



HAL
open science

In-depth characterization of sediment contamination in stormwater infiltration basins

Damien Tedoldi, Julien Couvidat, Mathieu Gautier, Qiufang Zhan, Thierry Winiarski, Gislain Lipeme Kouyi, Cécile Delolme, Vincent Chatain

► **To cite this version:**

Damien Tedoldi, Julien Couvidat, Mathieu Gautier, Qiufang Zhan, Thierry Winiarski, et al.. In-depth characterization of sediment contamination in stormwater infiltration basins. *Blue-Green Systems*, 2024, 6 (1), pp.1-19. 10.2166/bgs.2023.039 . hal-04440355

HAL Id: hal-04440355

<https://hal.science/hal-04440355v1>

Submitted on 4 Nov 2024







HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

In-depth characterization of sediment contamination in stormwater infiltration basins

Damien Tedoldi ^a, Julien Couvidat ^a, Mathieu Gautier ^a, Qiufang Zhan^a, Thierry Winiarski ^b, Gislain Lipeme Kouyi ^a, Cécile Delolme ^b and Vincent Chatain ^{a,*}

^a INSA Lyon, DEEP, UR7429, 69621 Villeurbanne, France

^b Université Claude Bernard Lyon 1, CNRS, ENTPE, UMR 5023 LEHNA, F-69622, Villeurbanne, France

*Corresponding author. E-mail: vincent.chatain@insa-lyon.fr

 DT, 0000-0001-7125-9544; JC, 0000-0003-4077-6973; MG, 0000-0003-0054-6532; TW, 0000-0001-9947-5038; GL, 0000-0002-0513-9496; CD, 0000-0001-9054-3727; VC, 0000-0003-2349-873X

ABSTRACT

Managing stormwater in infiltration-based systems enables the interception of runoff suspended solids. Accumulated particles form a layer of *stormwater sediments*, the contamination of which presents a critical challenge for maintenance operations but is still insufficiently understood. This study therefore aims to characterize the main contamination patterns of stormwater sediments. Sediments were sampled from 18 infiltration basins encompassing a diversity of catchments. Eighty-five substances, including metals and six families of organic micropollutants, were targeted, almost all of which were consistently quantified. A significant accumulation relative to baseline levels was ascertained in all sites. The geochemical signature of stormwater sediments was relatively close to that of road dust, but different from continental and marine sediments, revealing the contribution of urban-specific sources of contaminants. Common emission and transfer dynamics were inferred from strong correlations between metals; same observations were made for alkylphenols and the group formed by polycyclic aromatic hydrocarbons, polychlorinated biphenyls, dioxins, and furans. The predominant land use was generally insufficient to explain the inter-site variability of sediment contamination, which could still be understood from an in-depth study of each catchment and the identification of site-specific emission sources. These results constitute a milestone toward stormwater sediment reuse as a resource.

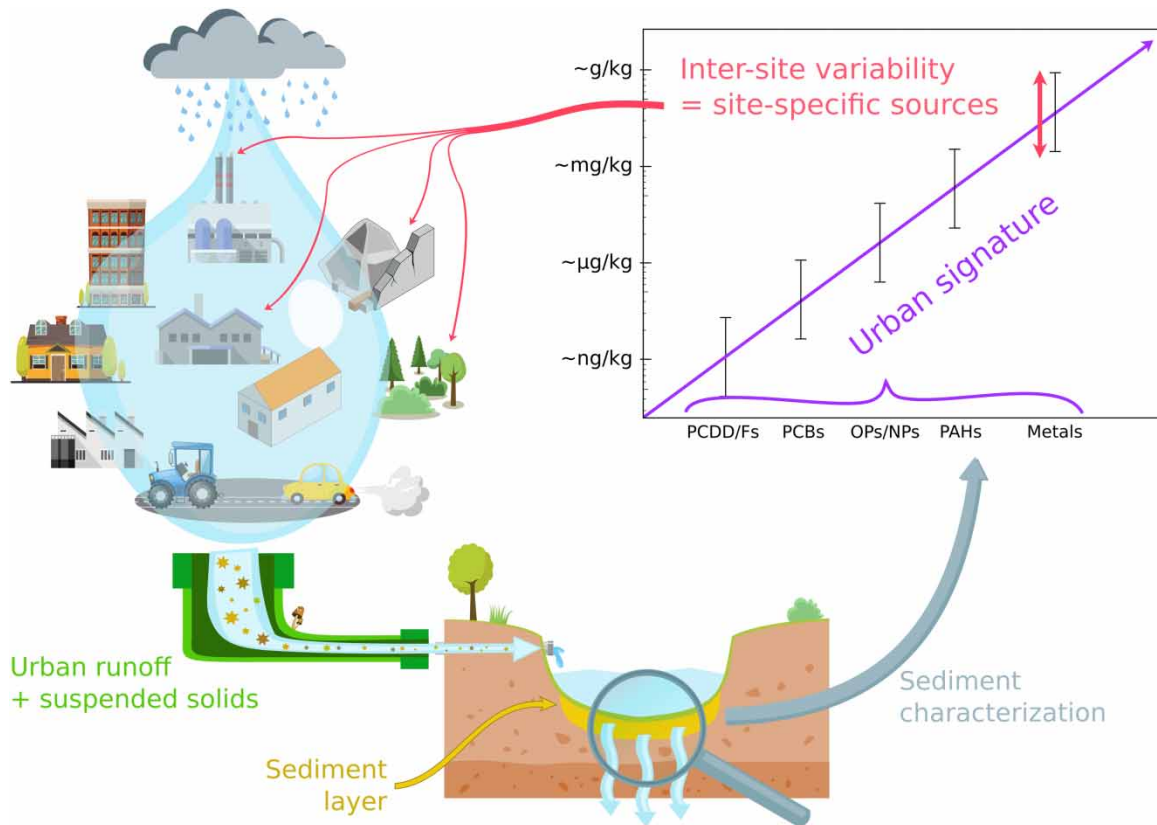
Key words: infiltration-based systems, micropollutants, runoff, sediment, sustainable drainage systems (SuDS), urban catchment

HIGHLIGHTS

- Legacy and emerging micropollutants were analyzed in sediments from 18 infiltration basins.
- Almost all targeted substances exceed the baseline levels in local soils.
- Stormwater sediments have an urban geochemical signature different from other sediments.
- The dominant land use of the catchment is not a good predictor of sediment composition.
- High geochemical anomalies may nevertheless be explained by specific emission sources.

This is an Open Access article distributed under the terms of the Creative Commons Attribution Licence (CC BY 4.0), which permits copying, adaptation and redistribution, provided the original work is properly cited (<http://creativecommons.org/licenses/by/4.0/>).

GRAPHICAL ABSTRACT



1. INTRODUCTION

According to the United Nations, the proportion of the world population living in urban areas has risen from 30 to 50% between 1950 and 2008, and it is expected to exceed 65% by 2050 (United Nations 2019). Urban sprawl leads to land artificialization and generally to soil surface sealing, which in turn induces strong disturbances on the natural hydrological cycle. Furthermore, urban development and human activities multiply both diffuse and point sources of contaminants of various kinds, so that runoff conveys increasing amounts of urban- and traffic-derived substances (Müller *et al.* 2020; Wicke *et al.* 2021). It is notably loaded with suspended particles originating from multiple sources: combustion, road traffic (gas exhaust, brakes, tires and asphalt wear, vehicle corrosion), industrial emissions, and also from animal or plant waste and soil or building erosion (Loganathan *et al.* 2013; Müller *et al.* 2020). These particles act as vectors for a wide range of metals, metalloids, organic micropollutants, and microorganisms including pathogens (Marti *et al.* 2017): they are thus responsible for a significant part of the urban contaminant loads into surface waters during wet weather (Hannouche *et al.* 2017). Urban stormwater discharges are now recognized as 'a barrier to maintaining high quality water resources' (Saifur & Gardner 2021) and a major environmental and health concern (Pal *et al.* 2014).

To cope with the above problems, sustainable drainage systems (SuDS) have benefited from growing interest worldwide and have been increasingly implemented in urban areas. These facilities collect runoff water, store it temporarily to alleviate the sewer system during storm events, and help re-establish a more natural water cycle through infiltration and/or evapotranspiration. Beyond these hydraulic and hydrologic improvements, SuDS also offer multiple co-benefits, especially for pollution mitigation. In particular, soil- or substrate-based systems enable most suspended solids to be intercepted by settling and filtration (Flanagan *et al.* 2019). Accumulated particles are thus progressively incorporated to the surface horizon (Mitchell Ayers & Kangas 2018), until in some cases they form a layer of *stormwater sediments* – the latter being presently defined as exogenous particles brought by runoff and apparently distinct from the underlying soil (El-Mufleh *et al.* 2014). The sedimentation rate in various stormwater basins has been estimated between 1

and 4 cm/year (Yousef *et al.* 1994; Ahilan *et al.* 2019) although this value likely depends on SuDS design and catchment type.

Over the long term, sediment accumulation may compromise the proper functioning of SuDS by clogging the infiltration area (which gradually decreases the surface permeability and limits the recharge of groundwater aquifers) or by filling up the storage volume (Taguchi *et al.* 2020). Maintenance operations are therefore periodically necessary, mostly consisting of sediment dredging, which generates large albeit discontinuous amounts of solid materials to be subsequently dealt with. In France, a study estimated the annual amount of sediments dredged from road ditches and road/highway stormwater ponds at 5.7 Mt nationwide, i.e., ~ 10 t/year/km² (Ruban *et al.* 2003). At a regional scale, the conurbation of the *Greater Lyon* (540 km²) has been implementing a sustainable stormwater management policy for several decades, so that there are currently >300 infiltration and retention basins on its territory: on average, 670 t/year of sediments have been dredged from all of the territory's stormwater facilities (including pretreatment devices) between 2012 and 2020 (Couvidat *et al.* 2021). Although regulations do exist at the national and international levels for other kinds of dredged sediments, e.g., from rivers or harbors (OSPAR or Barcelona conventions), the comparatively recent emergence of stormwater sediment-related issues, as well as the relative lack of knowledge about these materials, have led to non-regulatory and mostly empirical management practices. *De facto*, as soon as they have been dredged, they are commonly considered as a solid waste and managed as such, i.e., sent to a disposal site; yet it may become practically interesting, in a context of depletion of certain resources, to consider recovery pathways for these materials (Jang *et al.* 2010).

