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Spatial distribution of heavy metals in the surface soil of source-control stormwater infiltration devices

– Inter-site comparison

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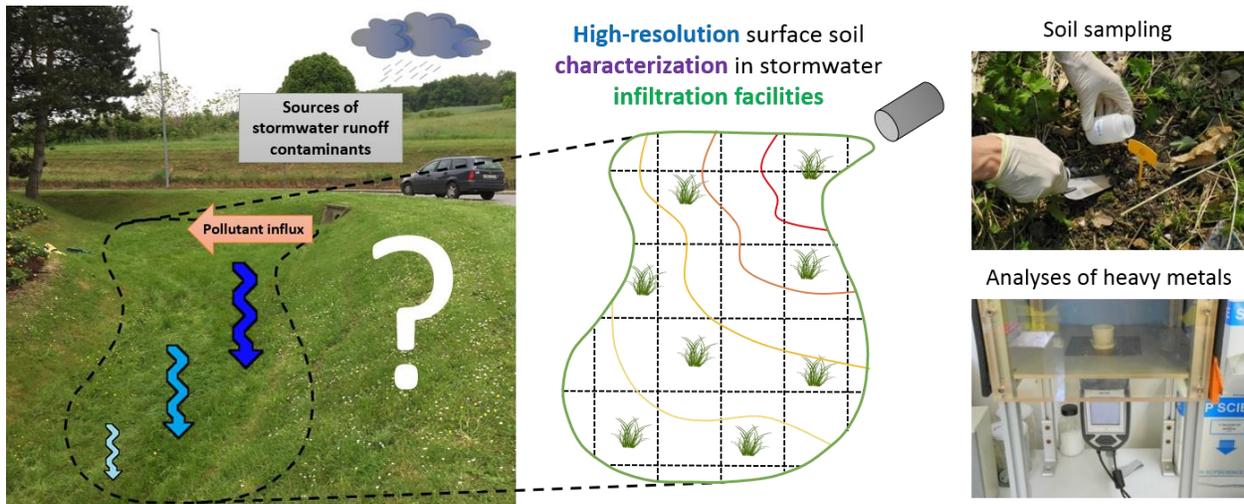
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ABSTRACT. Stormwater runoff infiltration brings about some concerns regarding its potential impact on both soil and groundwater quality; besides, the fate of contaminants in source-control devices somewhat suffers from a lack of documentation. The present study was dedicated to assessing the spatial distribution of three heavy metals (copper, lead, zinc) in the surface soil of ten small-scale infiltration facilities, along with several physical parameters (soil moisture, volatile matter, variable thickness of the upper horizon). High-resolution samplings and *in-situ* measurements were undertaken, followed by X-ray fluorescence analyses and spatial interpolation. Highest metal accumulation was found in a relatively narrow area near the water inflow zone, from which concentrations markedly decreased with increasing distance. Maximum enrichment ratios amounted to > 20 in the most contaminated sites. Heavy metal patterns give a time-integrated vision of the non-uniform infiltration fluxes, sedimentation processes and surface flow pathways

within the devices. This element indicates that the lateral extent of contamination is mainly controlled by hydraulics. The evidenced spatial structure of soil concentrations restricts the area where remediation measures would be necessary in these systems, and suggests possible optimization of their hydraulic functioning towards an easier maintenance. Heterogeneous upper boundary conditions should be taken into account when studying the fate of micropollutants in infiltration facilities with either mathematical modeling or soil coring field surveys.

GRAPHICAL ABSTRACT



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, and lessens infiltration into soils (Miller et al., 2014). Sustainable Urban Drainage Systems (SUDS), which contribute to the decentralization of stormwater management, have been proven to efficiently mitigate certain adverse impacts of urbanization on the water cycle, as they help control urban flooding, reduce combined sewer overflows, and participate in groundwater recharge (Dierkes et al., 2015; Zhou, 2014). While the use of facilities allowing for water infiltration is becoming a widespread approach in areas whose hydrogeological context enables it, their increasing implementation brings about some concerns about the fate of contaminants within these devices: given the micropollutant loads generated by urban watersheds (Gasperi et al., 2014), and the conservative behavior of several chemical species, long-term runoff infiltration may impair soil and/or groundwater quality (Mikkelsen et al., 1994; Pitt et al., 1999; Werkenthin et al., 2014). Operationally, the potential needs for soil maintenance or remediation to ensure a proper and sustainable functioning of infiltration-based SUDS are not clearly identified.

Previous experimental work led on such facilities revealed a significant accumulation of heavy metals (copper, lead, and zinc being among the most mentioned species) and hydrocarbons in the upper horizon of soil (El-Mufleh et al., 2014; Jones and Davis, 2013; Mikkelsen et al., 1996; Winiarski et al., 2006). It was often suggested that these systems exhibit a good potential for short- and mid-term pollution retention (Barraud et al., 2005; Napier et al., 2009). However, in most investigations, the sampling locations were not always based on a preliminary analysis of the contaminant distribution in the surface soil – as recommended for example in the standard ISO 10381-5 (2005). Since surface concentrations have been shown to exhibit high variability at the scale of a whole infiltration basin (Le Coustumer et al., 2007; Dechesne et al., 2004a), the derived contamination profiles may not have the same representativeness from one study to another.

The studies which specifically addressed the horizontal distribution of soil contamination in infiltration systems are scarce (Tedoldi et al., 2016); moreover, authors who investigated the question generally used a rather “loose” sampling grid – *i.e.* less than 2 sampling points/100 m² (Le Coustumer et al., 2007; Dechesne et al., 2004a), 2.5 points/100 m² (Napier et al., 2009), and about 6 points/100 m² (Kluge and Wessolek, 2012) – which may be insufficient to capture the small-scale variability of the concentrations. Additionally, most of these assessments were carried out in

large-scale or centralized facilities, as a result of which the spatial distribution of contaminants in “source-control” SUDS still suffers from a lack of documentation. Only Jones and Davis (2013) achieved a high-resolution characterization of a bioretention cell (about 75 sampling points/100 m²), and thus evidenced noteworthy relationships between metal concentrations, distance from the inlet, and modeled cumulative infiltration. Although several sources of variability have been identified, among which topography, soil heterogeneities, “historical” accumulation, or the presence of technical installations (*e.g.* street lamps or barriers), the present literature does not allow to draw general conclusions regarding the contamination levels and typical size of the polluted areas in the surface soil of SUDS.

Better appraising the pollutants’ accumulation, and resulting distribution, in the surface soil of infiltration devices, would be of great value to (i) provide practical guidance regarding SUDS operation and potential needs for soil maintenance, (ii) optimize the representativeness of further vertical soil samplings, and (iii) understand the mechanisms controlling this distribution and accordingly derive possible improvements of the current modeling tools. For these purposes, the present work aimed to achieve high-resolution cartographies of the soil contamination, in a series of source-control infiltration devices with various hydrologic behaviors and runoff contamination potentials, focusing on heavy metals chronically associated to the urban- and traffic-sourced pollution (Gromaire-Mertz et al., 1999; Huber et al., 2016; Kayhanian et al., 2012).

