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Record of pharmaceutical products in river sediments: A powerful tool to assess the environmental impact of urban management?

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ABSTRACT

The occurrence of eight pharmaceutical products (PPs) from various therapeutic classes was studied in urban sediments collected upstream a small dam in Orleans, France. Since PPs are globally distributed contaminants since the 1950s, their spatial and historical distribution was documented in order to better understand the impact of recent urban management on the chemical quality of sediments. Concentrations of the PPs mainly ranged between 1 and 10 ng g⁻¹ within the two cores. The chronology of core LOI13-2, based on radionuclide analysis (137 Cs and 210 Pb), enabled the characterization of the changes or shift in PPs over the last 50 years, impacted by the effluent deflections i) of a large wastewater treatment plant in 1989, and ii) of 3 smaller wastewater treatment plants between 2003 and 2009. In most cases the deepest occurrence of each PP in the core matched the market authorization date of different pharmaceuticals, indicating that some PPs can be used as chronomarkers. The study reveals that recent management of effluent discharges within the watershed improved the chemical quality of these sediments. In view of the persistence of PP pollution in trapped sediments, a total stock of 763 ± 565 g for the selected PPs was estimated at the millstream scale. Superficial sediments therefore represent a potential source of downstream pollution in the event of the removal of a small existing dam.

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1. Introduction

The growing use of pharmaceutical products (PPs) for human and animal therapies since the 1950s raises concerns about the environmental fate of these products and their metabolites within the aquatic environment because of their non-negligible excretion via urine and faeces (Daughton and Ternes, 1999; Lienert et al., 2007; Farré et al., 2008). Moreover, because they are exclusively generated by human activities, PPs could provide unique stratigraphic markers to allow the assessment and dating of anthropogenic impacts on the environment in a similar manner to how microplastics, synthetic fibers or pesticides have been used (Waters et al., 2016; Zalasiewicz et al., 2016). Nevertheless, the occurrence of PPs is under reported within sedimentary archives

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http://dx.doi.org/10.1016/j.ancene.2017.05.006 2213-3054/© 2017 Elsevier Ltd. All rights reserved. (Bernhardt et al., 2017), despite their significant concentrations in numerous water compartments. The question about their longterm fate also represents a challenge for both an effective water resource management (Loos et al., 2009; Mompelat et al., 2009; Petrie et al., 2015) and a better tracking of the impacts of human activities on the environment.

Pharmaceutical products contain a wide variety of organic compounds (i.e. more than 3000 in France according to the French National Agency for Medicines and Health Products Safety, ANSM, 2014). These pharmaceuticals are designed to prevent and treat various diseases and to improve health in general. The main sources of PPs are domestic, hospital and industrial wastewaters (specifically pharmaceutical plants) that reach water bodies through sewage systems (Fick et al., 2009; Verlicchi et al., 2010) due to the inefficiency of classical or advanced wastewater treatment plants (WTP) to totally remove PPs (Busetti et al., 2015; Petrie et al., 2015). Accordingly, PPs enter the natural environment and potentially cause severe ecological issues and/or







affect human health. Widespread contamination has been observed in surface, marine and ground waters (e.g. Heberer, 2002; McEneff et al., 2014; Lopez et al., 2015) as well as soils and sediments (e.g. Da Silva et al., 2011; Tamtam et al., 2011; Vazquez-Roig et al., 2012). Furthermore, the early studies related to the ecotoxicity of PPs at field-relevant concentrations have demonstrated that these pollutants are harmful to both invertebrates and vertebrates, even in low or natural concentrations (Brodin et al., 2013; De Castro-Català et al., 2016; Välitalo et al., 2017).

Beyond their hydrological impact on rivers (Hubbard et al., 2016), WTPs contribute a constant discharge of PPs in natural waters. Nevertheless, this flux is not random. For example, Klaminder et al. (2015) demonstrated a good correlation between the effluent and the downstream sediment concentrations of oxazepam (an anxiolytic drug), indicating that the impact of WTPs on the environment can be monitored (i) by the effluent concentration, and (ii) since the market authorization date of each compound.

The occurrence of PPs is closely related to the worldwide increase in population and the ongoing urbanization since World War II and therefore represents a major environmental issue for urban ecosystems (Heberer, 2002; Taylor and Owens, 2009). While numerous studies have been published concerning the occurrence of PPs in natural waters, there is still a lack of research on the spatial and temporal trends of this widespread aquatic pollution in sediments (Petrie et al., 2015). Sediments are however a relevant compartment for this type of tracking thanks to their ability to record the evolution of contamination over several decades and beyond (Tamtam et al., 2011; Dubois and Jacob, 2016; Lorgeoux et al., 2016). Furthermore, PPs can be considered as very reactive contaminants, exhibiting some affinity for both organic and inorganic surfaces (Stein et al., 2008; Zhou and Broodbank, 2014; Thiebault et al., 2016a,b). Moreover, several PPs have recently been added to the list of priority substances in the amended Water Framework Directive of the European Union (European Commission, 2013), indicating that PPs are now considered as a potential threat for various organisms, including humans (De Jongh et al., 2012). Finally, an assessment of the stock of contaminated sediments accumulated in small ponds and reservoirs should be considered since the Water Framework Directive (European Commission, 2000) has recommended the removal of dams without any practical function in order to improve the ecological continuity of superficial water bodies. Such an assessment is required to prevent the potential downstream propagation of PPs induced by physical modifications within the reservoir such as dredging or removal, or by chemical changes in the water column.

In view of the potential hazards of these PPs and the need to acquire data to assess and date the impact of anthropogenic activities on the environment, eight PPs were selected among the most frequently detected compounds in EU waters (Tamtam et al., 2008; Loos et al., 2009; Joigneaux, 2011), namely acetaminophen, atenolol, codeine, metoprolol, oxazepam, ofloxacin, trimethoprim and sulfamethoxazole. In the study presented here, the presence of PPs in sediments collected in a small millstream located in the urban area of Orleans, France, was investigated. This work aimed to (i) quantify PPs within a sedimentary archive to assess their potential as new chronomarkers; (ii) establish the long-term chronology of PP contamination related to the evolution of WTPs in the catchment in order to evaluate the link between the discharge

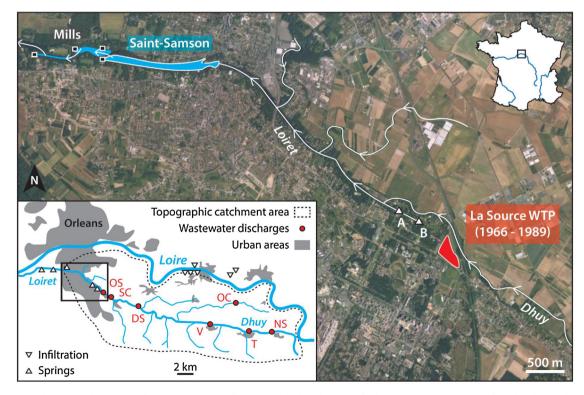


Fig. 1. Aerial photograph showing the location of the Saint-Samson millstream within the urban area of Orleans (www.geoportail.gouv.fr). White lines indicate rivers. A = Les Abîmes spring; B = Le Bouillon spring. The inset illustrates the hydrographic context of the Loire karst upstream of Orleans, with groundwater circulation between infiltrations and springs. The effluent inputs from WTPs to the Dhug River are marked by red dots: OS = Orleans-la-Source - SC = Saint-Cyr-en-Val - DS = Darvoy and Sandillon - OC = Ouvrouer-les-Champs - V = Vienne-en-Val - T = Tigy - NS = Neuvy-en-Sullias (see Table 1).

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