Making informed decisions on the fate of dredged stormwater sediments requires a good understanding of their characteristics; in particular, a crucial matter appears to be their contamination levels relative to the different substances encountered in urban runoff. Even though sediment quality in SuDS has already been addressed in several research works, most of these studies targeted either stormwater ponds, i.e., structures with a permanent water level (Bishop *et al.* 2000; Liebens 2001; Crawford *et al.* 2010; Jang *et al.* 2010; Crane 2014, 2019; Flanagan *et al.* 2021), or small-scale, 'source control' facilities (Tedoldi *et al.* 2017; Cao *et al.* 2019; Lenormand *et al.* 2022). The main conclusions of these studies are detailed and discussed in Section 4.1.2. In brief, trace metals were consistently quantified in sediments, but their concentrations sometimes fell within background values in soils and sometimes reached several hundred mg/kg. Contamination by polycyclic aromatic hydrocarbons (PAHs) was pervasive, but again with a notable variability across sites. The detection of substances belonging to other families of micropollutants (e.g., polychlorinated biphenyls (PCBs) and pesticides) was generally less frequent and not necessarily related to the catchment land use. All studies exhibited at least one substance whose concentration exceeded regulatory and/or toxicity threshold values in soils or benthic environments.

Data are scarcer and older regarding centralized, dry infiltration systems, mainly focusing on trace metals and/or PAHs in a few basins (Dechesne *et al.* 2005; Karlsson *et al.* 2010; El-Mufleh *et al.* 2014) and less on priority substances and micropollutants of huge environmental and health concerns such as alkylphenols and pesticides (Wiest *et al.* 2018). Yet, such basins contribute to the global amount of dredged stormwater sediments, with a potentially different composition and reactivity due to mainly aerobic conditions (Drapeau *et al.* 2017). In addition, the above-mentioned references also reveal significant site-to-site disparities in the contents of metals and organic micropollutants, which may span over >2 orders of magnitude. Despite multiple investigations into the causes of this variability, no consensus has been reached regarding the most influential explanatory variables or the impact of the catchment type (Liebens 2001; Egemose *et al.* 2015; Sharley *et al.* 2017; Flanagan *et al.* 2021), which may suggest the existence of a global 'urban signature' smoothing the influence of site-specific factors.

The core objectives of the research project reported in this paper were, therefore, (i) to characterize and capture the key contamination pattern of stormwater sediments accumulated in dry infiltration basins and (ii) to examine possible links between sediment composition and specific features of the drained urban catchment. For that purpose, 18 infiltration basins with contrasting designs and a variety of catchments were retained as study sites, to constitute a representative panel of urban and peri-urban land uses. In each site, the sediment layer was sampled and analyzed for a wide range of pedological parameters and metallic and organic micropollutants. Understanding and characterizing this contamination pattern is believed to support a more sustainable way to manage urban sediments produced in these structures – notably by offering appropriate valorization pathways for dredged sediments.

2. MATERIAL AND METHODS

2.1. Description of the study sites

2.1.1. Presentation of the study area

The *Greater Lyon* conurbation is located at the confluence of the Rhône and Saone Rivers in France. The eastern part of the territory is a relatively flat area, where the surface geological formations are essentially composed of a glacial fluvial deposit formed from old and recent alluvium interspersed with a few moraine hills with, to the west, the alluvial plain of the Rhône formed by fluvial alluvium (Figure 1). A variety of land uses can be encountered, with residential and industrial catchments, traffic areas, large parking lots, and even agricultural watersheds (the latter notably appear at the southeastern edge of the area, which borders more rural regions), making the study area representative of most metropolitan areas.

The sector's topography, the absence of streams to serve as outlets for urban drainage systems, and the high permeability of the glaciofluvial deposits (generally $\geq 10^{-5}$ m/s, as reported by Dechesne *et al.* (2005)), have led to the wide implementation of infiltration facilities for stormwater management since the 1980s. This territory nowadays includes approximately 100 infiltration basins of various sizes and shapes, collecting runoff water from all types of catchments – except the dense city center of Lyon because of space constraints.

2.1.2. Selection of infiltration basins across land uses

To ensure the representativeness of the sediments in relation to the different urban activities, a typology was established according to the catchment characteristics. These were found in the following databases: (i) data provided by the *Greater Lyon* conurbation (T. Winiarski, Personal Communication): position and name, nature of the basin (enclosed or open), maximum volume capacity, accessibility, date of construction, and drainage network (position, elevation, slope, and length); (ii) data supplied by the *Water Development and Management Scheme*

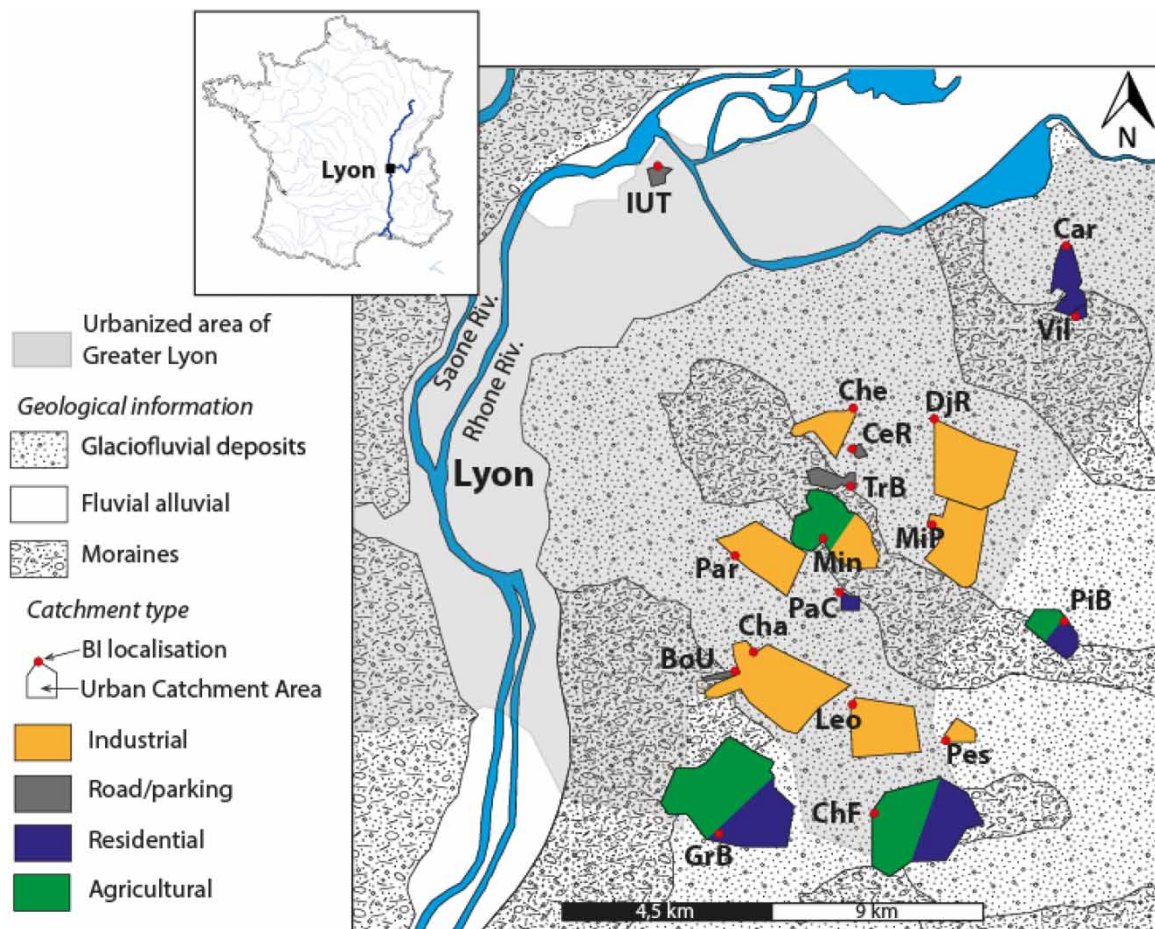


Figure 1 | Location of the 18 study sites in the *Greater Lyon* conurbation.

(SAGE) of the Eastern Lyon District (<https://www.sage-est-lyonnais.fr/>): project owner, facilities, type of urbanization and catchment size, groundwater vulnerability; (iii) data from the *European Environment Agency* (<https://land.copernicus.eu/en/products>): Corine Land Cover and soil sealing; and (iv) data from the *French National Institute of Geographic and Forestry Information* (IGN): BD ALTI® (<https://geoservices.ign.fr/bdalti>) and BD ORTHO® (<https://geoservices.ign.fr/bdortho>). This approach resulted in the selection of 18 representative infiltration basins implemented in relatively similar geological contexts (Figure 1).

2.1.3. Main characteristics of the selected sites

The area of the urban catchments varies from 2 to 434 ha, and the storage volumes from 500 to >60,000 m³ (Table S1 in the Supplementary Material). All basins can be described as centralized systems, with an infiltration/catchment area ratio ranging from 0.1 to 1.2% (median: 0.5%). Three basins are not connected to a sewer system (direct inflow of surface runoff). By combining the catchment surfaces with Corine Land Cover, the following typology was obtained: industrial and commercial areas, discontinuous urban fabric, agricultural land, cropping systems, road and rail networks and associated areas, and urban green areas. To simplify this classification, some classes were clustered as follows: industrial, road/parking, residential, and agricultural (Figure 1). Beyond their predominant land use, the catchments were then scrutinized using aerial views available via the French geographical database *Géoportail*, to identify – where present – punctual elements that may represent significant sources of several contaminants. The specific features that have been identified are summarized in Table S2 in the Supplementary Material.

Most basins had been in operation for more than 10 years when sampling campaigns were undertaken. The date of the last dredging operation was not always known exactly, but the accumulation period of sediments was in any case greater than 4 years (J. Couvidat, Personal Communication with operational teams from the *Greater Lyon* conurbation).

2.2. Sample collection

To get an overall vision of the composition of the sediment layer in each study site, a composite sample was formed by merging subsamples from different locations in the basin. Following the recommendations of a previously established sampling protocol (Hébrard-Labit 2006), the number of sampling points ranged from 8 to 18 with a linear progression according to the basin size – except in a few cases where the spatial extent of the sediment layer happened to be too limited to collect 8 independent samples. At each point, the sediment layer was sampled over its entire depth – which varied between a few centimeters and ≥ 20 cm – using a stainless steel shovel or a hand auger. The representative sample obtained after mixing the subsamples was again subdivided and packaged in clean bottles adapted to each type of physicochemical analysis (cf. Section 2.3).

2.3. Sediment characterization and elemental analysis

The sediment samples were sent to an analytical laboratory with COFRAC (French Accreditation Committee) certification. The different analyses that were carried out are summarized in Table 1, along with the corresponding method and standard/reference.

On the one hand, samples were analyzed for a series of pedological parameters: particle size distribution (PSD), organic matter (OM), cation exchange capacity (CEC), pH_{water}, and carbonates. The reasons for these choices are the following (Eggleton & Thomas 2004): the fine fraction of the sediment is known for concentrating contaminants of various kinds due to the high specific surface area of the particles that compose it; OM plays a crucial role in the sorption of dissolved inorganic elements and hydrophobic compounds; CEC expresses the total charge of exchangeable cations that 1 kg of dry soil is capable of holding and is therefore often used as a proxy for physisorption mechanisms; pH is also a fundamental parameter controlling the mobility of trace inorganic elements; carbonates may also play a role in metal retention because metal carbonates (CuCO₃, PbCO₃, etc.) tend to precipitate in calcareous materials.