2. MATERIAL AND METHODS

2.1 Description of the study sites

A series of 10 infiltration-based SUDS, located in the Paris region (France), which had been in operation for at least 10 years except for one of them, were selected for their contrasting watersheds, characteristics, and morphologies (Table 1). Among these are four infiltration basins with different sizes, five infiltration swales, and one grassed filter strip. Photographs of the study sites are supplied as *Supplementary data*. The watershed characteristics, including the use of metallic construction materials and anthropic activities, are indicated in Table 1; the annual average daily traffic in the vicinity of the study sites is also reported when available, as it has been demonstrated to have a significant impact on the metal contents in the topsoil of several roadside swales (Horstmeyer et al., 2016). Watershed delimitation was achieved *via* field inspection, as-

built drawings, and cadastral data supplied by the official French web mapping service *Géoportail*. The effective catchment area of each site – or sampled section in the case of longitudinal swales – was calculated as the weighted sum of the different surfaces composing the watershed, using the runoff coefficients proposed by Ellis et al. (2012). The average annual rainfall in the Paris region over the period 1981-2010 is ~640 mm (source: *Météo France*).

Inflow of water into the infiltration systems consists in either an inlet pipe (*Dourdan1*, *Greffiere*, *Alfortville*, *Dourdan2*, *Vaucresson*), or surface runoff directly flowing from the pavement (*Sausset1*, *Sausset2*, *Chanteraines*, *Vitry*, *Compans*). No dry weather flow was observed during the field campaigns, suggesting an absence (or limited amount) of illicit connections to the storm sewer system. In some devices, superficial outflow is possible in addition to infiltration (*Dourdan1*, *Chanteraines*, *Vitry*, *Compans*). In every study site except *Chanteraines* and *Sausset2*, a dark horizon – whose nature and formation process will be discussed later in this paper – could be distinguished at the soil surface (Figure 1), and its thickness was noticed to be variable in space within the devices (0-30 cm). Most facilities were constructed with flat bottoms and sharp embankments, except *Chanteraines*, *Vitry*, and *Compans*, where the surface soil displayed a 5 to 15% slope perpendicular to the pavement, and *Alfortville*, which had a V-shaped transversal section. Local differences in topography resulted from the history of the devices (*e.g.* vegetation growth or fauna activity), which might cause heterogeneous flow pathways at the soil surface. Since it appeared difficult to make a fine topographical survey, it was rather decided to visualize the water distribution in the upper horizon by performing high-resolution measurements of the soil moisture (*cf.* Section 2.2).



Figure 1. Soil core collected in the study site *Greffiere*: evidence of a dark horizon at the soil surface. Depths are given in cm starting from the surface.

2.2 Sampling and *in-situ* measurements

The field investigations were undertaken between April 2015 and May 2016. Samplings and measurements were carried out along a rectangular grid with $< 3 \text{ m}^2$ meshes whatever the study site. At each node: (i) the vegetation was removed if present, then approximately 50 g of surface soil (upper 2-3 cm) was composited from ≥ 4 subsamples surrounding the sampling location, using a stainless steel trowel which was subsequently cleaned and rinsed twice with ultrapure water; (ii) soil moisture in the first 8 cm was measured (in triplicates, retaining the mean value) with a time-domain reflectometer (*Spectrum Technologies*, FieldScout probe TDR 100); (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. In *Greffiere*, *Chanteraines*, *Vitry*, *Vaucresson*, and *Compans*, additional samples of raw sediment were collected on the nearby road pavement; such deposits could not be found in the immediate vicinity of the other study sites. All samples were conserved in individual high-density polyethylene flasks prior to analyses.

Table 1. Main characteristics of the investigated infiltration facilities

Site name	<i>Dourdan1</i>	<i>Greffiere</i>	<i>Alfortville</i>	<i>Sausset1</i>	<i>Sausset2</i>
Type of device	Retention-infiltration basin	Infiltration basin	Infiltration basin	Infiltration basin	Small swale
Watershed characteristics	Two-lane departmental road (4900 veh/day) + car parking lot adjacent to a waste recycling center	2-ha residential catchment. 40 houses with tile roofs, Zn gutters (40%), metallic rooftops and valleys (including Pb)	Industrial and tertiary activities (concrete roofs) + parking lots and service roads + logistics area (4700 veh/day, mostly trucks)	Car parking lot (210 veh/day)	Car parking lot (210 veh/day)
Effective catchment area	7000 m ²	5000 m ²	20000 m ²	400 m ²	160 m ²
Device area[†]	120 m ²	65 m ²	130 m ²	68 m ²	10 m ²
Inflow of water	Pipe (Ø600 mm) followed by a concrete apron	Pipe (Ø300 mm)	Pipe (Ø800 mm)	Direct runoff, large opening (90 cm)	Direct runoff, small opening (15 cm)
Superficial outlet	Elevated pipe (30 cm above the ground)	None	None	None	None
Operating time	> 20 years	> 20 years	16 years	14 years	14 years
Soil texture	Sandy loam	Sandy clay loam	Clay loam	Silt loam	Silt loam
Vegetation	Spontaneous vegetation	Spontaneous vegetation	Spontaneous vegetation	Shrubs and grass	Herbaceous plants
Soil pH_{water} (N = 3)	7.4 ± 0.1	7.8 ± 0.2	7.7 ± 0.1	7.9 ± 0.2	7.9 ± 0.1

Table 1 (continued). Main characteristics of the investigated infiltration facilities

Site name	<i>Chanteraines</i>	<i>Vitry</i>	<i>Dourdan2</i>	<i>Vaucresson</i>	<i>Compans</i>
Type of device	Swale	Swale	Swale	Swale	Filter strip
Watershed characteristics	Service road to a logistics area (<1500 veh/day, mostly trucks and utility vehicles) + car parking lot	T-junction within an industrial catchment including a tar factory, and a coal-fired power plant (in operation until April 2015)	Roundabout (7300 veh/day) + connected roads + 5 houses with tile roofs	Straight portion of a two-lane departmental road (4000 veh/day)	Highway (22000 veh/day) in the vicinity of an airport and an industrial area. Zinc-coated guardrail between the pavement and the filter strip
Effective catchment area	470 m ²	350 m ²	1600 m ²	400 m ²	250 m ²
Device area [†]	54 m ²	19 m ²	30 m ²	12 m ²	33 m ²
Inflow of water	Direct runoff	Multiple lateral openings	Pipe (Ø500 mm)	Pipe (Ø200 mm)	Direct runoff
Superficial outlet	Elevated pipe (25 cm above the ground)	Elevated pipe (25 cm above the ground)	None	None	Longitudinal ditch
Operating time	10 years	10 years	11 years	> 25 years	3 years
Soil texture	Sandy loam	Loam	Potting soil	Clay loam	Sandy loam
Vegetation	Grass	Herbaceous plants	Grass	Spontaneous vegetation	Grass
Soil pH _{water} (N = 3)	7.9 ± 0.1	8.1 ± 0.1	6.8 ± 0.1	7.6 ± 0.1	7.9 ± 0.1

[†]In the case of swales, this value corresponds to the area of the sampled section.

2.3 Sample preparation and laboratory analyses

In accordance with the international standard on the preservation and pre-treatment of soil samples for the analysis of non-volatile species, the samples were oven-dried at 40°C for 7 days, crushed with a pestle, then passed through a 2-mm nylon sieve (ISO 11464, 2006). Elemental analysis was performed *via* X-ray fluorescence (XRF) on homogenized subsamples (*Thermo Scientific*, Niton™ analyzer XL3t). Among the analytical range of the apparatus, copper, lead, and zinc were retained as tracers of urban- and traffic-derived contamination, because of (i) their well-documented relevance in urban and highway stormwater runoff, (ii) the low detection limits of the analyzer, which enabled to quantify these elements in almost every sample, and (iii) their contrasting physico-chemical properties. Cu has the highest affinity with organic matter (both in dissolved and solid form); Pb has been shown to be predominantly particulate-bound in urban runoff; the greatest part of Zn is generally in dissolved form in roof runoff, but its particulate fraction may be more substantial in traffic area runoff (Huber et al., 2016; Kabata-Pendias, 2011).

