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Sediment-based epidemiology, an utopia?

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Highlights

- Decantation tank sediments record a large diversity of organic contaminants.
- The quantitative link between use and sediment concentration must be understood.
- Ionization state is an important driving factor of the sedimentary occurrence.

Introduction

The occurrence of drug target residues (DTRs) of pharmaceutically active compounds and illicit drugs in numerous environmental compartments raises serious concerns about their potential hazard for living beings (Brodin et al., 2013; Richmond et al., 2018). Hence, a significant part of the current literature focuses on both assessing the removal of organic contaminants in classical wastewater treatment plants (e.g. Thiebault et al., 2017), and on optimizing their removal based on tertiary treatments (e.g. Altmann et al., 2015).

However, wastewater treatment plant influents (WWI) could also represent a tremendous source of information, due to the link between their chemical composition and the excretion of various chemical compounds by the population in the catchment (Castiglioni et al., 2014; Choi et al., 2018). Several studies have demonstrated the consistency of such analyses on illicit drugs and pharmaceutically active compounds in order to back-calculate the consumption of the population (e.g. Baker et al., 2014). This new scientific discipline has been called wastewater-based epidemiology and currently investigates not only chemicals consumed as pharmaceuticals, but also passive exposure to other markers such as industrial chemicals and pyrethroid pesticides (Lopardo et al., 2019; Rousis et al., 2017) among many examples. Such studies provide near real time dynamic of practices whereas the assessment of past uses is not accessible. Yet, as performed in environmental sedimentary archives, sediment decantation tank are present in urban sewer system, collecting mostly coarse particles considered as not favorable for recording organic contaminants as DTRs.

In this study, we analyzed the occurrence of DTRs in a decantation tank with an initial objective to perform sediment-based epidemiology, considering that decantation tank are close to the excretion sources, before the WWTP removal effect, and therefore potentially record a wide diversity of DTRs. Several challenges accompanied the study of such archives, among which the dating and the type of recorded sediments.

Methodology

The selected site is a decantation tank that collects wastewater and stormwater from the sewer network of the northern part of Orléans (France). This 20 m deep underground building of cylindrical and conical shape was constructed in 1942 and was reputed to have not been subjected to any clean out operation (COO) since that time. This constitutes unique opportunity to study the evolution of a large variety of thematic involved in urban metabolism such as nutrition, health, transport, energy, practises, biodiversity through organic molecules monitoring. Six cores were acquired from 2014 to 2017 in the 14 m depth sedimentary series. Five cores were retrieved by manually inserting a plastic tube in the outcropping sediments or with a

gravity corer prior to COO. One core was obtained in the oldest and deepest sediments that remained after partial COO.

All the 14 m sediments accumulated in the tank were collected by this mean. We focused our analyses on core CSA-02/2015-A that afforded the longest record, and two other cores with more limited record. After sampling, 152 sediment samples were directly extracted through accelerated solvent extraction with a mixture of water:methanol (1:1) in order to obtain concentration of 27 DTRs that were then quantified by HPLC-MS². Extracted sediments then underwent a large range of classical sedimentological analyses (granulometry, facies description, microscopic description, mineralogy) and Total Organic Carbon, Hydrogen Index, Oxygen Index and T_{max} were determined by Rock-Eval pyrolysis. The age model is based on 54 ¹⁴C dates obtained on the 6 cores and computed with Chronomodel using a modified Bomb13NH1 calibration curve to obtain a robust age model

Results and discussion

Oldest sediments are dated back to 1970. This reveals that COO certainly took place at the end of the seventies, with no administrative or written record subsisting. Sedimentation rates are then very high with 6 m deposited in two years, which corresponds to accumulation rates observed after the 2015 COO. Then, sedimentation rates slowed down with only 2.5 m accumulated from 1980 to 2014 because the decantation tank was saturated and could not accumulate more sediments. Hence, repeated episodes of erosion compensated by deposition are suspected over that period. Sediments are stratigraphically organised into organic (TOC > 2%) and mineral (TOC < 2%) layers. During rain episodes, roads and sidewalks are washed and bring coarse and mineral particles to the tank whereas during dry days, sediment is essentially built from organic materials carried by the wastewater (Jacob et al., 2020). Other markers identified in the sediments confirm these assertions. Glass microsphere that originate from road paintings are abundant in mineral sediments and rare in organic sediments (Jacob et al., 2020). Reversely and evidently, bile acids are rather concentrated in organic sediments and diluted in mineral sediments. The picture is different for drugs and illicit drugs.