On the other hand, samples were analyzed for elemental composition: major elements (silicon, iron, calcium, aluminum, titanium), trace metals (copper, lead, zinc, cadmium, nickel, chromium), and six families of organic micropollutants: 16 PAHs; 7 PCBs; 12 dioxin-like PCBs (DL-PCBs); 17 polychlorinated dibenzodioxins and furans (PCDD/Fs); p-tert-Octylphenol (OP), and 4-nonylphenol (NP); and 25 pesticides. The comprehensive list of analyzed compounds is provided as Supplementary Material (Table S3).

Table 1 | Summary of the physicochemical analyses performed on the 18 sediment samples

Analysis	Method/device	Standard/reference
PSD	Laser diffraction method	ISO 13320 (2009)
OM	Loss on ignition (LOI) at 550 °C for 4 h	NF EN 15169 (2007)
pH _{water}	Soil suspension in an aqueous solution (ratio 1:5)	ISO 10390 (2005)
Carbonates	LOI at 950 °C for 2 hours	Heiri <i>et al.</i> (2001)
CEC	Hexammincobalt trichloride solution	NF EN ISO 23470 (2018)
Major elements	X-ray fluorescence	<i>Internal method</i>
6 trace metals	Flame atomic absorption spectroscopy (FAAS) following acid digestion with <i>aqua regia</i> (HCl + HNO ₃)	FD T90-112 (1998)
16 PAHs	Gas chromatography-mass spectrometry (GC-MS)	ISO 18287 (2006)
7 PCBs	GC-MS	NF EN 16167 (2018)
12 DL-PCBs	GC-MS	Webster <i>et al.</i> (2013)
17 PCDD/Fs	GC-MS	DIN 38414-24 (2000)
2 alkylphenols	GC-MS	ISO/TS 13907 (2012)
25 pesticides	Gas chromatography-time-of-flight mass spectrometry (GC-TOF) and High-performance liquid chromatography-tandem mass spectrometry (HPLC-MS/MS)	<i>Internal method</i>

2.4. Estimation of the reference contents in local soils

Characterizing the site-specific contamination requires evaluating the baseline levels in the soils of the study area. For metals, usual content ranges were estimated from a national database of trace metals in arable topsoils called 'BDETM'. The latter contains data acquired for two decades (1990–2010) on eight metals, grouped by 'small agricultural regions' according to the French territorial nomenclature. In the region closest to our study sites where measurements were available (*Bas Dauphiné*), the first and ninth deciles of the measurements were 0.18–0.32 mg/kg for cadmium, 16–24 mg/kg for nickel, 30–47 mg/kg for chromium, 8–13 mg/kg for copper, 23–37 mg/kg for lead, and 42–60 mg/kg for zinc.

Regarding organic micropollutants, detailed data are not available at such a local scale; however, there exists a measurement network at a national scale called 'RMQS', which conducts long-term monitoring of soil quality based on a regular sampling grid with 2,200 points over the entire metropolitan territory (Saby *et al.* 2020). The first and ninth deciles of soil concentrations were 130–390 µg/kg for \sum^{16} PAHs, <LOQ (limit of quantification)–1.6 µg/kg for \sum^7 PCBs, and <LOQ ≤ 65 ng/kg for \sum^{17} PCDD/Fs. The latter value was itself estimated as an upper bound, as several compounds were still <LOQ.

For each substance (or group of substances), it was considered that exceeding the ninth decile of reference contents could be reasonably attributed to site-specific sources of contamination.

2.5. Data analysis

The data have been made available in a data repository (Tedoldi *et al.* 2023).

Due to the non-Gaussian distributions of some variables, non-parametric statistics were preferred over parametric statistics (Whitley & Ball 2002; Dodge 2008). Correlations between variables were assessed via Spearman's rank correlation coefficient and the associated non-parametric test of significance. The existence of a difference between groups was assessed via the Wilcoxon–Mann–Whitney test (two groups) or the Kruskal–Wallis test (three or more groups), the null hypothesis of which states that all groups originate from the same distribution.

Hierarchical cluster analysis (HCA) was also carried out. Due to substantial differences in the orders of magnitude of contaminant concentrations, normalization via *z*-scores was first applied to the dataset. Euclidean distance was used as a measure of the dissimilarity between two samples, and Ward's method was used as an agglomeration criterion for HCA.

3. RESULTS

3.1. Pedological parameters and major elements

The main results of the sediments' textural and pedological characterization are presented in Table S4 in the Supplementary Material. Most samples were relatively fine-grained, with a median grain diameter $\leq 40 \mu\text{m}$ for 14 sites out of 18. This fine fraction was dominated by silt, as the clay fraction did not exceed 14% across all sites and remained $\leq 8\%$ in half of them. A smaller group of sites had sediments with a coarser texture, which were categorized as *sandy loam* according to the USDA classification.

Overall, the sediments were rich in OM, with OM contents up to $\sim 30\%$; the median value almost reached 18%, and only four sites had OM contents below 10%. To set some orders of magnitude, the median OM content in the nearest agricultural soils that were surveyed within the 'BDETM' research program is between 2 and 3%. Two groups of sites stood out from the distribution of carbonates: half of the values ranged from 1 to 2%, while the upper half of the distribution spanned between 2.6 and 6.5%. pH_{water} was consistently neutral to slightly alkaline in sediments (6.8–8.1 with a median of 7.9). The samples with the lowest pH values all had a carbonate content $\leq 2\%$, but not the other way around. CEC was quite high, with most values falling between 20 and 50 cmol_+/ kg , underlining the sediments' reactivity. For comparison, the median CEC value measured in the soils of the alluvial area east of Lyon is about 8 cmol_+/ kg .

The elemental characterization of the sediments revealed a relative stability of their composition with respect to major elements (Figure S1 in the Supplementary Material): across all sites, the first and ninth deciles of the distributions differed only by a factor of 2, except for calcium which showed a higher dispersion. The sediments were mainly of siliceous type, with Si contents ranging from 120 to 220 g/kg except for one site ($\sim 80 \text{ g/kg}$). Fe and Ca were measured in quite similar proportions (median of 25 and 21 g/kg, respectively), while Al and Ti presented a lower enrichment in the sediments (median of 11 and 3 g/kg, respectively).

3.2. Trace metals

Trace metals were consistently quantified in the sediment samples at levels ranging from a few mg/kg to several thousand mg/kg; the inter-site variability of metal levels is displayed in Figure 2(a). In order of increasing concentrations, the six metals were ranked as follows (median value in parentheses): Cd (0.87 mg/kg) < Ni (54 mg/kg) \approx Cr (57 mg/kg) < Cu (150 mg/kg) \approx Pb (150 mg/kg) < Zn (570 mg/kg). Overall, the values exhibited a higher dispersion than the contents of major elements, as exemplified by the ratio between the third and first quartiles of the distributions, which was around 2 for Ni, Cr, and Pb; 2.5 for Cd; and >4 for Cu and Zn. Similarly, the ninth-to-first decile ratio remained <3 for Ni and Cr, while it rose up to 5.5 for Pb, 9 for Cu, and 15 for Cd and Zn.

3.3. Organic micropollutants

Figure 2(b) presents the total contents (i.e., the sum of all individual compounds) of 17 PCDD/Fs, 7 PCBs, 12 DL-PCBs, and 16 PAHs, as well as octylphenol and nonylphenol levels. A large part of the molecules from this list were consistently quantified in the sediment samples, with the following exceptions.

- For PCDD/Fs, 1,2,3,4,7,8-hexachlorodibenzo-*p*-dioxin (1,2,3,4,7,8-HxCDD) and 1,2,3,7,8,9-hexachlorodibenzo-*p*-dioxin (1,2,3,7,8,9-HxCDD) were quantified in 16–17 samples out of 18, so was 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (often referred to as TCDD) in 13 out of 18 samples; conversely, 1,2,3,7,8,9-hexachlorodibenzofuran (1,2,3,7,8,9-HxCDF) was systematically below the LOQ. Overall, the non-quantified levels represented a maximum of 3% of the total PCDD/F content and most often $<1\%$.
- Among the 7 indicator PCBs and the 12 DL-PCBs, PCB 28, on the one hand, and PCBs 81 and 169, on the other hand, were the only ones to present $<\text{LOQ}$ levels in a maximum of two samples, amounting to $\leq 3\%$ of their total content.
- The situation was more contrasted for PAHs. Four compounds were systematically quantified (naphthalene, fluoranthene, pyrene, and benzo(a)anthracene), and five others in all but one to three samples (phenanthrene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, and benzo(a)pyrene). The frequency of quantification dropped to 2/3 for anthracene, indeno(1,2,3-*cd*)pyrene, and benzo(ghi)perylene and to $<1/2$ for the remaining five PAHs. Non-quantified values accounted for $<4\%$ of the total PAH content in half of the samples, but this proportion rose to 15% or more in a quarter of the samples.

The orders of magnitude of these micropollutants' contents were markedly different from those of trace metals, as they were in the $\mu\text{g/kg}$ range – and up to several mg/kg for PAHs in a few samples. The concentration levels