All soil concentrations are given in milligrams per kilogram of dry matter. The limits of quantification (LoQ) 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⁻¹ for copper, lead, and zinc, respectively. Four measurements were carried out on different subsamples, then it was checked that the coefficient of variation of the concentrations was < 15%, otherwise two additional measurements were done; the following developments of this paper consider the mean values for each soil sample. The values < LoQ were not taken into account, which only concerned Cu and/or Pb in *Dourdan1*, *Sausset1* and 2, *Chanteraines*, and *Compans*; *Sausset1* was the only site where the proportion of samples < LoQ was higher than 10%. 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 (indicated in Table 1) was determined in three different composite surface samples, in a solution of soil and ultrapure water (volumetric ratio of 1:5) after 1h of equilibration (standard ISO 10390, 2005).

2.4 Blanks

So as to evaluate the contamination potentially induced by the sampling instruments, storing flasks, and soil preparation method, the whole procedure described hereinabove was applied to three supposedly non-contaminated soils. In total, 12 additional samples were collected in a forest, a garden, and a field in the eastern Paris region (corresponding to the “*small agricultural region*” of *Goele et Mmultien* in the French territorial nomenclature). The concentrations measured in these samples correspond to the sum of the geochemical background, the (unknown) initial contamination, and the induced contamination; they were thus used to establish a conservative estimate of the latter.

2.5 Calibration curves of the XRF analyzer

So as to assess the relationships between XRF measurements and total concentrations, chemical analyses were performed on several subsamples collected in *Greffiere*, *Alfortville*, and *Sausset2*, by a laboratory with COFRAC (French Accreditation Committee) certification. These three sites were chosen because they correspond to different ranges of concentrations (as presented below) and different soil textures. The samples were acid-digested (HF + HClO₄), according to the standard NF X31-147 (1996), then Cu and Zn were analyzed by Inductively Coupled Plasma Atomic Emission Spectrometry, and Pb was analyzed by Inductively Coupled Plasma Mass

Spectrometry. In parallel, XRF measurements were carried out on the corresponding samples in 8 replicates, in order to estimate the associated uncertainty from the same number of measurements for each point; the samples below the limits of quantification of the XRF analyzer were not taken into consideration.

2.6 Spatial interpolation and cartography

The different scalar fields were interpolated using universal kriging, with a local first-order trend model to account for the deterministic component of these variables (Chauvet and Galli, 1982). Cartographies of the heavy metal contents, soil moisture, volatile matter and thickness of the upper horizon were generated using a 10 cm × 10 cm interpolation grid over the whole area of the devices. In the subsequent results and discussion, unless otherwise specified, the mention of the SUDS “*surface soil*” will correspond to the sampled layer, *i.e.* the upper 2-3 cm.

2.7 Correlation statistics

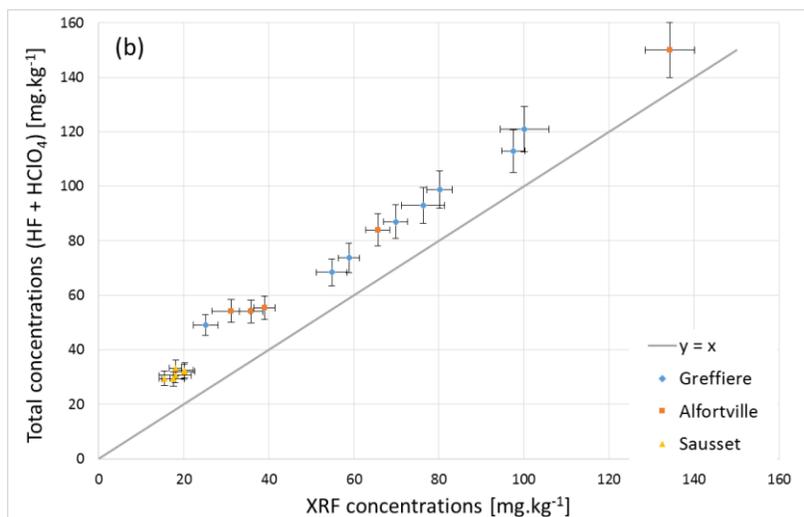
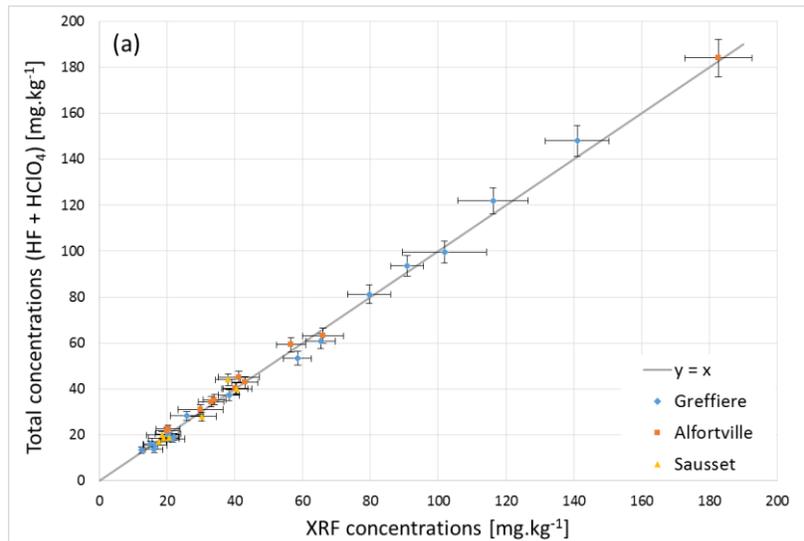
As some variables could not be considered as normally distributed (Shapiro-Wilk test, $p < 0.01$), the correlation between variables was assessed using Spearman’s rank correlation coefficient ρ , and the associated non-parametric test of significance. ρ_{XY} corresponds to the Pearson’s correlation coefficient between the rank values of the two variables X and Y .

3. RESULTS

3.1 Measurement and interpolation uncertainties

For each analyzed sample, the coefficient of variation of the XRF measurements was $< 15\%$, and in most cases $< 10\%$. Figure 2 displays the calibration curves of the XRF analyzer for each metal; the uncertainties associated to the total concentrations were provided by the analytical laboratory. These relationships do not seem to be site-dependent, except for Zn which seems to follow a slightly different trend in *Sausset*. It appears that XRF measurements provide a reliable estimation of the Cu total concentrations, but consistently underestimate the Pb concentrations (average difference of 16 mg.kg⁻¹), and the Zn concentrations below 250 mg.kg⁻¹ in *Greffiere* and *Alfortville* (21 mg.kg⁻¹). The results given in the following sections correspond to XRF concentrations without correction.

The lowest mean concentrations measured in the blank samples were 28 mg.kg⁻¹ for Zn, and < LoQ for Pb and Cu. Considering the maximum values of the LoQ given in Section 2.3, and accounting for the correction of XRF measurements, it follows that Geochemical background + Initial contamination + Induced contamination ≤ 20, 26, and 49 mg.kg⁻¹ for Cu, Pb, and Zn, respectively. Background concentrations were estimated from a national database on trace metals concentrations in non-contaminated soils (Duigou and Baize, 2010). In *Goele et Mmultien*, the Cu, Pb, and Zn concentrations are 15.7 ± 5.3 mg.kg⁻¹, 19.3 ± 7.4 mg.kg⁻¹, and 56.8 ± 13.2 mg.kg⁻¹, respectively (*N* = 226), which suggests that the contamination deriving from the experimental protocol is negligible. In every study site, the maximum kriging standard deviation – which quantifies the potential interpolation error as a function of the distance from the data locations – represents less than 13% of the maximum concentrations.



