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Sewer sediment-bound antibiotics as a potential environmental risk: Adsorption and desorption affinity of 14 antibiotics and one metabolite☆

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ABSTRACT

In this study, 14 antibiotics and one metabolite were determined in sewages and size-dependent sewer sediments at three sampling sites in the city of Dresden, Germany. Adsorption and desorption experiments were conducted with fractionated sediments. All antibiotics and the metabolite investigated were determined in the sewages; 9 of 14 antibiotics and the metabolite were adsorbed to sewer sediments. The adsorbed antibiotic loads in ng of antibiotic per g of sediment correlated with antibiotic concentrations in ng of antibiotic per litre of sewage. The size fractions <63 μm, 63–100 μm and 100–200 μm had significantly higher loads of adsorbed antibiotics than bigger size fractions. In general, the adsorbed load decreased with an increasing size fraction, but size fractions >200 μm had similar levels of adsorbed antibiotic loads. An antibiotic-specific adsorption coefficient, normalized to organic content, was calculated: four antibiotics exceeded 10.0 L g^{-1}, three antibiotics fell below 1.0 L g^{-1} and all residual antibiotics and the metabolite were in the range of 1.0–10.0 L g^{-1}. The adsorbed antibiotic load and the organic matter increased with time, generally. The mineral composition had a minor effect on the adsorption coefficients. Desorption dynamics of five antibiotics and the metabolite were quantified. Regardless of the size fraction, the predominant part of the equilibrium antibiotic concentration was desorbed after 10 min. The calculated desorption distribution coefficient indicated adsorption as irreversible at the pH investigated (7.5 ± 0.5).

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1. Introduction

In the modern life of humans, antibiotics are widely prescribed for therapy against bacterial diseases to prevent the onset of bacterial infections during a viral disease (Goossens et al., 2005; Van Boeckel et al., 2014). They have been massively administered and persist in the environment (Carvalho and Santos, 2016). Their potential to promote antibiotic resistance genes (ARGs) and bacteria (ARB) is a major public-health problem worldwide (Berendonk et al., 2015). Antibiotics and ARGs have been continuously detected in the aquatic environment (Gao et al., 2018; Kümmerer, 2009a; b; Xu et al., 2016). In urban systems, the main anthropogenic source of antibiotics is human excretion. In particular, antibiotics prescribed for humans are partly metabolized in the human body and enter the sewage system via excreted urine and faeces. In Germany, 70% of the antibiotics consumed is excreted unchanged (Kümmerer, 2009a). Sewers have been regarded as one of the most important sinks for antibiotics, ARGs and ARB (Auguet et al., 2017; Wunder et al., 2011). Although several studies have reported adsorption (Guo et al., 2017; Hou et al., 2010; Maier and Tjeerdema, 2018; Pan et al., 2012; Wang et al., 2017c) and desorption behaviour of antibiotics in soil sciences (D'Angelo and Starnes, 2016; Fernandez-

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Calvino et al., 2015; Li and Zhang, 2016, 2017; Li et al., 2015; Wu et al., 2013), or adsorption in sludge sediments at wastewater treatment plants (WWTPs) (Marx et al., 2015a; Polesel et al. 2015, 2016; Wang et al., 2017a), reports about the adsorption/desorption kinetics of sewer sediment-bound antibiotics are scarce.

Although most wastewater is drained into WWTPs, conventional WWTPs are not sufficient to prevent the release of antibiotics into adjacent surface waters (Menz et al., 2017; Michael et al., 2013; Wang et al., 2017b). In addition, the discharge of wastewater, a composite of sewage and stormwater, through combined sewer overflow (CSO) structures into receiving waters is inevitable due to the capacity limitations of urban drainage systems. Consequently, urban drainage discharge contains dissolved and particulate-bound antibiotics. Dissolved antibiotics adsorb into the water environment, particulate-bound antibiotics are remobilized when a certain shear stress is exceeded and desorption of adsorbed antibiotics depends mainly on the pH values in the surrounding water body. Decryption of these complex nexuses is a big step in tackling the promotion and spread of antibiotic resistance in the environment.

Furthermore, particle size distribution is a crucial physical characteristic of particulate compounds (Zhang et al. 2015a, 2015b). In terms of sewer sediment, particle size distribution is related to fluid transport theory (Bridge, 2009; Xu et al., 2018). Finer particles can stay in suspension longer and they are transported farther by runoff than larger particles (McKenzie et al., 2008). Additionally, the literature on contaminants suggests that there is particle size control of metal adsorption/desorption (Zhang et al., 2016); reports on antibiotics are missing.

Consequently, this study focuses on the adsorption/desorption affinity of 14 antibiotics and one metabolite for particulate components in urban drainage systems. To the best of the authors' knowledge, this is the first look at sewer sediment-bound antibiotics from a stormwater pollution aspect. The following investigations were conducted to (i) quantify the antibiotic concentrations in sewage and size-fractionated sewer sediments; (ii) define antibiotic-specific adsorption coefficients, which consider organic material content; (iii) quantify portions of antibiotics which desorb into the water phase; (iv) quantify antibiotic-specific desorption coefficients and (v) identify the physicochemical factors related to the adsorption and desorption phenomena.

