies – but typically organised with respect to the water inflow point/zone. Figure 1.a gives an illustration of the contamination cartography for the study site *Dourdan1*. In infiltration basins with one point-source inflow, the highest concentrations are found near the inlet, from which they tend to decrease with increasing distance; at the opposite extremity, the measured values are close to the limits of quantification of the analyser, and even lower in the case of Pb. Similarly, all along the swale with lateral openings, the metal concentrations decrease with increasing distance from the pavement, even if the values near the road are rather variable. At the scale of the whole systems, the relative standard deviation of the metal contents ranges between 27 and 106%: this is similar to the values found by Le Coustumer et al. (2007) in a wider "centralized" basin, except that zinc is the metal which shows highest dispersion in most assessed devices, whereas in the above-mentioned study the order was Zn < Cu ≤ Pb. Globally, metal accumulation appears to be restricted to a limited region of the devices, which is probably due to the fact that they have been designed for a ≥ 10-year storm event: thus, runoff water is likely to infiltrate before spreading over the entire facilities for frequent rainfall events. This was confirmed in several sites by the spatial distribution of soil moisture (data not shown).

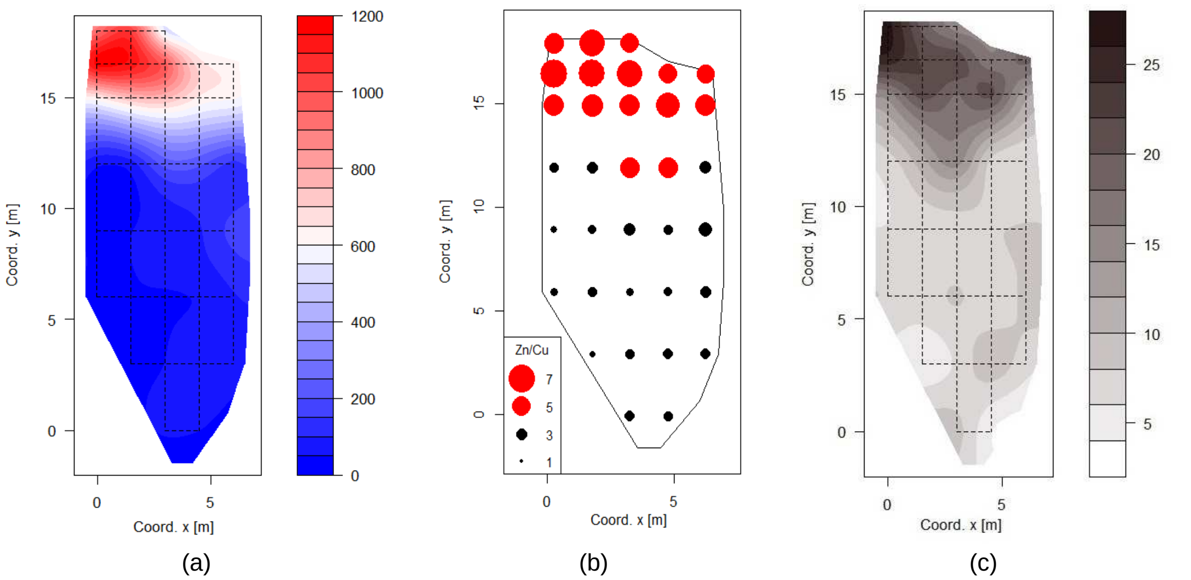

Figure 1. Some results for the study site *Dourdan1*. Cartographies of (a) the concentration of zinc [mg/kg], (b) the ratio Zn/Cu in each sample (red circles indicate the points where Zn/Cu ≥ 4), and (c) the volatile matter content [%]. Samples were taken at each node of the dotted grid. The coordinates of the inflow point are (0,18).