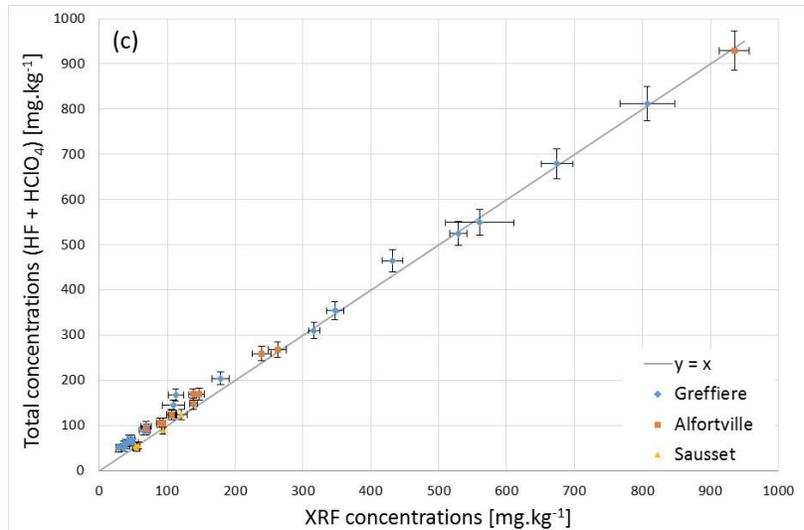


Figure 2. Relationships between the XRF measurements ($N = 8$ for each point) and the total concentration analyses following acid digestion, for (a) Cu, (b) Pb, and (c) Zn. Error bars represent the 95% confidence interval of each value.

3.2 Contamination patterns

The metal concentrations may exhibit an important dispersion within a device (the relative standard deviation varies between 26 and 106%, and is generally higher for Zn), however their spatial distribution displays a typical structure in the surface soil. The highest concentrations are always detected in the area surrounding the water inflow point/zone, from which they tend to decrease with increasing distance. Figure 3 illustrates this point in the specific case of Zn, with four sites that synopsise every encountered configuration. The swale with diffuse runoff inflow *Vitry* (3.a) is analogous to *Chanteraines* and *Compans*; the swale with one point-source inflow *Dourdan2* (3.b) is comparable to *Sausset1* and *Vaucresson*, with a longer contaminated zone; the contamination pattern observed in *Sausset2* (3.c) is similar to *Greffiere* and *Dourdan1*. *Alfortville* (3.d) has an atypical behavior that will be discussed further. In general, the decrease in metal concentrations is rather sharp, as the maximum lateral gradient is found close to the facilities' inlet, and amounts to respectively 860, 540, and 170 $\text{mg.kg}^{-1}.\text{m}^{-1}$ in the first three examples of Figure 3. Hence, in small-scale SUDS (*Sausset1*, *Sausset2*, *Chanteraines*, *Vitry*, *Vaucresson*, and *Compans*), surface concentrations are divided by a factor of 2 within less than 1 m from the inflow area; as a result, metal accumulation appears to be horizontally restricted to a limited region of the devices. In almost every investigated site, the three metals have similar spatial distributions (as illustrated in Figure 4.a-c for the infiltration basin *Dourdan1*).

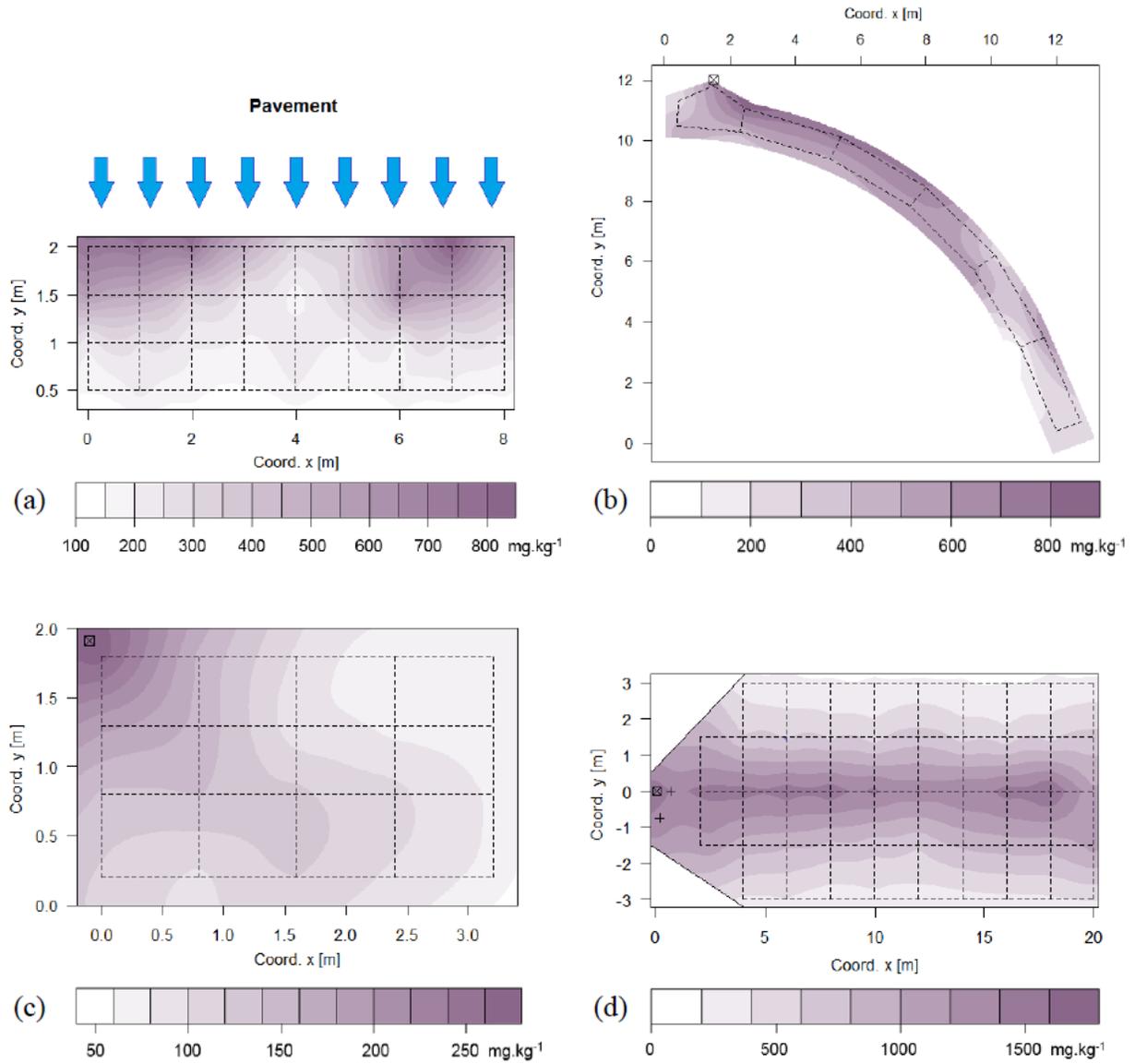


Figure 3. Spatial distribution of zinc [mg.kg^{-1}] in the surface soil of the study sites (a) *Vitry*, (b) *Dourdan2*, (c) *Sausset2*, and (d) *Alfortville*. Samples were taken at each node of the dotted grid and at the points +. The symbol \boxtimes indicates (when appropriate) the location of the inflow point.

