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## Contribution of combined sewer overflows to trace contaminant loads in urban streams

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#### ABSTRACT

The present study examines the contribution of combined sewer overflows (CSO) to loads and concentrations of trace contaminants in receiving surface water. A simple method to assess the ratio of CSO to wastewater treatment plant (WWTP) effluents was applied to the urban River Spree in Berlin, Germany. The assessment indicated that annual loads are dominated by CSO for substances with removal in WWTP above ~95%. Moreover, it showed that substances with high removal in WWTP can lead to concentration peaks in the river during CSO events. The calculated results could be verified based on eight years of monitoring data from the River Spree, collected between 2000 and 2007. Substances that are well removed in WWTP such as NTA (nitrilotriacetic acid) were found to occur in significantly increased concentration during CSO, while the concentration of substances that are poorly removable in WWTP such as EDTA (ethylenediaminetetraacetic acid) decreased in CSO-influenced samples due to dilution effects. The overall results indicate the potential importance of the CSO pathway of well-removable sewage-based trace contaminants to rivers. In particular, high concentrations during CSO events may be relevant for aquatic organisms. Given the results, it is suggested to include well-removable, sewage-based trace contaminants, a substance group often neglected in the past, in future studies on urban rivers in case of combined sewer systems. The presented methodology is suggested for a first assessment, since it is based solely on urban drainage data, which is available in most cities.

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#### 1. Introduction

Urban streams are affected by trace contaminants from wastewater treatment plants (WWTP), stormwater effluents and — in the case of combined sewer system — combined sewer overflows (CSO). In particular, pollutants that persist in WWTP are an ongoing issue. For instance persistent

carbamazepine, sulfamethoxazole and diclofenac have been identified recently as top priority pharmaceutical residues by the Global Water Research Coalition (2008). A second group of substances that receive increasing attention are both persistent and well-removable contaminants in stormwater runoff from impervious urban surfaces, which can have a significant impact on receiving waters (Grapentine et al., 2008; Hwang

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and Foster, 2008). Especially polycyclic aromatic hydrocarbons (PAH), polychlorinated biphenyls (PCB) and trace metals were identified as priority pollutants from stormwater effluents (Eriksson et al., 2007).

In comparison, well-removable substances, which are predominantly found in municipal wastewater (in the following referred to as sewage-based substances), have received little attention, since they are retained in WWTP and - if measured - only rarely detected at high concentrations in river monitoring programs. However, they can be contained in CSO, which bypass WWTP, in concentrations exceeding those in treated wastewater (Buerge et al., 2006; Phillips and Chalmers, 2009). As a result, an important CSO contribution of such non-persistent substances would be expected in surface waters after rain storms. The general effect was confirmed by Benotti and Brownawell (2007), who found that concentrations in a bay influenced by WWTP and CSO remained high for well-removable paracetamol (CAS 103-90-2) and caffeine (CAS 58-08-2) after a major storm event, while concentrations decreased clearly for all other sewage-based trace organics. Similarly, Buerge et al. (2006) showed that wellremovable caffeine can be used as a tracer for untreated wastewater in streams. Their results imply that the impact of rain events on the CSO share of total annual loads of such well-removable substances may be significant. Apart from the annual contaminant influx, peak concentrations in water bodies, such as rivers or reservoirs, might be caused by CSO. Potential peak concentrations during CSO could be an important issue, since non-persistent substances typically show high biological activity. For instance the synthetic hormone ethinylestradiol (CAS 57-63-6), which is removed in WWTP by ~90% (Joss et al., 2004), can have an endocrinal effect on aquatic organisms already at very low concentrations and short exposures (Caldwell et al., 2008).

The impact of trace substances from CSO in receiving rivers is difficult to assess, since quasi-continuous measurements would be required during storm events. However, even occasional measurements of trace substances during CSO events in receiving rivers or CSO require a high sampling and analytical effort and are, therefore, rarely performed (Welker, 2007).

This article presents two simple methods, which allow (a) the estimation of CSO contribution to annual mass balances of sewage-based substances of varying WWTP removal in receiving rivers and (b) the estimation of a critical WWTP removal fraction, above which peak concentrations are expected during CSO in the receiving river. Both methods do not require actual measurements of concentrations but are based on volumetric flows via WWTP and CSO, data which are typically available today for most larger cities.

In a second step, methods (a) and (b) are exemplified for the Berlin urban drainage system. The results are compared with a unique dataset of trace contaminant concentrations in the receiving River Spree during CSO. A particular focus lies on sewage-based trace contaminants EDTA and NTA, but a selected number of other substances of varying origin (stormwater versus sewage) as well as hygienic parameters are included. Finally local and general implications of sewage-based trace contaminants from CSO are discussed.

Abbreviations and mathematical variables are explained in Table 1.

#### 2. Materials and methods

#### 2.1. Study site

The combined sewer system in the centre of Berlin, Germany, drains an area of ~100 km<sup>2</sup> with 1.4 million inhabitants (Table 2). This corresponds to about 20% of the total drained area of Berlin, but almost 40% of the city's population. The remainder of the area is drained by a separated sewer system (Fig. 1). In the combined sewer area, wastewater is collected together with stormwater and pumped to the WWTP, which are mostly located at the edge or beyond the city limits. If the storage volume of the combined sewer system is exceeded during storm events, combined sewage overflows to the River Spree and its side channels via 179 CSO discharge points over a stretch of  $\sim$  18 km (Table 2). CSO were found to occur typically for rain events (separated by at least 6 h without rain) exceeding a total amount of 4.7 mm for the most sensitive areas (Riechel, 2009). On average this comparably low margin was exceeded 37 times per year between 2000 and 2007, with large interannual variability (Fig. 2). For instance, 48 rain events with a total height > 4.7 mm occurred in exceptionally wet year 2007 compared to 30 events in dry year 2003. In terms of volume, currently about seven million cubic meters combined sewage enter the River Spree each year, with an average sewage to stormwater ratio of 1:11 (unpublished data, Berliner Wasserbetriebe). In order to meet legislative requirements a long term pollution control plan has been set up, which involves different measures to upgrade the combined sewer system. Until 2020 these measures will be realized including, e.g., full utilization of the static in-pipe storage capacities by heightening of CSO crests (under consideration of flood prevention), implementation of actuators like weirs, sluices and throttles for real-time control and construction of additional stormwater tanks.

Whereas effluents from WWTP are measured, total CSO loads and contained raw sewage were available from simulations with the urban drainage network model KOSMO, which requires input data on precipitation, canalization network, hydrological data, inhabitant density and sewage loads (Schmitt, 1993). For the Berlin case study, hydrology and pollutant loads were simulated based on a verified simplified canalization network for the years 1961–1981 (unpublished data, Berliner Wasserbetriebe, 2008). Simulation results were available for two scenarios, (i) before realization of rehabilitation measures and (ii) after complete realization of rehabilitation measures in 2020. Table 3 contains estimated raw sewage effluents from CSO based on simulations for the time period 2000–2007 and for 2020.

The receiving River Spree flows through the combined sewer area before it joins the River Havel, which leaves Berlin in the South West (Fig. 1). Average monthly flow rates before the confluence (at the monitoring station in Fig. 1) vary between 12 m³ s⁻¹ in summer and 42 m³ s⁻¹ in spring (long-time averages published by the Senate of Berlin). Apart from CSO the River Spree receives stormwater effluents upstream of the city centre and the treated wastewater of WWTP 1 (Fig. 1). WWTP 2 was also discharging into the River Spree, before it was decommissioned in March 2003 after having gradually decreased its operation since November 2002. As

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