DTRs were mostly detected within organic layers, whereas some of them, as cocaine, were exclusively detected in mineral layers. It therefore appears that the organo-mineral characteristics of the sediments strongly control the adsorption capacity of DTRs, screening the potential for consumption back-calculation. It is necessary to understand the respective impact of organic content and mineral contribution to the sorption before using such archives for sediment based epidemiology. Our results show a strong control of speciation on the occurrence and concentration of DTRs (Thiebault et al., 2021b).

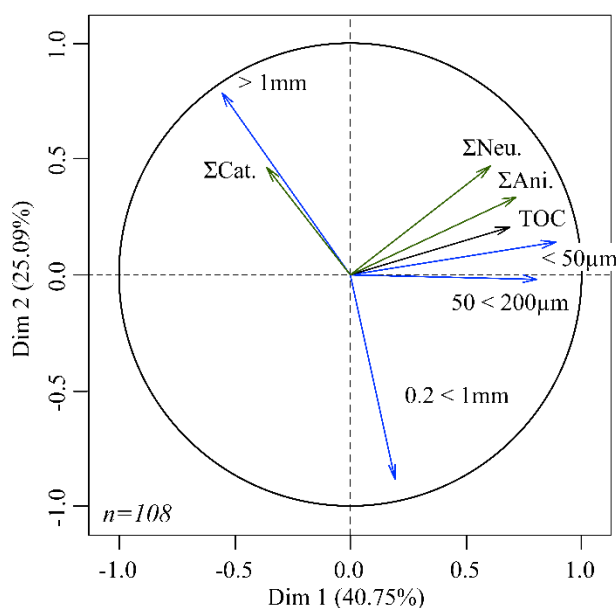


Figure 1: Principal component analysis performed with the sum of the concentrations of cationic ($\Sigma\text{Cat.}$), anionic ($\Sigma\text{Ani.}$) and neutral ($\Sigma\text{Neu.}$) licit and illicit drugs, the grain size (>1mm, 0.2<1mm, 50<200 μm and <50 μm respectively) and the total organic carbon (TOC).

Neutral and anionic compounds (e.g. carbamazepine, salicylic acid) present higher concentrations in the lower part of the core whereas cationic compounds (e.g. codeine, benzoylcegonine) display higher concentrations in the upper part. For the formers, their distributions are directly correlated with the distribution of organic carbon, whereas for the latter, a correlation with the relative abundance of inorganic surfaces is evidenced (Figure 1). As a result, the interaction mechanisms between targeted compounds and the sedimentary composition appears to be the most important control of their occurrences (Figure 2).

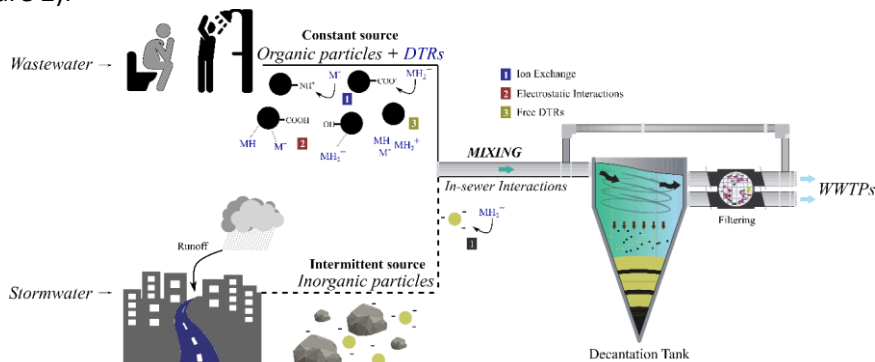


Figure 2: Schematic representation of the origin and possible reactivity of solids sampled in the decantation tank. Beyond the consumption back-calculation, such archive displays the potential to record the first use of such molecule within the population. To do so, specific dating through Chronomodel was performed and allowed to date the first occurrence of various DTRs and compare it to their market authorization date (MAD), as realized within riverine or lacustrine sedimentary archives (Thiebault et al., 2021a) with a wider diversity of DTRs. The example of methadone, which was never detected in riverine sediments, is provided with a good fit between first occurrence and MAD in France in 1995 (Figure 3).

