



Building an isotopic hydrogeochemical indicator of anthropogenic pressure on urban rivers

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ABSTRACT

This study provides evidence on the sources and pathways of trace-metal contamination using isotopic tracers, $\delta^{34}\text{S}$ of dissolved sulfate and $^{206}\text{Pb}/^{207}\text{Pb}$ of suspended particulate matter (SPM), in an urban French river. The trace-metal contamination of the Orge River watershed was correlated with a highly contrasted land-use pattern with mainly forests and agricultural lands upstream and a highly dense urban area (up to 8000 inhabitants/km²) downstream in the suburbs of the Parisian megacity. The increase in the sulfate concentrations (0.25–1 mmol/l) of the dissolved compartment (<0.45 μm) monitored during a hydrological year (2010/2011) correlated with the dissolved sulfate $\delta^{34}\text{S}$ and $\delta^{18}\text{O}$ ratios (+2 to +12 and +6 to +13‰, respectively). This result indicated that the runoff and sewage waters are important sources of water in the river, particularly for the highly urbanized sites. The lead enrichment factor in the SPM was observed to increase according to the urbanization gradient (1 to 6), and the associated $^{206}\text{Pb}/^{207}\text{Pb}$ ratio indicated that the signature of the lead-bearing particles transformed from a three-endmember system (2001 samples) to a two-endmember one (2010/2011 samples) over the last decade. These results suggest that gasoline-originated lead has disappeared from particulate fluxes since the lead additive prohibition in 2000. The lead-bearing species in the Orge River were also observed to originate mainly from urban activities, and the reactivity of selected urban lead-bearing species is discussed based on microscopic observations. Finally, coupling the $\delta^{34}\text{S}$ and $^{206}\text{Pb}/^{207}\text{Pb}$ ratios provided a powerful indicator of urban influence as a function of both river hydrodynamics and physico-chemical conditions.

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1. Introduction

Urban sprawl has led to the release of various contaminants into the environment, including trace metals, which may induce perturbations in ecosystems (Wong et al., 2006). In densely urbanized areas, urban rivers are the main outlet for contaminants collected from runoff on non-permeable surfaces and hydrodynamic transport. In a watershed, the frequent mixing of multiple contamination sources makes it difficult to determine the origin of the contaminants. To overcome this obstacle, specific isotopic tracers can be used to characterize both the sources and pathways of contamination, thus defining the degree of anthropogenic influence on different temporal and spatial scales.

In urban environments, processes such as bacterial development or precipitation/dissolution reactions involve sulfur compounds, mainly sulfide or sulfate (Garrels and Thompson, 1960; Szykiewicz et al., 2011; Priadi et al., 2012). In rivers, variations in the dissolved sulfate

concentration result from atmospheric inputs (Novak et al., 2000), rock weathering reactions, in particular *via* oxidation of sulfide minerals (Hitchon and Krouse, 1972; Robinson and Bottrell, 1997; Moncaster et al., 2000), dissolution of sulfate evaporites (Otero et al., 2008), biological activities (Kaplan and Rittenberg, 1964; Pisapia et al., 2007), and anthropogenic sources such as fertilizers (Brenot et al., 2007). Precipitation/dissolution reactions occurring in the water column, which affect the sulfur-containing dissolved and solid compounds, induce fractionation between sulfur isotopes (Seal, 2006). Consequently, the origin of surface water bodies can be discriminated and traced using the $\delta^{34}\text{S}$ ratio (Bottrell et al., 2008). Thus, the stable isotope composition of these compounds is a key tracer of specific reactions (Trust and Fry, 1992; Heidel et al., 2011).

Within the water column, inorganic contaminants are often bound to suspended particulate matter (SPM) of either natural or “urban” origin (Owens and Walling, 2003; Collins et al., 2005; Audry et al., 2006; Priadi et al., 2011). The contaminants are then transported by the water, inducing a potential availability and toxicity for the aquatic ecosystem. To study the origin of contaminant-bearing particles in urban rivers, the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio is a powerful tracer. Indeed, lead is a common contaminant, known for its abundance in urban environments.

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In addition, it is also a potentially harmful element. In environmental sciences, lead stable isotopes have been widely used (Komárek et al., 2008), particularly as source tracers in soils (e.g., Emmanuel and Erel, 2002), sediments, or the atmosphere (e.g., Carignan et al., 2005). Particulate isotopic lead is thus recognized as an indicator of urban pressure on rivers (Duzgoren-Aydin, 2007; Ayrault et al., 2012; Négrel and Petelet-Giraud, 2012). Consequently, the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio allows particle fluxes to be discriminated as a function of their natural or anthropogenic origin.