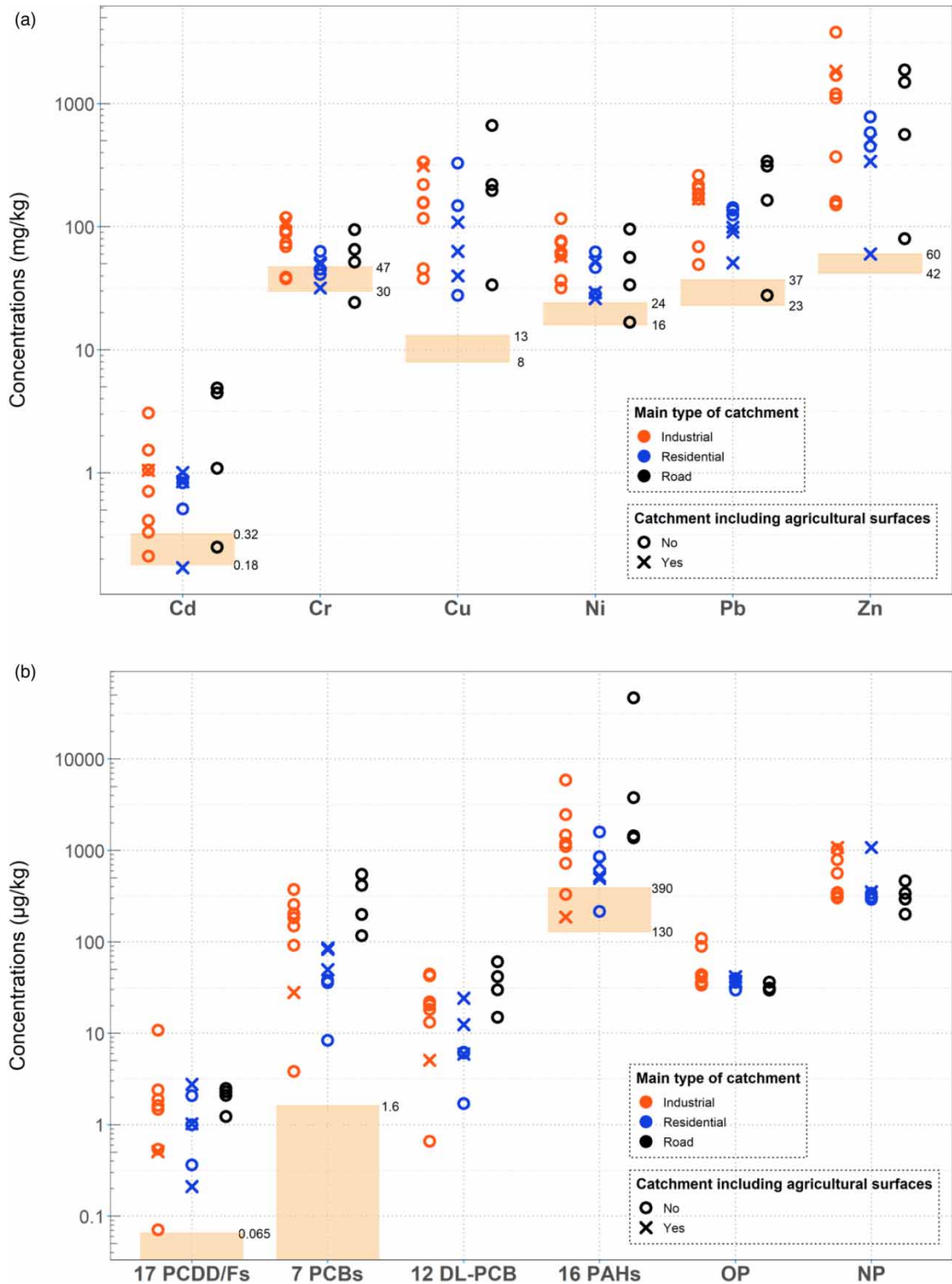


Figure 2 | Distributions of (a) trace metals and (b) organic micropollutants in the sediments from the 18 study sites; comparison with usual contents in local topsoils (when available) displayed as first-to-ninth deciles intervals (colored rectangles).

scaled as follows (median value in parentheses): \sum^{17} PCDD/Fs (1.6 µg/kg) < \sum^{12} DL-PCBs (16 µg/kg) < OP (36 µg/kg) < \sum^7 PCBs (100 µg/kg) < NP (340 µg/kg) < \sum^{16} PAHs (1,100 µg/kg). OP and NP presented the lowest dispersion across all sites, with a very narrow central distribution around the median values characterized by a third-to-first-quartile ratio of 1.2 and 1.7, respectively. This dispersion parameter was ~ 3 for PAHs and PCDD/Fs, and between 4 and 5 for PCBs and DL-PCBs.

Contrary to the above-mentioned substances, most of the 25 targeted pesticides were not detected in all sediment samples. Isoproturon and chlorpyrifos occasionally exceeded the limit of detection but remained <LOQ (9.6 and 1.7 µg/kg, respectively). Alachlor was quantified in two sites (1.6 µg/kg in *GrB* and 9.6 µg/kg in *Car*). Diuron was the most frequently quantified pesticide (10/18 sites), with contents in the range <0.8–20 µg/kg and a median value of 3.2 µg/kg. Noticeably, an accumulation of uncommon pesticides was observed in the *Cha* basin, with 80 µg/kg dichlorodiphenyltrichloroethane (DDT), 150 µg/kg dichlorodiphenyldichloroethylene (DDE), 21 µg/kg dichlorodiphenyldichloroethane (DDD) and 15 µg/kg trifluralin.

4. DISCUSSION

4.1. Evidence of catchment-induced contamination of the sediments

4.1.1. Comparison to reference contents

The sediment samples from almost all study sites displayed higher metal contents than the baseline levels in local soils (Figure 2(a)). This was systematically the case for copper, as well as for zinc, lead, and nickel in all but one site; cadmium and chromium contents fell more frequently within common values in soils (in three and six sites, respectively). With respect to the upper range of reference contents, the median levels of copper and zinc in sediments represented an enrichment by one order of magnitude, and those of lead, nickel, and cadmium by a factor of 2–4.

The observation was similar for organic micropollutants – at least those monitored within the ‘RMQS’ measurement network – whose contents in stormwater sediments generally exceeded the national anthropogenic background in soils by up to two orders of magnitude (Figure 2(b)). This was notably the case for PCDD/Fs and PCBs, which were systematically more concentrated than the ninth decile of measurements in French topsoils. For PAHs, the three least concentrated sediment samples showed total contents below 360 µg/kg, which remained within usual ranges in French topsoils, but the most contaminated sites were 10–100 times above the reference values. No data were available at the national scale for alkylphenols, which constitute contaminants of emerging concern, nor for DL-PCBs.

Hence, whatever the site considered and the characteristics of its catchment, the accumulation of stormwater sediments over and/or within the soil that forms the substrate of infiltration systems constitutes a *contamination*, in the sense that it causes an input of most – if not all – of the targeted substances at anomalous levels.

4.1.2. Comparison to literature values

To further contextualize the composition of the sediments collected at the 18 study sites, a comparison to published data on sediments from similar SuDS is provided in Table 2. Metal contents determined in this study appeared to be in the upper range or even higher than literature values, especially for nickel, copper, lead, and zinc – which were already the four metals with the largest anomalies relative to baseline levels in soils. Statistical distributions of PAH contents were fairly comparable – with the exception of extreme values – between the present results and the measurements by Bishop *et al.* (2000), Weinstein *et al.* (2010), and Flanagan *et al.* (2021). Conversely, Crane (2014) found higher PAH contents in stormwater sediments, especially in industrial areas from Minnesota, USA (the median value of \sum^{16} PAHs was approximately six times greater than in the present assessment). Based on the fewer studies that targeted PCBs and DL-PCBs in stormwater sediments, the present results were found to be one or two orders of magnitude higher than literature values.

Contrariwise, the detection of pesticides – other than diuron – in sediment samples appeared less frequently in the present work than in most of the papers reviewed. At the one site where DDT, DDD, and DDE were quantified (*Cha*), the measured contents were of the same order of magnitude as in the studies by Jang *et al.* (2010) and Crawford *et al.* (2010), but substantially lower than what Flanagan *et al.* (2021) observed in a stormwater pond from Sweden. These studies targeted and quantified common pesticides with the present assessment (e.g., organochlorides such as endosulfan, dieldrin, endrin, and organophosphates such as chlorpyrifos), while all of these compounds remained <LOQ in the present study sites. Literature data on alkylphenols are even scarcer than for all above-mentioned contaminants, with only two studies investigating their presence in stormwater sediments (Crane 2019; Flanagan *et al.* 2021). In contrast to these authors’ observations, both OP and NP were consistently quantified in all collected sediment samples, and the measured NP contents spanned a noticeably narrower range than in these two studies.

Therefore, the contamination of stormwater sediments is subject to a variability that this literature survey proved to be quite significant for metals; nevertheless, despite a diversity of land uses, most sites under study belonged to the upper range of metal distributions compared to the previously documented sites worldwide, suggesting a shared

Table 2 | Summary statistics of the contaminant contents in stormwater sediments; comparison with literature data and French river sediments

	Metals	Ni	Cr	Cu	Pb	Zn	Organics	∑⁷ PCBs	Others
	Cd	mg/kg	mg/kg	mg/kg	mg/kg	mg/kg	∑¹⁶ PAHs	μg/kg	μg/kg
	mg/kg						mg/kg		
This study	0.17–4.90 (0.87)	17–116 (54)	24–119 (57)	28–663 (152)	28–341 (153)	58–3,800 (569)	<0.19–46.7 (1.15)	4–546 (104)	DDT: <6–80 {1/19} DDD: <5–21 {1/19} DDE: <3–152 {1/19} OP: 30–109 (36) NP: 200–1,080 (343) DL-PCBs: 0.65–60.7 (16.5)
15 stormwater ponds near Toronto, Canada (Bishop <i>et al.</i> 2000)	<0.10–1.2 (0.57)	4–22 (11)	7–817 (18)	11–63 (37)	3–68 (29)	29–535 (274)	0.16–98.4 (1.93)	∑ ⁵⁹ PCBs: <500–789 {2/15}	DDT: <5–8.9 {1/15} DDE: <1–3.8 {2/15} Dieldrin: <5
24 stormwater ponds in Florida, USA (Liebens 2001)	<0.1–3.13 (0.64)	2–24 (10)	<1–55 (17)	<1–55 (17)	5–777 ^a (60)	<1–622 (114)	–	–	–
22 stormwater ponds in Florida, USA (Jang <i>et al.</i> 2010)	<0.37–5.30 {1/73}	5–40 [9.7, σ = 0.3]	<2–175 [7.5, σ = 1.6]	4–90 [13.3, σ = 0.7]	<2–196 [7.6, σ = 2.0]	5–711 [47.5, σ = 1.2]	FLA: <5–59.3 PHE: <5– 27.8 PYR: <5–75.2 {3–4/65} ^b	–	DDT: <25–419 {20/65} DDD: <25–364 {3/65} DDE: <25–103 {3/65} Aldrin: <25–41 {1/65} Dieldrin: <25–495 {4/65}
18 stormwater ponds in South Carolina, USA (Crawford <i>et al.</i> 2010; Weinstein <i>et al.</i> 2010)	<1.5–4.3 (1.9)	–	–	<5–600 (30)	–	<1–580 (60)	0.1–159 (2.0)	–	DDD: <10–94 {7/36} DDE: <5–70 {7/36} Chlorpyrifos: <2–28 (6.5) Aldrin, dieldrin: <5
15 stormwater ponds in the Minneapolis–St. Paul area, USA (Crane 2014, 2019)	[1.5, σ = 0.5]	[23, σ = 7]	[42, σ = 15]	[32, σ = 10]	[39, σ = 26]	[154, σ = 80]	∑ ¹⁸ PAHs: 1.4–203 (6.3)	–	OP: < 2.5 NP: 32–6,300 + 1 outlier: 85,000
17 stormwater ponds in Sweden (Flanagan <i>et al.</i> 2021)	–	–	–	–	–	–	<0.20–6.4 (0.64)	<0.4–100 (3.2)	DDT: <10–1,580 {1/34} DDD: <10–869 {1/34} DDE: <10–216 {2/34} OP: <10–529 {13/34} NP: <100–30,500 {13/34}
7 different stormwater facilities in Maryland, USA (Cao <i>et al.</i> 2019)	–	–	–	–	–	–	–	0.7–4.1 (0.9)	DL-PCBs: 0.45–2.95 (1.81)
4 different infiltration facilities in Sweden (Karlsson <i>et al.</i> 2010)	0.1–1.0 (0.5)	14–35 (25)	25–70 (50)	25–250 (175)	30–80 (50)	100–1,400 (800)	–	–	–
Long-term monitoring of two infiltration basins in France (El-Mufleh <i>et al.</i> 2014)	0.5–3.7 1.5–11.4	–	100 –	84–366 97–313	30–819 ^a 56–234	683–2,463 703–2,500	1.7–4.3 4.6–36.4	– –	– –