2. Materials and methods

2.1. Study area

The city of Dresden is the hometown of about 540,000 inhabitants and is located in the eastern part of Germany. The central wastewater treatment plant (WWTP) has a design capacity of 740,000 population equivalents with a current load of approximately 650,000 PE. A 20% portion is from Freital, Heidenau, Pirna, eastern Radebeul and some bordering municipalities, which drain their wastewater into the sewer system of Dresden. Furthermore, the catchment of the central WWTP includes 14 hospitals (11 in the city of Dresden). Hence, antibiotics prescribed for outpatients and inpatients are present in the sewage (Marx et al., 2015b). About 64% of the city's sewer network is operated as a combined sewer system – sewage and stormwater are drained in the same pipe. The first sampling site (LOC#1) was located in the southwest of Dresden (N 51°04′33.1″, E 13°40′01.2″) with a clear sedimentation effect due to the backwater phenomenon. The second sampling site (LOC#2) was the grit chamber upstream of a culvert (N 51°03′46.7″, E 13°41′28.2″) with approximately two-thirds of the sewage load of the WWTP. The last site (LOC#3) was the grit chamber of the WWTP (N 51°04′15.6″, E 13°40′51.1″).

2.2. Sewer sediment and sewage sample collection

We collected sewer sediments and wastewater samples at all three locations (LOC#1, LOC#2 and LOC#3). The sediments at LOC#1 were collected twice – the first sediment (SED#1A) the day after a precipitation event and the second (SED#1B) after 20 antecedent dry-weather days – with a self-designed two-weir system in combination with a bypass for a normal 800/1200 ovoid cross-sectional sewer profile (**SI** Fig. S1). The weir partly blocks the profile during dry-weather flow, and sewage drains through the bypass; thus, no water enters the collection area. The sediments at LOC#2 (SED#2) and LOC#3 (SED#3) were excavated with a clamshell bucket grab. The sewage samples at LOC#1 and LOC#2 were collected using an Epic 1511 automatic sampler (BÜHLER MONTEC) configured for a sampling frequency of 10 min, and an ASP Station 2000 automatic sampler (ENDRESS + HAUSER) with flow proportional sampling was used at LOC#3. The flow was monitored with a DM 43 F electromagnetic flowmeter (ABB AUTOMATION PRODUCTS GMBH). The sewage samples from LOC#1 and LOC#2 were mixed to make 24-h composite samples and were delivered daily to the lab. The sewage samples from LOC#3 were stored at 4 °C in brown glass flasks for a maximum of 7 days until laboratory analysis, following the storage recommendation from a previous study (Marx et al., 2015c).

2.3. Sediment fractionation

The collected sediment samples were separated into seven particle size ranges. Particle sizes smaller than 63 μm were separated by wet sieving, and particle sizes of 63–100, 100–200, 200–400, 400–630, 630–1000 and 1000–2000 μm were separated by dry sieving according to German Norm DIN 18123 (2011). The duration of the dry sieving was 10 min with 0.4 revolutions per second.

2.4. Antibiotic determination in sewage and sediment

The antibiotic groups were selected according to the global prescription report of Van Boeckel et al. (2014). The most-prescribed antibiotics within these groups were selected according to one of the largest statutory health insurance companies in Germany (Allgemeine Ortskrankenkasse, AOK PLUS). As shown in Table 1, in total, 14 antibiotics and one human metabolism product were analysed by solid phase extraction (SPE) and LC-MS/MS according to Rossmann et al. (2014). Briefly, a 50 mL aliquot of a homogeneous sewage sample was filtered through a glass fibre filter (<0.9 μm; WICOM, Heppenheim, Germany). The samples were adjusted to a pH of 3.5 (±0.2) with formic acid (LC-MS grade; Sigma, St. Louis, MO, USA). Then, 2.5 mL of prepared water was extracted by solid phase extraction (SPE) onto a 30 mg Oasis HLB VacCartidge (Waters, Milford, MA, USA) using a Gilson ASPEC XL Automatic Sample Processor (Middleton, WI, USA). The extracts were analysed using a LC-MS/MS system. Chromatographic separation was performed with a Synergi Hydro RP 80A 4 μm column (150 mm × 2.0 mm) and a Security Guard cartridge for C18 HPLC columns with a 4 mm × 2 mm internal diameter (Phenomenex, Aschaffenburg, Germany). A 100 mm × 3 mm Nucleoshell HILIC 2.7 μm column (Machery-Nagel, Düren, Germany) was additionally used for the determination of amoxicillin, ciprofloxacin, doxycycline and levofloxacin. An API 4000 tandem mass spectrometer (ABSciex, Framingham, MA, USA) was equipped with an electrospray interface (ESI) in multiple reaction monitoring mode (MRM). The limit of detection (LoD) was in the range from 0.2 ng L^{-1} (azithromycin) to 29.7 ng L^{-1} (cefuroxime); the limit of quantification (LoQ) ranged between 0.8 ng L^{-1} (azithromycin) to 99.0 ng L^{-1}

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