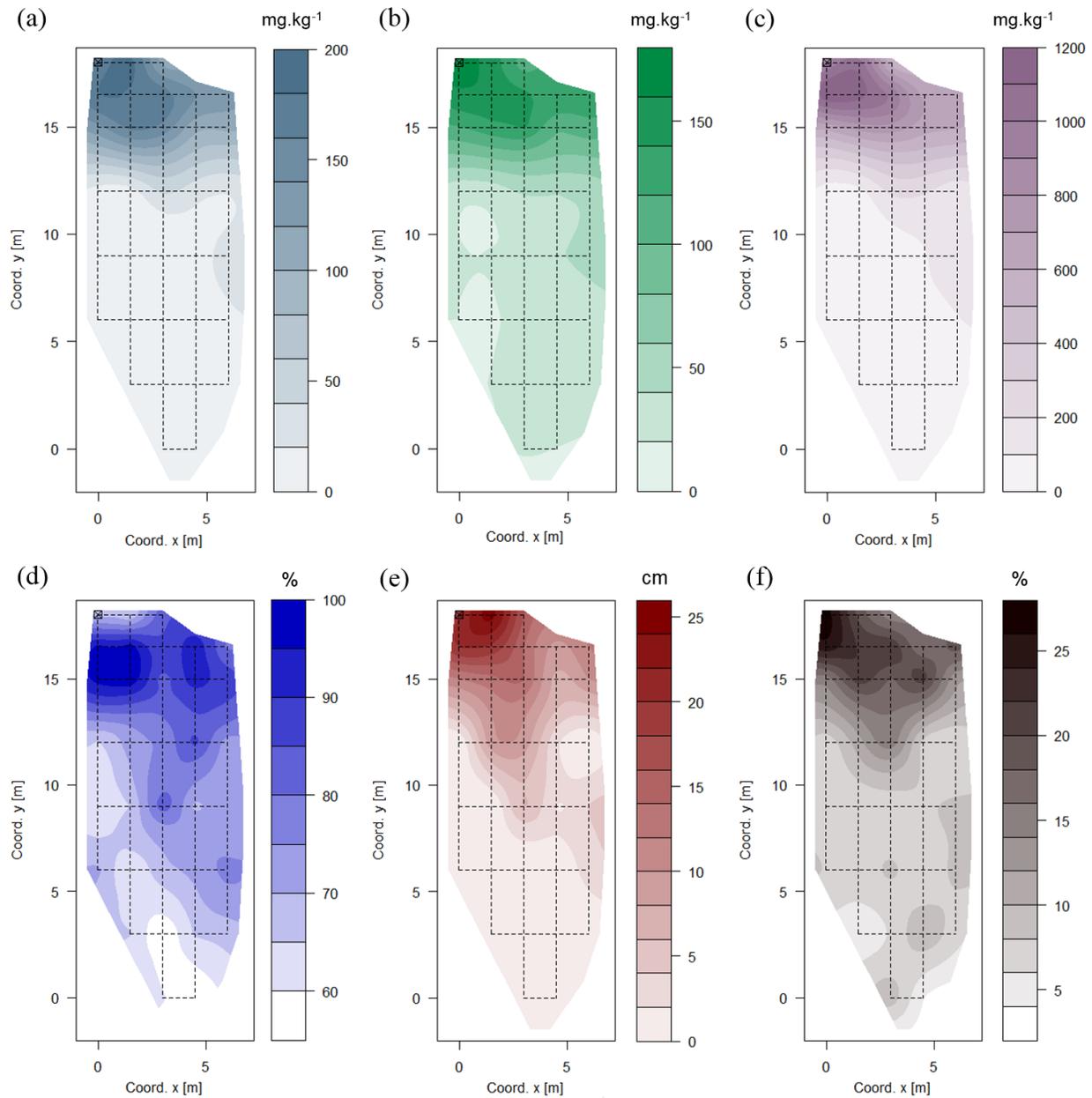


Figure 4. Complete results for the study site *Dourdan I*. Cartographies of the concentrations of (a) lead, (b) copper, and (c) zinc in the surface soil [mg.kg^{-1}], (d) the relative water content [%], (e) the thickness of the upper horizon [cm], and (f) the volatile matter content in the surface soil [%]. Samplings and measurements were carried out at each node of the dotted grid. The symbol \boxtimes indicates the location of the inflow point.

3.3 Heavy metal enrichment in soil

Insofar as the site-specific initial concentrations are hardly ever measured before the devices start operating, they have to be estimated by another method in order to accurately assess the metal enrichment due to runoff infiltration. The common assumption that nearby sampling points, supposedly “uninfluenced” by infiltration, may represent local background contents (Lind and Karro, 1995), is likely to be unsuitable for systems where the upper horizon has been either excavated (to provide a sufficient storage volume) or amended with topsoil (to facilitate vegetation growth) 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, 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 in each SUDS. Assuming that other sources of metals (such as atmospheric deposition) induce a homogeneous increase in the surface concentrations, the average value in the latter zone was considered as a “reference” concentration – which is likely not to be the same as the initial concentration. Figures 5.a to 5.c illustrate the significant increase in the metal contents with respect to the reference concentrations in all sites ($p < 0.05$), except for lead in *Sausset1*; this could be due to the measurements uncertainties associated to low concentrations, even in the most contaminated zone. Among the ten study sites, the infiltration basin *Alfortville* globally appeared as the most polluted device, however highest enrichment ratios were found in *Greffiere*, *Compans*, and *Dourdan1* (7-21 for Pb, 6-10 for Cu, and 8-28 for Zn); in the latter site, this was reinforced by low background levels. In comparison to the other devices, *Vitry* and *Vaucresson* exhibited high Pb concentrations near the inflow area, but lesser Cu and Zn contamination than average. Although metal accumulation was also evident in *Sausset1*, *Sausset2*, and *Chanteraines*, both absolute contents and enrichment ratios remained low. As regards proportions between metals, the ratio Zn/Cu in the most contaminated zone was fairly similar in *Alfortville*, *Sausset1* and *2*, *Dourdan2*, *Vaucresson*, and *Compans* (3.7-4.3); it was higher in *Greffiere*, *Vitry*, and *Dourdan1* (5.2-6.7), and lower in *Chanteraines* (2.3). Highest and lowest Cu/Pb ratios were respectively found in *Compans* (4.3), and in *Vitry* and *Vaucresson* (< 0.5).

