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Zirconium deficit as a tracer of urban sediment accumulation in Sustainable Urban Drainage Systems – Application to the calibration of a filtration model

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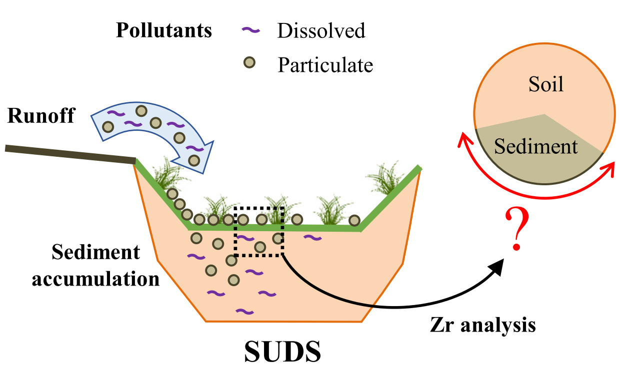
HIGHLIGHTS

- Method for the quantification of runoff-generated particles in the solid phase (soil samples)
- Horizontal and vertical sampling in 11 infiltration systems, analysis of Zr and metals
- Zr is immobile in soils, has few anthropogenic sources, and low levels in sediment
- Zr deficit in the most contaminated zone: "dilution" of the soil by sediment
- Sediment profiles: calibration of a filtration model without collecting effluent water

GRAPHICAL ABSTRACT



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ABSTRACT

Among the processes governing contaminant retention in soil-based *Sustainable Urban Drainage Systems* (SUDS), quantifying the relative contribution of particle settling and filtration requires a tracer of runoff-generated solids. Since zirconium (Zr) is a widely used geochemical invariant in pedological approaches, with few anthropogenic sources, the present investigation aims to assess whether its use may be extended to sediment identification in SUDS. High-resolution horizontal and vertical soil sampling was carried out in 11 infiltration systems, as well as in road-deposited sediment. Following elemental analysis *via* X-ray fluorescence spectrometry, the spatial distribution of both Zr and urban-derived metals could be determined. Zr content in sediment was found to be fairly stable and significantly lower than in soil. In most devices, Zr and metals exhibited "mirror" trends, both horizontally and vertically, *i.e.* a deficit of Zr could be observed in the most contaminated area. This indicated a "dilution-like" mixture of soil and sediment, the fraction of which could be calculated and appraised spatially. The vertical profiles proved the occurrence of bed filtration over 5 to 15 cm, and enabled the calibration of a simple filtration model. The uncertainties associated with the determined filter coefficient were found to be comparable to the other experimental methods – with the additional improvement that the present approach does not require water sampling.

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1. Introduction

Stormwater management close to the source, referred to variously as *Low Impact Development* (LID), *Best Management Practices* (BMPs), *Water Sensitive Urban Design* (WSUD), or *Sustainable Urban Drainage Systems* (SUDS), has gained popularity across the world (Fletcher et al., 2015), in order to mitigate the adverse effects of urban development and rising levels of impervious cover on the water cycle – namely, increased peak flows and annual volumes of runoff, faster hydrological response of the catchments, reduced infiltration and groundwater recharge (Fletcher et al., 2013; Miller et al., 2014), along with several qualitative impacts on the receiving water bodies (Hatt et al., 2004; McGrane, 2016). In addition to their widely recognized hydrologic and hydraulic benefits, these systems, and especially soil- or media-based devices, offer interesting perspectives towards the interception of diffuse pollutant fluxes in urban environments (Dierkes et al., 2015; Napier et al., 2009; Paus et al., 2013). On one hand, particle-bound contaminants in runoff are likely to be trapped with suspended solids *via* deposition and filtration, sometimes leading to the progressive formation of a sediment layer at the surface of infiltration systems (El-Mufleh et al., 2014; Erickson et al., 2013). On the other hand, dissolved species may undergo sorption onto various reactive constituents of the solid matrix (Sposito, 2008; Tedoldi et al., 2016). Since several contaminants such as trace metals are present in both dissolved and particulate forms in urban runoff, their accumulation in soil (or filter media) results from a combination of these physical and physico-chemical mechanisms.

Conventional experimental assessments of soil contamination in SUDS, which consist of collecting and analyzing soil samples after a known operation time, provide an overall and "time-integrated" vision of these retention processes (Kluge and Wessolek, 2012; Tedoldi et al., 2017a). However, they do not enable to differentiate between the relative contributions of settling/filtration and sorption, especially because runoff-generated solids – hereafter referred to as "sediment" – are not quantified *in the solid phase*.