Urban road dust, worldwide (Majumdar <i>et al.</i> 2017; Roy <i>et al.</i> 2022)	0.3–11.2 (1.8)	11–87 (30)	14–175 (76)	33–430 (100)	37–520 (120)	110–820 (270)	0.9–49.0 (6.3)	–	–
French river sediments (Padox <i>et al.</i> 2010b), $N > 5,600$	0.18–5.50 (0.65)	7–45 (19)	11–96 (36)	7–100 (22)	10–215 (33)	40–882 (130)	0.24–14.2 (1.49)	7–140 (57)	–
Marine sediments from French coastal waters (Padox <i>et al.</i> 2010a), $N > 460$	0.11–1.10 (0.33)	10–33 (20)	24–80 (51)	11–185 (41)	19–99 (41)	65–441 (150)	0.31–9.74 (1.49)	Ind. PCBs ^c : <3–25 to 50 (<10)	–

Data are presented as min–max (median), [mean, standard deviation (standard deviation is referred to as σ and presented when available)], or {frequency of quantification}, except for road dust and river/marine sediments where the range corresponds to the first and ninth deciles of the measurements.

^aHigh Pb contents in the data acquired prior to or during the 2000s constituted the signature of leaded gasoline, the use of which was banned in most countries at the turn of the 20th and 21st centuries.

^bThe low frequency of quantification that was reported by Jang *et al.* (2010) for PAHs is probably related to a relatively high detection limit (5 mg/kg for each individual PAH).

^cStatistics on PCBs in marine sediments are only available for individual compounds and not for Σ^7 PCBs.

urban specificity. Conversely, apart from extreme values, sediment impregnation by PAHs seemed more generalized and homogeneous between countries, which is probably linked to the ubiquity of their emission sources. The observed remanence of PCBs at elevated levels – compared to sediments from other countries – is of particular concern, as these substances were banned in France in 1987, but still build up in infiltration basins due to their well-documented persistence in the environment. This has been certainly reinforced by a local industrial context prevailing before the 1980s decade, which generated a historical PCB contamination in the Rhône River and Lyon area (Mourier *et al.* 2014). Alkylphenol contamination of sediments was pervasive albeit relatively uniform across all land uses, contrary to pesticides that were detected in few sites.

4.2. From the geochemical signature to the sources and fate of contaminants

4.2.1. Elucidation of common emission and transfer dynamics

By accumulating urban contaminants over time, infiltration basins provide an integrated pattern of their emission and transfer dynamics. The case of PAHs is somehow archetypal in this regard, as it has been well documented that their relative distribution in a given environmental media exhibits characteristic features depending on their major source(s). In practice, this is reflected in different *diagnostic ratios*, built from the concentrations of two or more compounds (for example: low- and high-molecular-weight PAHs (LMW and HMW, respectively), anthracene (ANT), phenanthrene (PHE), fluoranthene (FLA), pyrene (PYR), benzo(a)anthracene (BaA), chrysene (CHR), indeno(1,2,3-c,d)pyrene (IcdP), and benzo(g,h,i)peylene (BghiP)), which allow the identification of the predominant emission sources. Based on the ratios and thresholds proposed by Tobiszewski & Namieśnik (2012), the following conclusions could be drawn (Table S5 in the Supplementary Material). The two ratios used to discriminate between petrogenic and pyrogenic emissions ($\sum \text{LMW}/\sum \text{HMW}$ and $\text{ANT}/[\text{ANT} + \text{PHE}]$) systematically indicated a dominance of the latter category, so that combustion was prevalent ahead of, for example, fuel leaks. Pyrogenic emissions were primarily attributed to traffic (i.e., petrol/diesel combustion) in all industrial and road catchments based on the ratios $\text{BaA}/[\text{BaA} + \text{CHR}]$ and $\text{IcdP}/[\text{IcdP} + \text{BghiP}]$. Conversely, the ratio $\text{FLA}/[\text{FLA} + \text{PYR}]$, which distinguishes wood/coal combustion (>0.5) and fossil fuel combustion ($0.4\text{--}0.5$), was highest in residential areas ($0.56\text{--}0.58$), unraveling the contribution of heating systems.

Although the link between the geochemical signature of the other contaminants and their predominant sources and/or transport pathways is not as clear as for PAHs, the variety of study sites enables common patterns to be identified between different groups of substances. Three groups stood out from the correlation matrix (Figure S2 in the Supplementary Material), with significant positive correlations within a group, and almost consistently weak and non-significant correlations with the rest of the variables: (i) the six trace metals; (ii) the group formed by PAHs, PCBs, DL-PCBs, and PCDD/Fs; and (iii) alkylphenols. Shared similarities within each group, suggesting common emission and transfer dynamics, may be illustrated, for example, by land use- or traffic-related sources of metals (Müller *et al.* 2020), joint use of NP and OP as adhesives and tackifiers in the tire rubber industry (Lamprea *et al.* 2018), and possibly airborne PAHs, PCBs, and PCDD/Fs. Conversely, the lack of correlation between groups shows that the prevalence of emission sources of one contaminant type is largely independent of the other types. This had already been observed for metals and PAHs in infiltration systems (Tedoldi *et al.* 2017) but is an original result for the other micropollutants.

4.2.2. Why are pesticides encountered in stormwater sediments?

Pesticides were not included in the previous correlation matrix, as they were most often $<\text{LOQ}$. In addition, when quantified, their concentrations in sediments could hardly be interpreted and compared in absolute terms: most of them are indeed hydrophilic and therefore not well sorbed onto solid materials (according to the review by Weber *et al.* (2004), the sorption of diuron and alachlor onto various types of soil is characterized by an average K_d value of ~ 7 and 2 L/kg , respectively), in addition to being prone to degradation – with the notable exception of DDT and its metabolites that are known for being persistent in the environment. Still, pesticide occurrence in sediments may seem somewhat unexpected, since all quantified molecules have been prohibited in France – the oldest ban dating back to 1971 for DDT. Diuron, alachlor (the main component of an herbicide formerly marketed by Monsanto), and trifluralin were prohibited as *pesticides* in the 2000s, but the quantified levels in recently accumulated stormwater sediments did ascertain their presence in urban runoff. This remanence could be explained by the remaining allowance of certain uses in urban areas (in building materials for example), since pesticidal and biocidal uses are subject to distinct regulations. A recent study, which targeted surface waters from the Paris region, evidenced a decrease in diuron concentrations after its ban as a

pesticide, followed by stabilization at a non-negligible background attributed to urban inputs (Paijens *et al.* 2022). The present results corroborate these conclusions and emphasize the contribution of stormwater runoff to the transfer of biocidal substances in urban catchments.

4.2.3. Pedological and mineralogical descriptions enable a better understanding of contaminant dynamics

For most of the sediment samples, the median value of the PSD (D_{50} in Table S4) ranged from 18 to 40 μm . This order of magnitude is similar to, or slightly lower than, the characteristics of suspended solids in stormwater runoff (Flanagan *et al.* 2019). This observation supports the idea that coarser particles may settle prior to reaching the infiltration basin (e.g., in a retention structure where present) while the finer ones are intercepted and embedded into the surface layer. Few studies allow a mineralogical description of the different crystalline species present in these deposits. According to Badin *et al.* (2009) and Drapeau *et al.* (2017), who have investigated common study sites with the present assessment, sediments contain mainly carbonates and primary minerals such as quartz, calcite, albite, and chamosite that predominate in these materials. The precise mineralogical composition was found to depend essentially on the characteristics of the drained catchment and on the local geochemical context.

Of interest is the fact that pedological parameters – OM and pH, but also sand, silt, clay, carbonates, and CEC, not shown on Figure S2 – did not exhibit any correlation with contaminant levels in sediments, even though they are among the main factors driving the solid phase reactivity in natural environments (cf. Section 2.3). This observation illustrates a key feature of contaminant accumulation in stormwater management systems: the particles that constitute the sediment layer are already partially contaminated when they arrive in the devices, and these contamination levels appear to be independent of their ability to subsequently intercept dissolved contaminant fluxes. This is in line with the observations of Clozel *et al.* (2006), who suggested that metal speciation had been previously acquired in the urban catchment, before reaching the infiltration basin at the outlet. A similar conclusion was reached by Rommel *et al.* (2021) in the case of roadside sediments, the composition of which did not show a visible evolution between two sampling campaigns separated by a period of 2 years. This raises the question of the influence of the catchment type on the sediment contamination, which will be explored in the following section.

4.3. Catchment-specific features of the sediment contamination

4.3.1. The predominant land use is not a good predictor of sediment contamination

The existence of a catchment-induced difference in sediment contamination was first investigated via statistical testing, after splitting the 18 study sites into three groups according to their predominant land use (residential, industrial, and road). For all contaminants except PCBs, the Kruskal–Wallis test did not allow any difference to be detected between these three groups, as a result of a relatively high dispersion of the contamination levels (Figure 2). More specifically, a Wilcoxon–Mann–Whitney test subsequently demonstrated that the only significant difference occurred between \sum^7 PCBs in residential and road catchments. In other words, considering only the predominant land use generally did not enable the identification of contamination patterns that would be specific to one catchment type, as previously discussed by Flanagan *et al.* (2021). This observation stands in contrast to smaller scale, source control facilities where the link between land use and soil/sediment contamination has proved easier to establish (Egemose *et al.* 2015; Tedoldi *et al.* 2017).

4.3.2. Unraveling and understanding similarities in contamination patterns

The analysis was, therefore, refined by gathering sites according to their similarities in terms of contamination, rather than catchment land use. For that purpose, HCA was applied to the dataset containing (i) the six trace metals; (ii) total contents of PAHs, PCBs, DL-PCBs, and PCDD/Fs; and (iii) OP and NP. Based on a mathematical distance to represent the dissimilarity between sites (cf. Section 2.5), the latter were iteratively agglomerated into clusters, in order to reach the minimum within-cluster variance (Ward's method). The result is the hierarchical tree displayed in Figure 3, where the height of the separation between two branches indicates the distance (i.e., the magnitude of the difference) between two clusters.