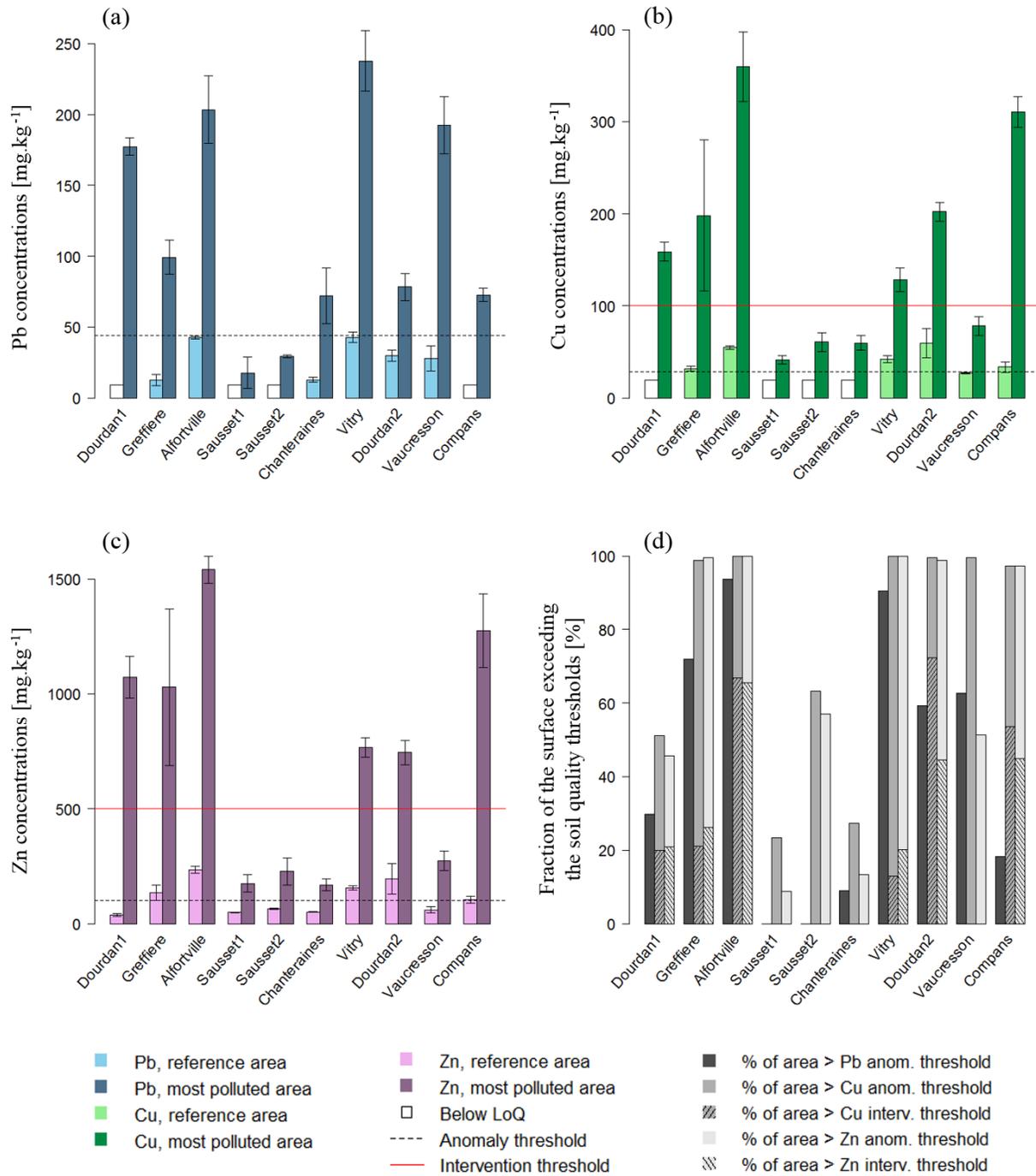


Figure 5. Mean surface concentrations (\pm standard deviation) of (a) lead, (b) copper, and (c) zinc [mg.kg^{-1}] measured in the most and the least polluted areas of each investigated device; comparison with the 9th decile of nationwide analyses in agricultural soils (Baize et al., 2007, dotted lines) and the Canadian intervention thresholds (Fouchécourt et al., 2005, solid lines). (d) Proportion of the SUDS area where the surface concentrations exceeded the anomaly (solid bars) and intervention (hatched bars) thresholds for each metal.

4. DISCUSSION

4.1 Comparison with soil quality criteria

Long-term pollutant buildup in the surface soil may result in two distinct problems: (i) the nature and levels of contamination might induce health and environmental hazards, which has to be taken into consideration in case SUDS are implemented within multi-functional spaces (Woods Ballard et al., 2015); (ii) the gradual exhaustion of the soil's sorption capacities is likely to facilitate downward transport, and increase the risks of groundwater contamination. In this context, stating whether a soil has to be considered "contaminated" or not, and defining appropriate measures, requires the introduction of generic soil quality criteria (Dechesne et al., 2004b). "Anomalous" soil concentrations, indicating a probable exogenous contamination, may be characterized with respect to the geochemical background. Conversely, "intervention" thresholds, whose exceedance entails land use restriction or soil excavation, generally originate from country-specific *regulatory guidelines*. In the absence of such standards in the French regulations – except in the specific case of sewage sludge spreading over agricultural lands – criteria from other countries will be presented and discussed. These intervention thresholds are usually defined with regards to a given soil use: among the available values, Table 2 displays those which may apply to either "recreational activities" or "groundwater protection".

The variability between thresholds originates from (i) the definition of the exposure scenario (*e.g.* dermal contact, ingestion of soil, or inhalation of dust), integrating chronic and/or acute harmful effects, and (ii) the consideration of ecotoxicological effects in addition to human health hazards in several countries. For example, the Dutch and the Swedish intervention values account for both a maximum permissible risk for humans, and an ecotoxicologically-based value (above which 50% of the tested species encounter adverse effects due to soil contamination). In the present study, the criteria retained as anomaly and intervention thresholds correspond respectively to (i) the 9th decile of a series of > 11000 nationwide heavy metals analyses in agricultural soils (Baize et al., 2007), and (ii) the "strictest" values among the presented guidelines, *i.e.* the Canadian standards.

In every study site, "anomalous" concentrations are observable in all or part of the surface soil (Figure 5), but different features can be distinguished. On the one hand, the soil of *Sausset1*, *Sausset2*, and *Chanteraines* may to a large extent be considered "uncontaminated". On the other

hand, intervention thresholds are locally exceeded in 6 devices for Zn and Cu, but never for Pb. Even considering the strictest thresholds, the corresponding area amounts to less than 25% of the whole surface in the basins *Dourdan1* and *Greffiere*, and in the swale *Vitry* (Figure 5.d). This proportion is higher in *Dourdan2*, *Compans* and *Alfortville* (45-72%). Replacing XRF measurements by corrected values according to the relationships displayed on Figure 2 increases the fraction of the SUDS area where Pb concentrations exceed the anomaly thresholds, but has no impact on the exceedance of the intervention thresholds.

Table 2. Examples of country-specific anomaly and intervention thresholds (expressed as total concentrations, in mg.kg⁻¹). The former usually entail further investigation when exceeded, whereas the latter require specific measures (*e.g.* soil excavation).

Country	Reference	Associated land use	Cu	Pb	Zn
Anomaly thresholds					
France	Baize et al., 2007	9 th decile of > 11000 heavy metals analyses in French agricultural soils	28	44	102
Canada	Fouchécourt et al., 2005	National background concentrations	40	50	110
The Netherlands	NMHSPE, 2000	National background concentrations	36 [†]	85 [†]	140 [†]
Switzerland	OSol, 1998	“Indicative values”	40	50	150
Intervention thresholds					
Canada	Fouchécourt et al., 2005	Residential, recreational and institutional sites	100	500	500
Sweden	Swedish EPA, 1997	“Sensitive use” + groundwater extraction occurring in the vicinity of the site	200	300	700
The Netherlands	NMHSPE, 2000	Serious impairment of the soil’s functional properties for humans, plant and animal life	190 [†]	530 [†]	720 [†]
Belgium	Walloon Parliament, 2009	Recreational and commercial sites	290	700	710
Denmark	Danish EPA, 2002	Land use includes “very sensitive purposes” (<i>e.g.</i> private gardens or day-care centers)	500	400	1000
Switzerland	OSol, 1998	Private gardens, playgrounds	1000	1000	2000

[†]These values correspond to a “standard” soil, and have to be corrected according to the volatile matter and clay content of the investigated soil.