As discussed by Clark and Pitt (2012), while infiltration practices are often designed according to empirical recommendations, and their treatment performance extrapolated from a limited number of field studies, a better understanding of the retention mechanisms taking place could valuably improve their choice, design and maintenance. Additionally, although soil clogging by runoff sediment has often been reported as a major concern in infiltration systems, characterizing and quantifying the state of clogging in a given device, or even anticipating the occurrence of hydraulic malfunction, remain complex objectives (Cannavo et al., 2018). Finally, if the long-term fate of contaminants in SUDS soil is to be appraised with a modelling approach, it seems necessary to consider the behavior of particle-bound species in addition to solute transport. Even if several filtration models are available for this purpose (Logan, 2001), few authors attempted to do so (Li and Davis, 2008a), possibly because of the difficulty to calibrate such models with usual field data (Tedoldi et al., 2016). To this end, the vertical distribution of filtered particles – provided it may be accurately determined – may constitute useful calibration data. All these points highlight the interest of identifying a tracer of urban sediment accumulation in SUDS.

The latter should (*i*) be conservative in a pedological acceptation, *i.e.* its content in soil should not be affected by runoff infiltration (neither *via* sorption nor *via* lixiviation), and (*ii*) display an anthropogenic signal different from the local background level in soils. The first point typically prevents the use of heavy metals such as copper or zinc, which are also present in the dissolved form in runoff (their average dissolved fractions reported by Huber et al. (2016) in road runoff were 38 and 31%, respectively) and have a strong affinity for several soil constituents (Tedoldi et al., 2016).

Due to their very low mobility in soils under almost all environmental conditions, resistance to weathering, and very low availability to plants, elements such as titanium (Ti) and zirconium (Zr) are generally considered as geochemical invariants (Kabata-Pendias, 2011; Schulz, 1965; Shahid et al., 2013); hence, they are commonly used as references to calculate enrichment or depletion factors of other elements (Egli and Fitze, 2000; Stockmann et al., 2016). However, Ti has been quantified at relatively high levels (>1 g/kg) in urban road dust (Apeagyei et al., 2011), and has numerous anthropogenic sources including paint pigments, car manufacturing, and metallic alloys (Salminen, 2005). Conversely, the anthropogenic sources of Zr are quite limited – nuclear fallout and ceramic dust being the most frequently cited ones (Salminen, 2005; Shahid et al., 2013) –, as a result of which the contents in road dust were found to be much lower (~200 mg/kg) (Apeagyei et al., 2011). It was observed that the presence of a dense urban area did not significantly impact Zr concentrations in both water and sediment from an urban stream (Mohiuddin et al., 2010). Furthermore, the lithogenic sources of Zr and its consequent abundance in most soils enable a systematic quantification with analytical methods such as X-ray fluorescence (Kabata-Pendias, 2011), thus providing the opportunity to extend the number of analyses and to achieve a finer description of its spatial variability.

Therefore, the objective of the present work is to assess the potential use of Zr as an unequivocal tracer of sediment accumulation in SUDS, and to illustrate a possible application with the calibration of a simple filtration model. The approach relies on high-resolution horizontal and vertical soil sampling from 11 infiltration systems, and subsequent analysis of both Zr and three typical urban- and traffic-derived trace metals (Cu, Pb, and Zn). The latter, which are ubiquitous in stormwater runoff (Göbel et al., 2007; Huber et al., 2016), will be considered as a signature of runoff-induced contamination, and used as a comparison basis.

2. Materials & methods

2.1. Description of the study sites

Four small sized infiltration basins, five swales, and two grassed filter strips (Table 1), located in the Paris region (Fig. 1), were selected for their contrasting hydraulic configurations, soil types, watersheds, and runoff contamination potentials (Tedoldi et al., 2017a). Inflow of water into the infiltration systems consists of either an inlet pipe (*Dourdan*, *Greffiere*, *Alfortville*, *Vaucresson*), or surface runoff directly flowing from the pavement, with a localized (*Sausset2*) or diffuse inflow (*Sausset1*, *Chanteraines*, *Vitry*, *Compans1*, *Compans2*, *Compans3*). In some devices, superficial outflow is possible in addition to infiltration: in the case of *Dourdan*, *Chanteraines*, and *Vitry*, this occurs when water ponding exceeds a given level, whereas in *Compans1* and *2*, water storage is supposedly achieved in a downstream longitudinal ditch; however, previous studies have shown that most water infiltrates in the roadside filter strips (Flanagan et al., 2017). *Compans3* is equipped with a V notch weir.

In *Chanteraines*, *Vitry*, *Compans1* and *2*, the surface soil displays a 5 to 15% slope perpendicular to the pavement; *Alfortville* and *Compans3* have a V-shaped transversal section; the rest of the facilities were constructed with flat bottoms and sharp embankments. *Chanteraines*, *Vitry*, and *Compans2* were excavated and backfilled with topsoil from another site during construction, as were *Compans1* and *3* with an engineered filter media (mixture of calcareous sand and loamy soil). The other systems were implemented upon the preexisting soil.

2.2. Sampling procedure

The field investigations were undertaken in two phases between April 2015 and May 2016, so as to consecutively obtain the distribution of the chemical species of interest at the surface and along vertical profiles (Fig. 2). Firstly, a systematic sampling of the surface horizon was carried out using a rectangular grid, with a mesh size adapted to each site so as to meet the strictest of these two criteria: (*i*) collect ≥20 samples per device, and (*ii*) collect ≥35 samples/100 m^2. At each node, the