The discriminating elements that caused the two main partitions of the dendrogram were the contamination levels and nature, respectively. More specifically, three clusters, as well as a single element, could be distinguished:

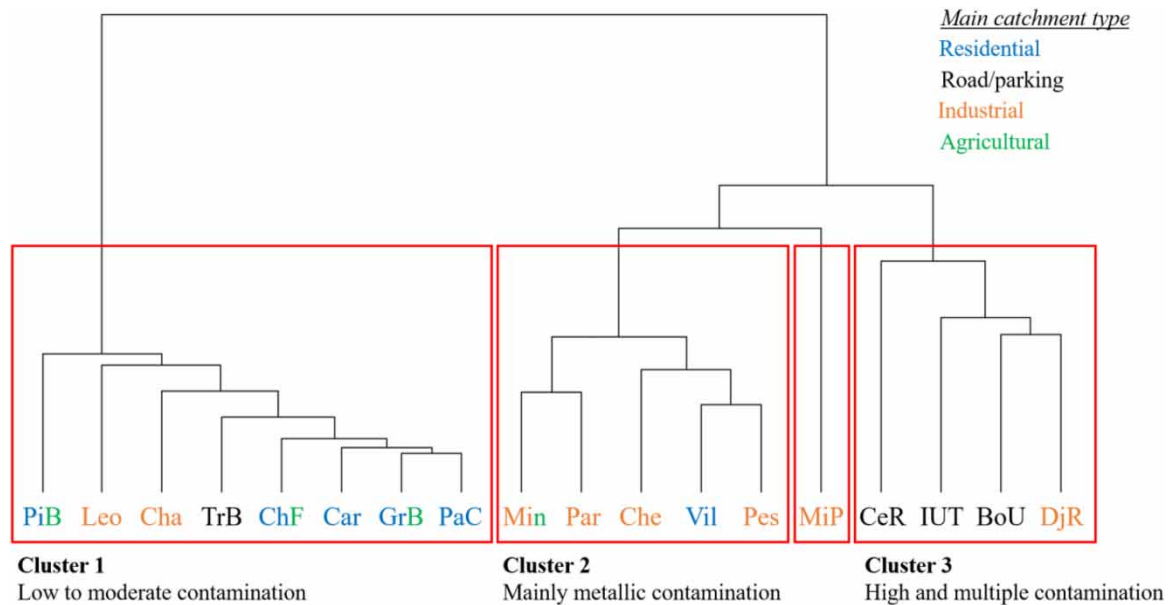


Figure 3 | Dendrogram resulting from HCA of the sediments from the 18 study sites, classified according to their contamination levels.

- Cluster 1: sites characterized by low to moderate contamination levels of the sediments, mainly comprising residential catchments, along with two industrial sites and one parking lot.
- Cluster 2: sites classifying in the upper half of most metal distributions, but exhibiting comparatively low levels of organic micropollutants: these correspond to four industrial catchments and one residential catchment.
- Cluster 3: sites with contamination levels falling within the upper range of the distributions for almost all targeted substances: three of them drain runoff water from road environments, and the fourth one from an industrial catchment.
- The remaining site (*MiP*, industrial) has an atypical contamination profile, which is essentially due to an anomaly in PCDD/Fs (11 µg/kg, 7 times the median value of the 18 sites) and OP concentrations (110 µg/kg, 3 times the median value).

The proximity between the sites from a given cluster could be understood from a detailed description of their catchments and the identification of specific emission sources (Table S2). The case of metals (cluster 2) is detailed as an illustration. The *Che* catchment had large buildings with uncoated zinc roofs, known for generating very high concentrations of dissolved zinc in stormwater runoff, on the order of mg/L or more (De Buyck *et al.* 2021): this latter is likely to be intercepted by the reactive sediment layer and thus induce high Zn contents (ca. 4 g/kg). The situation was similar, albeit to a lesser extent, for the residential *Vil* catchment, where most buildings had tile roofs but were equipped with metallic gutters and watertightness elements. The *Par* and *Min* catchments also presented a high potential for metallic emissions, respectively, due to the presence of a smelter in the middle of a truck production site, and a sheet metal workshop and a tram maintenance center. The last site from this cluster (*Pes*) had a catchment with various warehouses including metal reinforcement and concrete structures.

Hence, the lack of differences between catchment types, as observed in Section 4.3.1, must be reconsidered. On the one hand, most residential catchments were found to generate relatively uniform and non-extreme contamination (cluster 1). On the other hand, industrial areas and road environments apparently have a higher probability to generate high emissions of *at least* one contaminant, but they may as well fall among the least contaminated sites for other substances. For these two catchment types, the predominant land use may be insufficient to distinguish – where appropriate – urban configurations that are particularly emissive of certain contaminants.

4.4. What makes stormwater sediments specific from other solid materials?

The nature of stormwater sediments is somewhat ambiguous, especially considering the multiple origins of runoff particles. To situate them in a wider spectrum of solid materials, the composition of stormwater sediments was compared to that of road dust, on the one hand, and river and marine sediments, on the other hand.

4.4.1. Comparison to urban road dusts

Road dust refers to the complex mixture of particles accumulated on the roadway during dry weather; these are typically sampled by vacuum cleaning or street sweeping. Metal and PAH contents in urban road dust have been extensively documented in scientific literature, and the results have been synthesized in several review articles, e.g., by Roy *et al.* (2022) for metals and by Majumdar *et al.* (2017) for PAHs (Table 2). Compared to road dust, metals in sediments from the present study sites displayed a similar variability and contamination levels of the same order of magnitude – although the previously noted enrichment of nickel, copper, lead, and zinc also emerged from this comparison. The conclusions were slightly different when considering literature data, most of which indicated lower metal contents in sediments than in road dust (Table 2). PAH contents spanned approximately the same range in both types of materials, but the median value was approx. five times higher in road dust.

PCBs have been little studied in road dust; most notably, the available data were acquired in different world regions. In India, Nayak *et al.* (2021) reported values of \sum^9 PCBs (including the seven congeners under study + PCB 31 and 158) between 4 and 24 $\mu\text{g}/\text{kg}$ (median: 10 $\mu\text{g}/\text{kg}$); in Vietnam, Anh *et al.* (2019) observed levels of \sum^7 PCBs ranging from 0.5 to 22 $\mu\text{g}/\text{kg}$ (median: 3 $\mu\text{g}/\text{kg}$). These values are close to the results of Cao *et al.* (2019) and Flanagan *et al.* (2021) in stormwater sediments but are particularly low compared to the present observations. A similar discrepancy arose from the comparison of DL-PCBs: road dust contents ranged from 6 to 10 $\mu\text{g}/\text{kg}$, with a median of 7 $\mu\text{g}/\text{kg}$ (Nayak *et al.* 2021), and from 0.2 to 5 $\mu\text{g}/\text{kg}$, with a median of 1.5 $\mu\text{g}/\text{kg}$ (Anh *et al.* 2019), as opposed to the median content of 16 $\mu\text{g}/\text{kg}$ in the present case. This difference can be explained by the above-mentioned local industrial context, responsible for large emissions of PCBs in the 1970s and 1980s, and/or by a more limited use of PCBs in the countries from which literature values were drawn, before these compounds were proven to be hazardous and banned (McLachlan *et al.* 2018).

4.4.2. Comparison to river and marine sediments

As an alternative basis of comparison, the present data were put into perspective with the quality of river and marine sediments in France, which was synthesized in two reports published by the National Institute for the Industrial Environment and Risks (INERIS) (Padox *et al.* 2010a, 2010b). Summary statistics on >5,600 measurements nationwide and >460 measurements in coastal waters, respectively, are also presented in Table 2.

Overall, contaminant enrichment of stormwater sediments relative to river sediments followed the sequence: Cd, Cr, PAHs < Pb, Zn < Ni, PCBs < Cu. On the one hand, the distributions of cadmium, chromium, and PAH contents were relatively similar between river sediments and the present results. On the other hand, for lead and zinc, almost all our data were above the first quartile of measurements in river sediments, and 50% exceeded the third quartile. Likewise, nickel and PCBs appeared to be more concentrated in stormwater sediments, as 12 (resp. 14) sites out of 18 had higher PCB (resp. Ni) contents than the third quartile in river sediments. Finally, the present data on copper contents fell in the very upper range of the distribution in river sediments, the median value of which was consistently exceeded in our samples; additionally, the 12 highest values were within the top 10% of river sediment contents.

The comparison was slightly different when considering marine sediments, as the latter exhibited lower Cd and higher Cu distributions than river sediments; median Cr, Pb, and Zn concentrations were also higher in marine sediments, but the distributions were less skewed – because of which the upper percentiles were lower – than for river sediments. Consequently, the enrichment of stormwater sediments compared to marine sediments followed the sequence: Cr, PAHs < Cd, Ni < Cu, Pb, Zn. Hence, whatever the reference they are compared to, stormwater sediments exhibit differences from other matrixes that shed light on urban-specific sources of contaminants. Despite the inter-site variability discussed above, these observations do support the idea of an ‘urban geochemical signature’ borne by stormwater sediments.

5. CONCLUSION: ‘URBAN SIGNATURE’ OF STORMWATER SEDIMENTS

The aim of this study was to provide a comprehensive picture of the characteristics of stormwater sediments, with particular emphasis on their contamination, and to understand what makes them specific from other materials. Eighteen contrasting infiltration basins, designed to manage stormwater from a variety of urban and peri-urban catchments, were chosen as study sites. After collecting a composite sample from the whole sediment layer of each site, the latter underwent a pedological characterization as well as a chemical analysis, targeting trace

metals and six families of organic micropollutants: PAHs, PCBs, DL-PCBs, PCDD/Fs, alkylphenols, and pesticides. The main findings can be summarized as follows.

- Some specific features of stormwater sediments are their fine texture, stable and slightly alkaline pH, and very high OM content, which induce a potential reactivity.
- Stormwater sediments exhibit the signature of an urban contamination, as a result of the accumulation of contaminated particles conveyed by urban runoff, and the sorption of dissolved substances onto the sediment layer. Except for pesticides, the emissions of which have a chiefly stochastic component, all the targeted contaminants are ubiquitous in the urban areas considered, whatever the type of catchment.
- The emission and transport dynamics of metals, alkylphenols, and other organic micropollutants (PAHs, PCBs, DL-PCBs, and PCDD/Fs) appear to be globally independent from each other, but common behaviors within each group can be inferred from the strong correlations observed. PAH profiles suggest pyrogenic and mostly traffic-derived emissions, even if wood or coal combustion also represents a significant contribution in residential catchments due to urban heating.
- Stormwater sediments share some similarities with continental and marine sediments regarding their characteristics and composition, but they present a higher enrichment in several contaminants – especially PCBs, nickel, lead, zinc, and copper – highlighting the importance of urban-specific sources of contaminants. Their contamination pattern was found to be closer to road dusts.
- Managing runoff from large urban catchments in centralized infiltration basins induces a mixing and ‘averaging’ effect of contaminant loads: hence, their dominant sources may be difficult to trace individually without a careful examination of the catchment’s characteristics.