Generally, no data is available about the initial state of the soil, which is yet necessary to properly evaluate the contaminant enrichment due to water infiltration. Additionally, the common assumption that nearby sampling points, uninfluenced by water infiltration, may represent "background" concentrations (*e.g.* Legret, et al. (1992) and Lind & Karro (1995)), is likely to be unsuitable for systems where the surface soil has been either excavated or amended during the construction works, which is the case of most investigated facilities in the present study. Consequently, metal accumulation was rather appraised through a comparison between the concentrations found in the most and the least contaminated areas of the devices – assuming that other sources of metals (such as atmospheric deposition) induce a homogeneous increase in the topsoil concentrations. These two areas were defined as the zones where the metal concentrations are respectively higher than the 9th decile, and lower than the 1st decile of the whole measurements. The average concentration in the second zone is considered here as a "reference" concentration.

Figures 2.a to 2.c illustrate the significant increase in the metal contents with respect to the reference concentrations ($p < 0.05$), except for lead in the car park built in 2001 (*Sausset*), where minimal Pb contamination was expected since leaded-fuel was banned in 2000 in France. The highest Pb concentrations are found in *Vitry*, which collects runoff water from an industrial watershed. The infiltration basins *Fleury* and *Dourdan1* exhibit the highest enrichment ratios (respectively 6 and 8 for Cu, 8 and 21 for Pb, 8 and 28 for Zn). In the most polluted regions of the facilities, the concentrations generally exceed the “anomaly” thresholds of the French and Dutch standards (dotted lines on Figure 2), and even in several cases the “intervention” value (solid line), suggesting a need for local soil maintenance (AFNOR, 1985; NMHSPE, 2000).

The ratios between metals were found to be variable in space in several facilities. Figure 1.b displays the values of Zn/Cu at each sampling point of the site *Dourdan1*. Pb was not taken into consideration because of the samples below the limit of quantification. In the part of the device with lowest concentrations, Zn/Cu is fairly uniform and ranges between 2-3, which approximately corresponds to the ratio between background Zn and Cu contents in agricultural soils of this region (Baize, et al., 2007). Conversely, significantly higher values of Zn/Cu (5-7) are found in the entire contaminated zone, with a sharp transition from one region to the other. Similar trends are visible in the study sites *Vitry* and *Sausset* (Figure 2.d). This difference in ratios is an additional evidence that metal contamination does not originate from the natural soil environment, indicating the contribution of surface runoff, and it confirms (in three sites) that certain parts of the devices are not active during stormwater infiltration.