4.2 Factors governing the metals’ distribution

The contamination patterns are interpretable as the signature of (i) the amount of infiltration fluxes, (ii) the flow pathways at the soil surface, and (iii) the importance of settling processes. In the Paris region, the annual rainfall distribution is dominated by small events: for example, during

the period 1993-2008, 90% of the rainfall events were ≤ 3 mm, and 98% were ≤ 10 mm; these events contributed 40 and 75% of the total rainfall volume, respectively (see *Supplementary data*). For these frequent events, which are responsible for the major part of the contaminant fluxes in runoff, water is likely to infiltrate before spreading over the entire systems, all of which have been designed for a ≥ 10 -year storm. In other words, the actual drainage/infiltration area ratio is likely to be higher than expected during small rainfall events. As water is the carrier medium of solutes and particles, the contaminant fluxes would be essentially concentrated near the inflow area, while the fraction of the SUDS surface opposite to this zone would be seldom reached by neither particulate-bound nor dissolved metals. Besides, low flow rates are likely to favor settling processes close to the water inlet. Whatever the site, there was no evidence of soil or particle re-suspension due to infrequent storms, since the area surrounding the inflow zone did not show any trace of erosion. Insofar as the investigated devices are source-control systems, the water velocity is not likely to be as high as in centralized facilities, where erosion does occur (Cannavo et al., 2010).

In the study site *Vitry* (Figure 3.a), the decrease in the Zn concentrations observed from $x = 3$ to $x = 5$ m corresponds to a local difference in topography (~ 10 cm) which deviates runoff right or left from this mound. *Alfortville* (Figure 3.d) has a peculiar behavior, since a 30-cm-thick clay layer was found at 30 cm depth in the major part of the device, thus preventing water infiltration near the inlet pipe: all along the V-shaped basin, the lowest zone (located at $y = 0$) displayed nearly uniform and highest soil moisture (see *Supplementary data*), and was found to be quite evenly contaminated. The similarity between the contamination patterns of the three metals, in spite of potentially different speciations in urban runoff, also suggests that hydraulics is the factor responsible for most variability of the contaminant concentrations in the surface soil. This conclusion is corroborated by the spatial distribution of the soil moisture after a rain event (Figure 4.d), which reveals the flow pathways in the device, and displays a pattern similar to the metals' one. However, such a soil moisture distribution could not be observed in every study site, since it is strongly dependent on the weather conditions before the measurements were carried out. In the case of *Dourdan1*, the field campaign was undertaken few hours after a 11.5 mm rainfall event (source: *Météo France*). Other examples are provided as *Supplementary data*.

4.3 Correlations with soil organic matter

The nature and formation processes of the variably thick dark horizon which has been observed at the surface of most devices are not precisely identified. In the larger and older infiltration basins (*Dourdan1*, *Greffiere*, *Alfortville*), where native vegetation is allowed to grow with a limited number of cleaning operations, its thickness has been observed to be correlated to the volatile matter content at the soil surface (Figure 4.e-f): $\rho = 0.87, 0.78, \text{ and } 0.64$, for the three above-mentioned sites, respectively, which is statistically significant with $p < 10^{-5}$ in each case. Interestingly, similar correlations were obtained with the heavy metal contents: $\rho > 0.74$ whatever the metal ($p < 10^{-6}$ likewise).

This layer may be partially formed of deposited and/or filtered suspended solids (*e.g.* originating from tire or brake pad abrasion) with high heavy metal contents (Huber et al., 2016). Its variable thickness would thus be linked to the deposition rate of particulate matter within the infiltration systems. However, the sediment samples collected in the vicinity of the study sites (*cf.* Section 2.2) were not as organic-rich as the soil of most devices: their volatile matter content ranged between 7 and 14%, while the maximum values measured in *Dourdan1*, *Greffiere*, and *Alfortville* were higher than 27%. This consideration suggests that the upper layer cannot be only formed of deposited particles. It may also be composed of fresh humus originating from plant growth and decay, which are favored in the most frequently flooded areas of the systems (as confirmed by visual inspection of the sites), thus enhancing soil organic matter production. The correlations with metal concentrations might therefore be explained by hydraulics (*i.e.* differences in cumulative infiltration fluxes), which have been demonstrated to govern a large part of the contamination patterns, and by the ability of soil organic matter (and specifically humic substances) to enhance metal retention due to *chemisorption*, *i.e.* short-range interactions between metals and surface reactive groups acting as ligands (Bradl, 2004). The present results do not enable to state which of these two mechanisms is predominant in the formation of the upper horizon in each site. In any case, this finding is likely to have additional implications regarding the long-term evolution of the soil's hydrodynamic properties (*e.g.* due to clogging), and consequently the hydraulic performance of the devices (Cannavo et al., 2010; El-Mufleh et al., 2014). The previous observations hold to a lesser extent in the infiltration basin *Sausset1*, but no such trend was detected in smaller swales/filter strips, where the upper horizon was homogeneously amended with planting soil/filter media during or after the construction works, and the vegetation is regularly maintained.

4.4 Sources of contaminants

Metal concentrations in SUDS soil result from the combination of three distinct elements: (i) the hydraulics of the devices – as mentioned earlier, (ii) the loads originating from the watersheds, and (iii) the importance of the retention mechanisms in soil. Hence, the inter-site variability of concentrations has a catchment-dependent and a soil-dependent components. As the sorption properties of the different soils have not been characterized in the present study, only the point (ii) will be discussed. Pb contamination appears to be related to either industrial activities in the vicinity of the site (*Vitry, Alfortville*) or long operating time of the device (*Vaucresson, Dourdan1*). The former element is likely to induce a supplementary contamination due to atmospheric deposition and/or the transportation of raw materials by trucks (Brown and Peake, 2006), whereas the latter probably corresponds to the signature of leaded gasoline, whose use was banned in 2000 in France. In the residential catchment of *Greffiere*, where car traffic is low, lead rooftops and valleys constitute an additional source of Pb in stormwater runoff (Petrucci et al., 2014). Aside from roofing and siding materials (when present in the drainage area), the major sources of Cu and Zn have been shown to be brake and tire wear, respectively (Davis et al., 2001; Petrucci et al., 2014). Heavily trafficked areas such as *Compans* therefore exhibit a significant potential for runoff contamination, so does *Dourdan2* to a lesser extent. In addition to traffic-related sources, contamination in *Dourdan1* may originate from metal storage in the waste recycling center adjacent to the site, as suggested by higher Zn/Cu ratios than in the other traffic areas. Likewise in *Greffiere*, high Zn/Cu may indicate the contribution of zinc roofs and gutters as a significant source of Zn. Furthermore, in *Dourdan1, Greffiere, and Alfortville*, the extended drainage area increases the pollutant loadings received per unit area of soil, as well as the probability for unidentified point sources of metals within the watershed. Conversely, the source-control SUDS *Sausset1, Sausset2, and Chanteraines* display low to moderate contamination potentials, because of light traffic on their catchments, and small drainage/infiltration area ratios (6, 16, and 9, respectively), as a result of which little accumulation of metals is visible in these devices.

4.5 Comparison with previous assessments

Table 3 presents the range of Cu, Pb, and Zn concentrations measured in the surface soil of similar infiltration-based SUDS. The low contamination levels found by Achleitner et al. (2007), Ingvertsen et al. (2012), and Jones and Davis (2013) in small-scale devices are comparable to the metal contents in *Sausset* and *Chanteraines*. Conversely, the infiltration basins investigated by

Dechesne et al. (2004a), Barraud et al. (2005), and Napier et al. (2009) display concentrations similar to *Dourdan1*, *Greffiere*, *Alfortville*, and *Dourdan2*. The signature of Pb in “older” roadside filter strips is clearly visible (Dierkes and Geiger, 1999; Kluge and Wessolek, 2012; Norrström and Jacks, 1998), which is akin to *Vaucresson* but not to *Compans*, because the latter device is more recent.