Altogether, despite a potentially significant inter-site variability, stormwater sediments from infiltration basins do present an ‘urban signature’ that makes them distinct from other materials alike, concretely embodied in the sediments’ median contamination profile. Fluctuations around these median values result from a higher or lower occurrence of these contaminants’ sources within – or around – the catchment and appear to be unpredictable from the predominant land use alone. Developing a predictive model of the sediment contamination remains a challenging objective, which requires novel approaches toward the adequate spatial description of the catchments and new insights into the long-term evolution of the sediments (including, but not limited to, their reactivity and contamination patterns). Combined with the present findings, such research initiatives would support the selection of relevant valorization pathways for dredged stormwater sediments.

ACKNOWLEDGEMENTS

The authors acknowledge the AERMC for the fruitful discussions and financial support to the DESIR project. Also, the authors are grateful to the OTHU, and especially to Laëtitia Bacot, for their support and project management. This work was also realized within the Graduate School H2O’Lyon (ANR-17-EURE-0018) and Université de Lyon (UdL), as part of the program ‘Investissements d’Avenir’ run by Agence Nationale de la Recherche (ANR). The authors also want to thank all the contributors to the initial program and, in particular, J. P. Bedell, L. Lassabatere, J. Poulenard, and J. M. Dorioz. Finally, the authors acknowledge macrovector/Free-pik, Freepik, and Brgfx/Free-pik for their vectorial arts (available at www.freepik.com) used in the graphical abstract.

AUTHOR CONTRIBUTIONS

JC and DT: Conceptualization, Methodology, Formal analysis, Investigation, Writing – Original Draft, Writing – Review & Editing, Visualization. MG: Conceptualization, Formal analysis, Writing – Review & Editing. QZ: Investigation, Writing – Review & Editing. TW: Conceptualization, Methodology, Investigation, Writing – Review & Editing, Supervision, Funding acquisition. GLK: Validation, Investigation, Writing – Review & Editing, Resources. CD: Conceptualization, Methodology, Resources, Project administration, Supervision. VC: Conceptualization, Investigation, Writing – Original Draft, Writing – Review & Editing, Visualization, Supervision, Project administration, Funding acquisition.

FUNDING

This work was financially supported by the French Water Agency (Agence de l’Eau Rhône-Méditerranée-Corse/AERMC) through the DESIR project, within the cooperation framework set up between the AERMC and the

Field Observatory on Urban Hydrology (*Observatoire de Terrain en Hydrologie Urbaine*/OTHU – federative research structure NO. 4161).

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

REFERENCES

- Ahilan, S., Guan, M., Wright, N., Sleigh, A., Allen, D., Arthur, S., Haynes, H. & Krivtsov, V. 2019 *Modelling the long-term suspended sedimentological effects on stormwater pond performance in an urban catchment*. *Journal of Hydrology* **571**, 805–818. <https://doi.org/10/gm6srb>.
- Anh, H. Q., Watanabe, I., Tomioka, K., Minh, T. B. & Takahashi, S. 2019 *Characterization of 209 polychlorinated biphenyls in street dust from northern Vietnam: Contamination status, potential sources, and risk assessment*. *Science of The Total Environment* **652**, 345–355. <https://doi.org/10.1016/j.scitotenv.2018.10.240>.
- Badin, A.-L., Méderel, G., Béchet, B., Borschneck, D. & Delolme, C. 2009 *Study of the aggregation of the surface layer of Technosols from stormwater infiltration basins using grain size analyses with laser diffractometry*. *Geoderma* **153**, 163–171. <https://doi.org/10.1016/j.geoderma.2009.07.022>.
- Bishop, C. A., Struger, J., Shirose, L. J., Dunn, L. & Campbell, G. D. 2000 *Contamination and wildlife communities in stormwater detention ponds in Guelph and the Greater Toronto Area, Ontario, 1997 and 1998 part II – contamination and biological effects of contamination*. *Water Quality Research Journal* **35**, 437–474. <https://doi.org/10.2166/wqrj.2000.027>.
- Cao, S., Capozzi, S. L., Kjellerup, B. V. & Davis, A. P. 2019 *Polychlorinated biphenyls in stormwater sediments: Relationships with land use and particle characteristics*. *Water Research* **163**, 114865. <https://doi.org/10.1016/j.watres.2019.114865>.
- Clozel, B., Ruban, V., Durand, C. & Conil, P. 2006 *Origin and mobility of heavy metals in contaminated sediments from retention and infiltration ponds*. *Applied Geochemistry* **21**, 1781–1798. <https://doi.org/10.1016/j.apgeochem.2006.06.017>.
- Couvidat, J., Chabert, M. & Chatain, V. 2021 *Etat de l'Art sur la gestion opérationnelle des sédiments de l'assainissement pluvial* (Technical report). AERMC – OTHU.
- Crane, J. L. 2014 *Source apportionment and distribution of polycyclic aromatic hydrocarbons, risk considerations, and management implications for urban stormwater pond sediments in Minnesota, USA*. *Archives of Environment Contamination and Toxicology* **66**, 176–200. <https://doi.org/10.1007/s00244-013-9963-8>.
- Crane, J. L. 2019 *Distribution, toxic potential, and influence of land use on conventional and emerging contaminants in urban stormwater pond sediments*. *Archives of Environment Contamination and Toxicology* **76**, 265–294. <https://doi.org/10.1007/s00244-019-00598-w>.
- Crawford, K. D., Weinstein, J. E., Hemingway, R. E., Garner, T. R. & Globensky, G. 2010 *A survey of metal and pesticide levels in stormwater retention pond sediments in coastal South Carolina*. *Archives of Environment Contamination and Toxicology* **58**, 9–23. <https://doi.org/10.1007/s00244-009-9347-2>.
- De Buyck, P.-J., Van Hulle, S. W. H., Dumoulin, A. & Rousseau, D. P. L. 2021 *Roof runoff contamination: A review on pollutant nature, material leaching and deposition*. *Reviews in Environmental Science and Biotechnology* **20**, 549–606. <https://doi.org/10.1007/s11157-021-09567-z>.
- Dechesne, M., Barraud, S. & Bardin, J.-P. 2005 *Experimental assessment of stormwater infiltration basin evolution*. *Journal of Environmental Engineering* **131**, 1090–1098. [https://doi.org/10.1061/\(ASCE\)0733-9572\(2005\)131:7\(1090\)](https://doi.org/10.1061/(ASCE)0733-9572(2005)131:7(1090)).
- Dodge, Y. 2008 *The Concise Encyclopedia of Statistics*, 1st edn, Springer Reference. Springer, New York.
- Drapeau, C., Delolme, C., Chatain, V., Gautier, M., Blanc, D., Benzazoua, M. & Lassabatère, L. 2017 *Spatial and temporal stability of major and trace element leaching in urban stormwater sediments*. *Open Journal of Soil Science* **7**, 347–365. <https://doi.org/10.4236/ojss.2017.711025>.
- Egemose, S., Sønderup, M. J., Grudinina, A., Hansen, A. S. & Flindt, M. R. 2015 *Heavy metal composition in stormwater and retention in ponds dependent on pond age, design and catchment type*. *Environmental Technology* **36**, 959–969. <https://doi.org/10.1080/09593330.2014.970584>.
- Eggleton, J. & Thomas, K. V. 2004 *A review of factors affecting the release and bioavailability of contaminants during sediment disturbance events*. *Environment International* **30**, 973–980. <https://doi.org/10.1016/j.envint.2004.03.001>.
- El-Mulleh, A., Béchet, B., Ruban, V., Legret, M., Clozel, B., Barraud, S., Gonzalez-Merchan, C., Bedell, J.-P. & Delolme, C. 2014 *Review on physical and chemical characterizations of contaminated sediments from urban stormwater infiltration basins within the framework of the French observatory for urban hydrology (SOERE URBIS)*. *Environmental Science and Pollution Research* **21**, 5329–5346. <https://doi.org/10.1007/s11356-013-2490-3>.
- Flanagan, K., Branchu, P., Boudahmane, L., Caupos, E., Demare, D., Deshayes, S., Dubois, P., Meffray, L., Partibane, C., Saad, M. & Gromaire, M.-C. 2019 *Retention and transport processes of particulate and dissolved micropollutants in stormwater biofilters treating road runoff*. *Science of the Total Environment* **656**, 1178–1190. <https://doi.org/10.1016/j.scitotenv.2018.11.304>.