In this study, the behavior of lead and sulfur isotopic tracers ($^{206}\text{Pb}/^{207}\text{Pb}$ and $\delta^{34}\text{S}$, respectively) was monitored in the Orge River catchment, which is located in the upper part of the hugely anthropized Seine River watershed (North-western France). Coupling the analyses of $^{206}\text{Pb}/^{207}\text{Pb}$ ratio and $\delta^{34}\text{S}$, which are tracers of SPM and water bodies, respectively, represents an original approach. To the best of our knowledge, this strategy has been used previously but only on the particulate component (Yang et al., 2010) to trace the origin of soils and sediments. Samples were collected along

the Orge River, including two of its tributaries, to examine the spatio-temporal trends of urban contamination in this watershed. The approach developed here aimed to accurately identify the river watershed areas that are the most exposed to anthropogenic pressure, in association with specific hydrogeochemical contexts, providing information on contamination sources for both dissolved and particulate components. This objective was achieved by building an isotopic hydrogeochemical indicator of urban contamination, based on the monitoring of $^{206}\text{Pb}/^{207}\text{Pb}$ and $\delta^{34}\text{S}$ in the dissolved and SPM compartments, respectively in the Orge River.

2. Material and methods

2.1. Location

Details about the sampling procedures and site characteristics were provided in Le Pape et al. (2012), and location of the study site is provided in Fig. 1. The sampling locations along the Orge

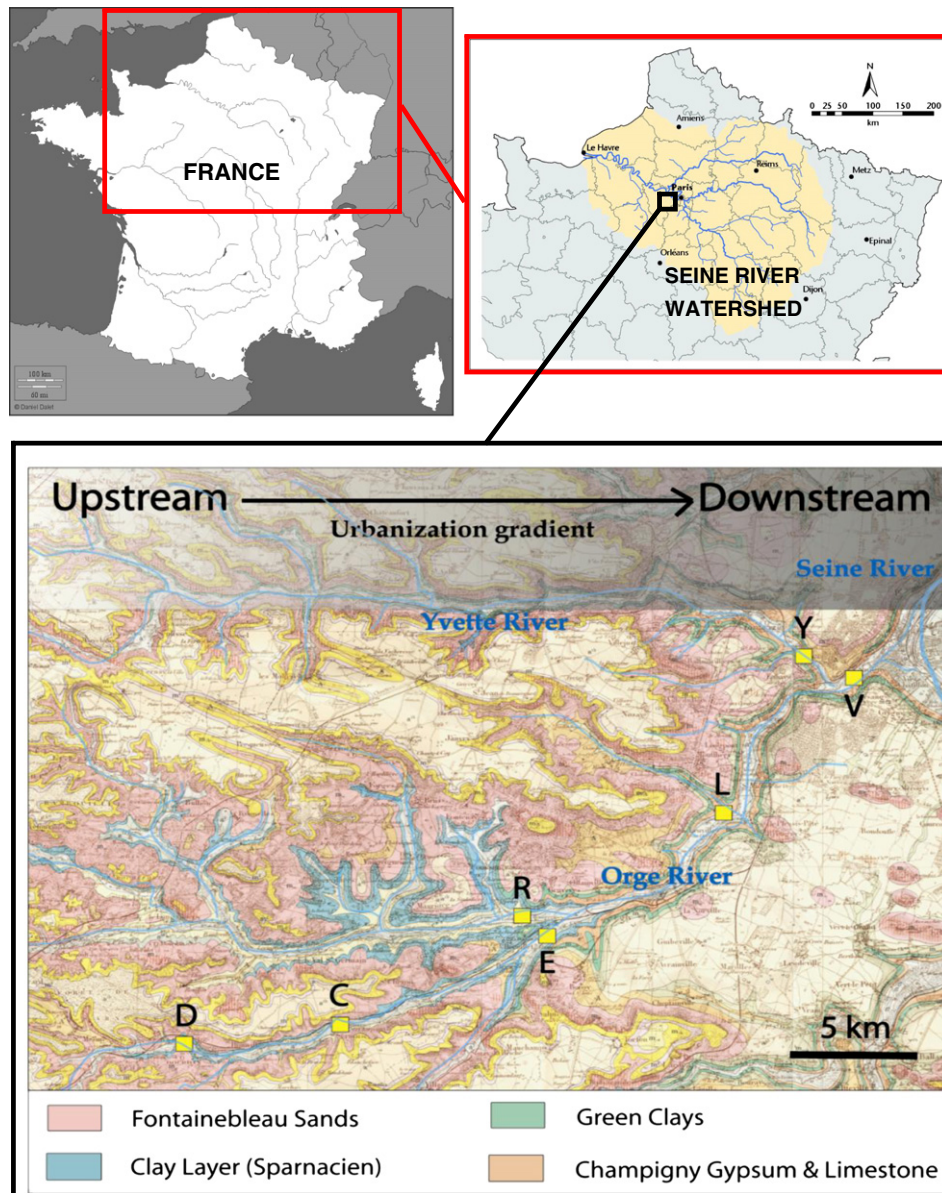


Fig. 1. Geological map of the Orge River catchment with the locations of the sampling sites (from Dollfus et al., 1968, modified). "D" stands for Dourdan, "C" stands for St Chéron, "R" stands for Rémarde, "E" stands for Egly, "L" stands for Longpont-sur-Orge, "Y" stands for "Yvette", "V" stands for Viry-Chatillon.

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