As regards the spatial distribution of heavy metals, the existing studies had led to contrasting conclusions. Similar to the present findings, several authors detected highest contamination near the inlet pipe (Le Coustumer et al., 2007; El-Mufleh et al., 2014; Napier et al., 2009) or inflow zone (Norrström and Jacks, 1998), with an overall decrease in concentrations along the flow pathway (Jones and Davis, 2013). Conversely, the most contaminated location of a mid-scale infiltration basin was found to be the lowest point (Dechesne et al., 2004a), and the highest concentrations in several filter strips were found at 0.5 to 1 m from the pavement, despite the presence of a permeable shoulder (Boivin et al., 2008; Dierkes and Geiger, 1999). This may be due to the slope of the facility (unspecified in these articles), the infiltration rate, and/or soil clogging near the inflow area.

Table 3. Range of metal concentrations [$\text{mg}\cdot\text{kg}^{-1}$] found in the surface soil of similar devices in previous investigations.

Reference	Device and catchment	Age	Cu	Pb	Zn
Dechesne et al. (2004a)	Infiltration basin, truck parking lot	14 years	25-176	68-223	458-1566
Barraud et al. (2005)	Infiltration basin, mixed urban land uses	12 years	87-256	78-191	861-2605
Napier et al. (2009)	Roadside infiltration basins	7 years	8-198	18-107	63-1050
Ingvertsen et al. (2012)	Swales, light urban area or car parking lot	6-16 years	20-100	35-120	70-400
Achleitner et al. (2007)	Vegetated swale, parking lot	2-10 years	26-131	28-196	66-229
Jones and Davis (2013)	Bioretention cell, parking lot	4 years	16-50	28-70	120-250
Norrström and Jacks (1998)	Roadside filter strip, highway	29 years	19-57	205-542	96-140
Dierkes and Geiger (1999)	Roadside filter strip, highway	16 years	27-413	65-239	155-527
Kluge and Wessolek (2012)	Roadside filter strip, highway	> 50 years	3-565	11-426	8-804

4.6 Operational and scientific outcomes

Soil sampling. The foregoing analysis highlights the uttermost importance of a careful and rational selection of the sampling points for experimental assessments of SUDS, given the substantial dispersion and variability of surface concentrations. Identifying areas with

homogeneous contamination levels at the surface constitutes an objective criterion to collect composite core samples and guarantee their optimal representativeness (ISO 10381-5, 2005). In this respect, X-ray fluorescence has been proven to be an interesting technique, as it provides a cost-effective and rapid way to analyze soil samples with a controlled accuracy, extends the number of analyses that can be carried out during the preliminary investigation, and thus enables to achieve a finer cartography of heavy metals. In a different context, XRF measurements have been extensively used to produce maps of soil contamination in urban allotment gardens: the authors concluded that this method was useful for better soil management as well (Bechet et al., 2016).

SUDS maintenance. The present observations suggest possible improvements of maintenance operations to be undertaken in order to ensure a sustainable functioning of these devices. Traditionally, SUDS soil has been considered as part of technical infrastructures serving as treatment facilities and, as such, designed to accumulate contaminants until it is entirely disposed as waste. As discussed previously, the localized enrichment of metals restricts the area requiring soil scrapping or renewal, systematically situated around the water inlet. Hence, the needs for intervention may be assessed through a simplified two-step procedure: (i) compositing and analyzing one bulk sample from several locations immediately surrounding the inflow zone; (ii) in case concentrations exceed intervention thresholds, collecting additional samples at various distances along transects starting from this zone. This would enable to capture the decrease of concentrations, and thus determine the extent of the contaminated area. Besides, visual observations of the water flow paths may also be helpful to identify *a priori* “hot spots” of concentrations and optimize the sampling strategy. In catchments with high contamination potential such as *Compans*, this procedure should be undertaken at least every 3 years, since this duration has been proven sufficient to reach Zn and Cu concentrations requiring surface soil remediation.

SUDS design. The hydraulic functioning of infiltration systems could be optimized towards an easier maintenance, with an excavated area near the water inlet designed to intercept a known fraction of the annual pluviometry. This would concentrate most pollutant fluxes in a clearly identified zone, where intervention could be carried out with a defined frequency according to the migration rate of contaminants. Accessibility to this area should be limited so as to avoid any public health risk; conversely, as important storage volumes are basically required for infrequent

events, the corresponding space, which is likely to display lower surface contamination, appears suitable for multi-functional purposes.

Furthermore, in the current practices on SUDS design and planning, small-scale “treatment trains” consisting of consecutive facilities are increasingly implemented to combine stormwater management with pollutant removal (Woods Ballard et al., 2015). In these chains of devices, the first permeable surface reached by stormwater runoff is likely to intercept the greatest part of the contaminant fluxes – especially in geographical areas whose rainfall distribution is dominated by small events. This might prevent a proper and effective functioning of a treatment device (*e.g.* a bioretention system) in case it is not located ahead of the “treatment train”.

SUDS modeling. Presently, most attempts to model the fate and transport of contaminants in the soil/filter media of SUDS have adopted a one-dimensional framework, assuming that the water fluxes spread homogeneously over the soil surface whatever the rainfall intensity, and that the spatial distribution of contaminants only varies with depth (Li and Davis, 2008; Quinn and Dussailant, 2014). These two hypotheses have been proven to be inaccurate for frequent rainfall events, at least in the case of infiltration systems without permanent ponding. The modeling framework should therefore be adapted in further studies, so as to take into account the heterogeneous distribution of infiltration fluxes as an upper boundary condition of the model.

5. CONCLUSIONS AND RESEARCH NEEDS

This study has investigated the spatial distribution of three characteristically urban-sourced heavy metals in the surface soil of ten infiltration-based SUDS with contrasting designs and catchments. Overall, more than 450 soil samples have been collected and analyzed *via* X-ray fluorescence spectrometry, thus enabling to get a precise visualization of the small-scale variability of concentrations in each device. Cartographies of heavy metals evidence that most accumulation typically occurs near the inflow area, with a sharp decrease in concentrations with increasing distance from this zone. Lead, copper, and zinc behave the same in most facilities, while their particulate fraction in urban runoff has been shown to be quite different. These observations emphasize the role of hydraulics, and more specifically spatial differences in the cumulative infiltration fluxes, as a factor controlling the horizontal extent of soil contamination in SUDS. In the larger infiltration basins, the most polluted zone has been observed to have the highest volatile

matter content, with the maximum thickness of the dark upper horizon, which is likely to enhance the surface soil's retention capacities in the vicinity of the water inlet.

Focusing on surface contamination, the present results have demonstrated that most infiltration – and consequently micropollutant – fluxes are concentrated in a relatively small area of the infiltration systems. This element may have noteworthy consequences on vertical contaminant transfer: besides limiting the residence time of degradable hydrophilic pollutants in the biologically active zone, it could locally favor downward migration of solute species following the exhaustion of the surface soil's sorption capacities – all the more so as deeper horizons are likely to have different compositions and potentially lower sorption potential. With this in mind, the vertical extent of contamination should be further investigated in similar infiltration devices, and if urban pollutants are proven to have reached a significant depth under surface “hot spots”, then it may be valuable to consider other inflow modes than point-source inlets. Such systems would be aimed to divide the water fluxes and facilitate water spreading over a larger surface, for instance *via* several small pipes, which is a common design for on-site sanitation facilities. Such a practice would probably reduce the risks of vertical transport, but undoubtedly extend the contaminated area in the surface soil. Furthermore, it should also be explored whether the spatial distribution of metals at lower depths follows similar trends, or whether several sources of variability at the soil surface do not impact vertical transport.

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