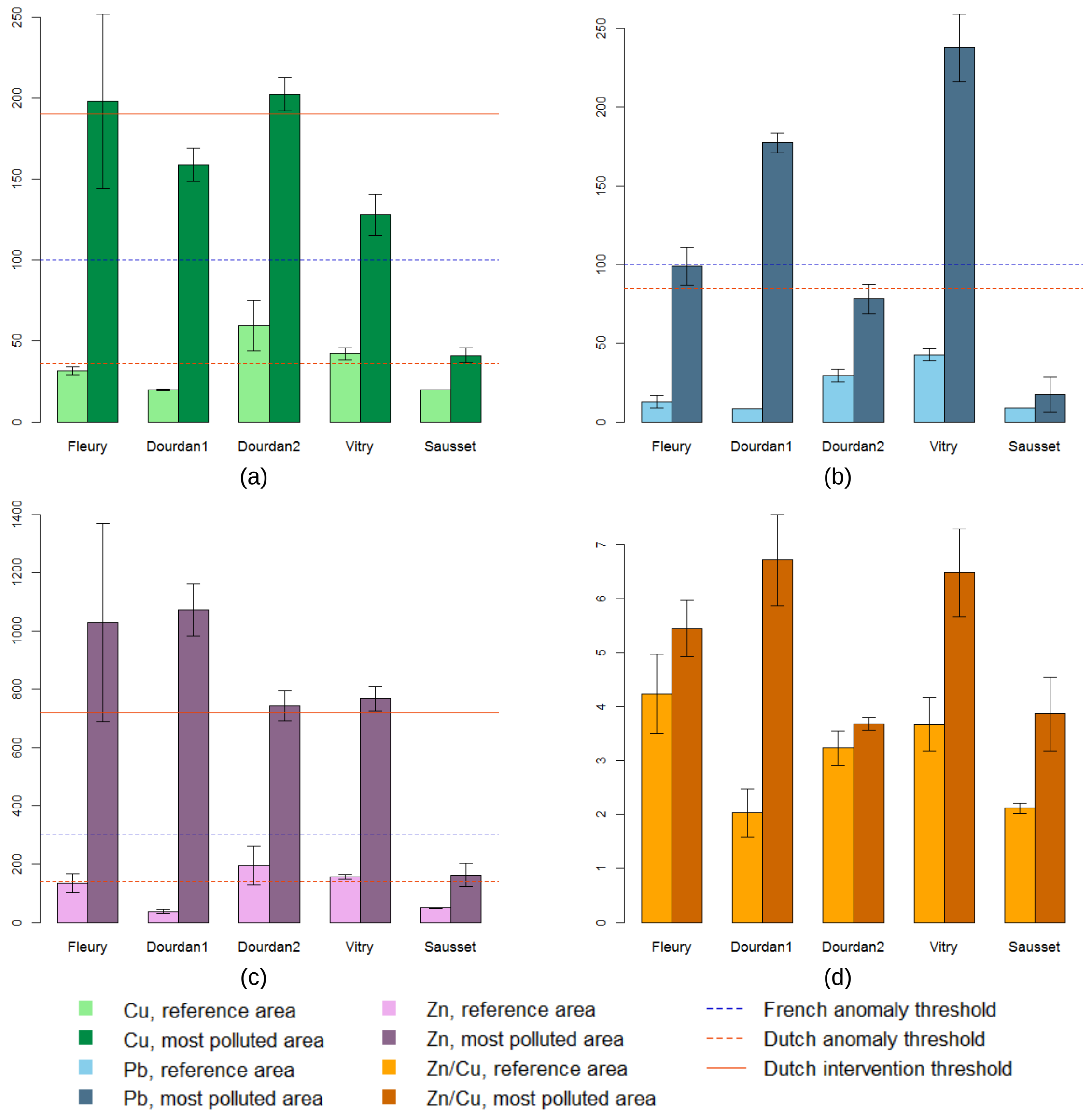

Figure 2. Mean concentrations of (a) copper, (b) lead, and (c) zinc [mg/kg] measured in the most and the least polluted areas of each investigated device. Comparison with the French (blue lines) and Dutch (red lines) standards (AFNOR, 1985; NMHSPE, 2000). (d) Mean ratio Zn/Cu [-] in these two areas.

In the larger infiltration basins, the concentrations of trace metals appear to be significantly correlated to both the volatile matter content (Figure 1.c) and the thickness of the upper horizon of soil (data not shown); Pearson's *r* coefficient was higher than 0.8 whatever the metal. This might indicate that the contaminants in runoff are mostly bound to organic-rich suspended solids, which settle rapidly when they enter the basin. However the previous observation does not hold for the smaller devices such as vegetated swales, in which the upper horizon was homogeneously amended with planting soil during and/or after the construction works.

4 CONCLUSIONS

This study has addressed the spatial distribution of trace metal concentrations (Cu, Pb, and Zn) in the surface soil of various infiltration systems, through a series of high-resolution samplings. Contamination may exhibit a high variability within a facility, however it appears to be spatially structured according to the distribution of the infiltration fluxes. Hence the area surrounding the inflow point(s) is significantly more polluted than the other extremity of the device, since runoff water is not likely to spread over the entire soil surface during common rainfall events. In order to get a representative estimate of the global contamination level in a SUDS, this "typical" variability should be taken into consideration for the *a priori* selection of the samplings points.

The ratio between metals, which was found to be significantly different in the contaminated and the non-contaminated zones of several devices, with a sharp transition between these two areas, may represent the signature of runoff-sourced contamination. In "older" infiltration basins, which have been operating for more than 20 years without soil renewal, both volatile matter content and thickness of the dark upper horizon of soil have been observed to be variable in space; furthermore, interesting correlations have been highlighted between the concentrations of metals and these two parameters.

These results have practical implications concerning soil maintenance, insofar as the area which would require an intervention only corresponds to a small part of the systems (less than 20% of the whole area). Additionally, the basic assumption of 1-D modelling, which implies that the distribution of contaminants only varies with depth, has been proven to be unsuitable in the case of infiltration systems without a permanent pool of water, and should therefore be adapted for prospective evaluations of SUDS.

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