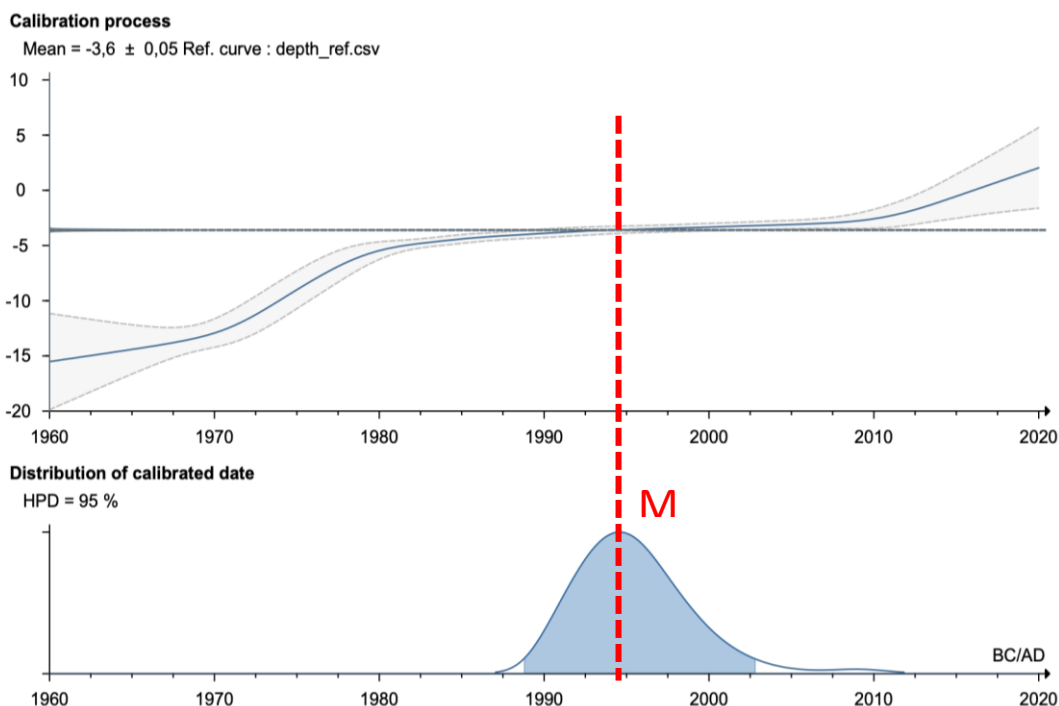


Figure 3: Comparison between first occurrence of methadone and sedimentary dating based on Chronomodel

Conclusions and future work

In this study, the sampled sediments were very close to the primary source of DTRs (i.e. households), making it possible to detect a wider variety of compounds such as, for the first time, illicit drugs compared to riverine or lacustrine sediments, in which only highly concentrated or persistent DTRs can be detected. This wider DTR diversity emphasized the potential of such sediment for the reconstruction of the

consumption/excretion history. However, due to sudden vertical variations in the composition of the sedimentary layers that are attributed to respective inputs of wastewater and stormwater materials depending on meteorological events, the affinity of DTRs with sediments was obviously impacted as well. Specific vertical distributions of DTRs according to their molecular charge were revealed by statistical analyses and evaluated based on general knowledge of the adsorption mechanisms of organic contaminants onto organic and inorganic surfaces. As a result, before expecting a complete understanding of the historical excretion within this catchment based on sedimentary accumulations in sewer networks, several corrections have to be performed on these vertical distributions, based on the affinity between DTRs and sediments/suspended solids.

However, it was possible to date the first use of each DTR based on sediment dating through ¹⁴C assessment and adapted model, which is an important information related to the time between MAD and citizen use.

Finally, although this archive displays the first distribution pattern for most DTRs, further work remains necessary in order to better understand the historical deposition dynamics and potential local remobilization.

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