- Flanagan, K., Blecken, G.-T., Österlund, H., Nordqvist, K. & Viklander, M. 2021 Contamination of urban stormwater pond sediments: A study of 259 legacy and contemporary organic substances. *Environmental Science & Technology* **55**, 3009–3020. <https://doi.org/10.1021/acs.est.0c07782>.
- Hannouche, A., Chebbo, G., Joannis, C., Gasperi, J., Gromaire, M.-C., Moilleron, R., Barraud, S. & Ruban, V. 2017 Stochastic evaluation of annual micropollutant loads and their uncertainties in separate storm sewers. *Environmental Science and Pollution Research* **24**, 28205–28219. <https://doi.org/10.1007/s11356-017-0384-5>.
- Hébrard-Labit, C. 2006 Guide technique: Recommandations pratiques pour la gestion des produits de l'assainissement pluvial (Guide technique). Laboratoire Central des Ponts et Chaussées (LCPC).
- Heiri, O., Lotter, A. F. & Lemcke, G. 2001 Loss on ignition as a method for estimating organic and carbonate content in sediments: Reproducibility and comparability of results. *Journal of Paleolimnology* **25**, 101–110. <https://doi.org/10.1023/A:1008119611481>.
- Jang, Y.-C., Jain, P., Tolaymat, T., Dubey, B., Singh, S. & Townsend, T. 2010 Characterization of roadway stormwater system residuals for reuse and disposal options. *Science of the Total Environment* **408**, 1878–1887. <https://doi.org/10/dwtpkd>.
- Karlsson, K., Viklander, M., Scholes, L. & Revitt, M. 2010 Heavy metal concentrations and toxicity in water and sediment from stormwater ponds and sedimentation tanks. *Journal of Hazardous Materials* **178**, 612–618. <https://doi.org/10.1016/j.jhazmat.2010.01.129>.
- Lamprea, K., Bressy, A., Mirande-Bret, C., Caupos, E. & Gromaire, M.-C. 2018 Alkylphenol and bisphenol A contamination of urban runoff: An evaluation of the emission potentials of various construction materials and automotive supplies. *Environmental Science and Pollution Research* **25**, 21887–21900. <https://doi.org/10.1007/s11356-018-2272-z>.
- Lenormand, É., Kustner, C., Combroux, I., Bois, P. & Wanko, A. 2022 Diagnosing trace metals contamination in ageing stormwater constructed wetlands by portable x-ray fluorescence analyzer (pXRF). *Science of The Total Environment* **844**, 157097. <https://doi.org/10.1016/j.scitotenv.2022.157097>.
- Liebens, J. 2001 Heavy metal contamination of sediments in stormwater management systems: The effect of land use, particle size, and age. *Environmental Geology* **41**, 341–351. <https://doi.org/10.1007/s002540100392>.
- Loganathan, P., Vigneswaran, S. & Kandasamy, J. 2013 Road-deposited sediment pollutants: A critical review of their characteristics, source apportionment, and management. *Critical Reviews in Environmental Science and Technology* **43**, 1315–1348. <https://doi.org/10.1080/10643389.2011.644222>.
- Majumdar, D., Rajaram, B., Meshram, S., Suryawanshi, P. & Chalapati Rao, C. V. 2017 Worldwide distribution of polycyclic aromatic hydrocarbons in urban road dust. *International Journal of Environmental Science and Technology* **14**, 397–420. <https://doi.org/10.1007/s13762-016-1084-2>.
- Marti, R., Bécouze-Lareure, C., Ribun, S., Marjolet, L., Bernardin Souibgui, C., Aubin, J.-B., Lipeme Kouyi, G., Wiest, L., Blaha, D. & Cournoyer, B. 2017 Bacteriome genetic structures of urban deposits are indicative of their origin and impacted by chemical pollutants. *Scientific Reports* **7**, 13219. <https://doi.org/10.1038/s41598-017-13594-8>.
- McLachlan, M. S., Undeman, E., Zhao, F. & MacLeod, M. 2018 Predicting global scale exposure of humans to PCB 153 from historical emissions. *Environmental Science: Processes Impacts* **20**, 747–756. <https://doi.org/10.1039/C8EM00023A>.
- Mitchell Ayers, E. & Kangas, P. 2018 Soil layer development and biota in bioretention. *Water* **10**, 1587. <https://doi.org/10.3390/w10111587>.
- Mourier, B., Desmet, M., Van Metre, P. C., Mahler, B. J., Perrodin, Y., Roux, G., Bedell, J.-P., Lefèvre, I. & Babut, M. 2014 Historical records, sources, and spatial trends of PCBs along the Rhône River (France). *Science of the Total Environment* **476–477**, 568–576. <https://doi.org/10.1016/j.scitotenv.2014.01.026>.
- Müller, A., Österlund, H., Marsalek, J. & Viklander, M. 2020 The pollution conveyed by urban runoff: A review of sources. *Science of the Total Environment* **709**, 136125. <https://doi.org/10/gjbwzf>.
- Nayak, Y., Sahu, Y. K., Patel, K. S., Sharma, S., Hung, C.-C., Martín-Ramos, P. & Yurdakul, S. 2021 Distribution and sources of polychlorinated biphenyls in air, dust, and sediment from India. *Journal of Hazardous, Toxic, and Radioactive Waste* **25**, 05020004. [https://doi.org/10.1061/\(ASCE\)HZ.2153-5515.0000560](https://doi.org/10.1061/(ASCE)HZ.2153-5515.0000560).
- Padox, J.-M., Hennebert, P., Benard, A. & Mancioffi, L. 2010a Qualité chimique des sédiments marins en France : Synthèse des bases de données disponibles – Chemical quality of marine sediments in France: Synthesis of available databases.
- Padox, J.-M., Hennebert, P. & Mancioffi, L. 2010b Qualité chimique des sédiments fluviaux en France – Synthèse des bases de données disponibles (No. INERIS-DRC-10-105335-04971A). INERIS, Aix-en-Provence.
- Paijens, C., Tedoldi, D., Frère, B., Mailler, R., Rocher, V., Moilleron, R. & Bressy, A. 2022 Biocidal substances in the Seine River: Contribution from urban sources in the Paris megacity. *Environmental Science: Water Research & Technology* **8**, 2358–2372. <https://doi.org/10.1039/D2EW00253A>.
- Pal, A., He, Y., Jekel, M., Reinhard, M. & Gin, K. Y.-H. 2014 Emerging contaminants of public health significance as water quality indicator compounds in the urban water cycle. *Environment International* **71**, 46–62. <https://doi.org/10.1016/j.envint.2014.05.025>.
- Rommel, S. H., Stinshoff, P. & Helmreich, B. 2021 Sequential extraction of heavy metals from sorptive filter media and sediments trapped in stormwater quality improvement devices for road runoff. *Science of the Total Environment* **782**, 146875. <https://doi.org/10.1016/j.scitotenv.2021.146875>.
- Roy, S., Gupta, S. K., Prakash, J., Habib, G. & Kumar, P. 2022 A global perspective of the current state of heavy metal contamination in road dust. *Environmental Science and Pollution Research* **29**, 33230–33251. <https://doi.org/10.1007/s11356-022-18583-7>.

- Ruban, V., Clozel, B., Conil, C. & Durand, D. 2003 Origine, caractérisation et gestion des boues de l'assainissement pluvial routier et urbain. Points sur les connaissances actuelles et perspectives In Bulletin des Laboratoires des Ponts et Chaussées 246, 117–126.
- Saby, N., Bertouy, B., Boulonne, L., Toutain, B., Caria, G. & Jolivet, C. 2020 Summary statistics of permanent organic pollutants concentration in French topsoils. <https://doi.org/10.15454/LQVMNN>.
- Saifur, S. & Gardner, C. M. 2021 Loading, transport, and treatment of emerging chemical and biological contaminants of concern in stormwater. *Water Science and Technology* **83**, 2863–2885. <https://doi.org/10.2166/wst.2021.187>.
- Sharley, D. J., Sharp, S. M., Marshall, S., Jeppe, K. & Pettigrove, V. J. 2017 Linking urban land use to pollutants in constructed wetlands: Implications for stormwater and urban planning. *Landscape and Urban Planning* **162**, 80–91. <https://doi.org/10.1016/j.landurbplan.2016.12.016>.
- Taguchi, V., Weiss, P., Gulliver, J., Kleiner, M., Hozalski, R., Baker, L., Finlay, J., Keeler, B. & Nieber, J. 2020 It is not easy being green: recognizing unintended consequences of green stormwater infrastructure. *Water* **12**, 522. <https://doi.org/10.3390/w12020522>.
- Tedoldi, D., Chebbo, G., Pierlot, D., Kovacs, Y. & Gromaire, M.-C. 2017 Assessment of metal and PAH profiles in SUDS soil based on an improved experimental procedure. *Journal of Environmental Management* **202**, 151–166. <https://doi.org/10.1016/j.jenvman.2017.06.063>.
- Tobiszewski, M. & Namieśnik, J. 2012 PAH diagnostic ratios for the identification of pollution emission sources. *Environmental Pollution* **162**, 110–119. <https://doi.org/10.1016/j.envpol.2011.10.025>.
- United Nations, Department of Economic and Social Affairs, Population Division 2019 *World Urbanization Prospects: The 2018 Revision (No. ST/ESA/SER.A/420)*. United Nations, New York.
- Weber, J. B., Wilkerson, G. G. & Reinhardt, C. F. 2004 Calculating pesticide sorption coefficients (Kd) using selected soil properties. *Chemosphere* **55**, 157–166. <https://doi.org/10.1016/j.chemosphere.2003.10.049>.
- Webster, L., Roose, P., Bersuder, P., Kotterman, M., Haarich, M. & Vorkamp, K. 2013 Determination of polychlorinated biphenyls (PCBs) in sediment and biota. <https://doi.org/10.17895/ICES.PUB.5078>.
- Weinstein, J. E., Crawford, K. D., Garner, T. R. & Flemming, A. J. 2010 Screening-level ecological and human health risk assessment of polycyclic aromatic hydrocarbons in stormwater detention pond sediments of Coastal South Carolina, USA. *Journal of Hazardous Materials* **178**, 906–916. <https://doi.org/10.1016/j.jhazmat.2010.02.024>.
- Whitley, E. & Ball, J. 2002 Statistics review 6: Nonparametric methods. *Critical Care* **6**, 509–513. <https://doi.org/10.1186/cc1820>.
- Wicke, D., Matzinger, A., Sonnenberg, H., Caradot, N., Schubert, R.-L., Dick, R., Heinzmann, B., Dünnbier, U., von Seggern, D. & Rouault, P. 2021 Micropollutants in urban stormwater runoff of different land uses. *Water* **13**, 1312. <https://doi.org/10.3390/w13091312>.
- Wiest, L., Baudot, R., Lafay, F., Bonjour, E., Becouze-Lareure, C., Aubin, J.-B., Jame, P., Barraud, S., Kouyi, G. L., Sébastien, C. & Vulliet, E. 2018 Priority substances in accumulated sediments in a stormwater detention basin from an industrial area. *Environmental Pollution* **243**, 1669–1678. <https://doi.org/10.1016/j.envpol.2018.09.138>.
- Yousef, Y. A., Hvitved-Jacobsen, T., Sloat, J. & Lindeman, W. 1994 Sediment accumulation in detention or retention ponds. *Science of the Total Environment, Highway Pollution* **146–147**, 451–456. <https://doi.org/10/ckmctd>.
- Analytical methods**
- DIN 38414-24 2000 German standard methods for the examination of water, waste water and sludge – Sludge and sediments (group S) – Part 24: Determination of polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofuranes (PCDFs) (S 24).
- FD T90-112 1998 Water quality – Determination of eight metallic elements (Mn, Fe, Co, Ni, Cu, Zn, Ag, Pb) by flame atomic absorption spectrometry – Direct and after complexing and extraction determination methods.
- ISO 10390 2005 Soil quality – Determination of pH.
- ISO 13320 2009 Particle size analysis – Laser diffraction methods.
- ISO 18287 2006 Soil quality – Determination of polycyclic aromatic hydrocarbons (PAH) – Gas chromatographic method with mass spectrometric detection (GC-MS).
- ISO/TS 13907 2012 Soil quality – Determination of nonylphenols (NP) and nonylphenol-mono- and diethoxylates – Method by gas chromatography with mass selective detection (GC-MS).
- NF EN 15169 2007 Characterization of waste – Determination of loss on ignition in waste, sludge and sediments.
- NF EN 16167 2018 Soil, treated biowaste and sludge – Determination of polychlorinated biphenyls (PCB) by gas chromatography with mass selective detection (GC-MS) and gas chromatography with electron-capture detection (GC-ECD).
- NF EN ISO 23470 2018 Soil quality – Determination of effective cation exchange capacity (CEC) and exchangeable cations using a hexamminecobalt(III)chloride solution.
- Dataset**
- Tedoldi, D., Couvidat, J., Chatain, V., Gautier, M., Bedell, J.-P., Lassabatere, L., Delolme, C. & Winiarski, T. 2023 Data for: 'In-depth characterization of sediment contamination in stormwater infiltration basins', Mendeley Data, V1, doi: 10.17632/dw4rrzhkft.1.

First received 28 July 2023; accepted in revised form 11 December 2023. Available online 22 December 2023