Contribution of combined sewer overflows to micropollutant loads discharged into urban receiving water

Contribution des rejets de déversoirs d'orage aux flux de micropolluants émis dans un milieu récepteur urbain

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RÉSUMÉ

Très peu d'études ont été réalisées afin de déterminer les flux de micropolluants émis par les rejets de déversoirs d'orage (DO) et leur relative contribution aux flux émis par les systèmes d'assainissement. Afin de combler cette lacune scientifique, plusieurs campagnes de mesure ont été effectuées au niveau d'un rejet de déversoir d'orage principal et au sein d'une station de traitement des eaux usées (STEU) de 160 000 EH située dans le sud-ouest de l'Allemagne. Cette étude a porté sur l'analyse de 69 micropolluants organiques, représentant plusieurs sources d'émission et ayant différentes caractéristiques physico-chimiques : 17 résidus médicamenteux et de soins corporels, 5 produits de contraste rayons X, 18 produits chimiques industriels, 7 pesticides urbains, 6 PCB et 16 hydrocarbures aromatiques polycycliques. Bien que les rejets de DO à l'échelle du bassin versant représentent seulement 18 % du volume total annuel déversé (somme des rejets de DO et STEU), ils contribuent à hauteur de 30 % à 95 % aux flux annuels de 26 micropolluants (parmi eux, 16 composés ont des normes de qualité environnementale fixées par la DCE). Parallèlement, pour 16 autres substances, la contribution des rejets de DO aux flux annuels reste inférieure à 10 %. Afin de réduire efficacement les émissions de micropolluants vers le milieu récepteur, il est nécessaire de combiner plusieurs mesures concernant la station d'épuration et le réseau d'assainissement.

ABSTRACT

Only few studies deal with micropollutant loads discharged by combined sewer overflows (CSO) and their relative contribution to emissions from urban wastewater systems. To overcome this research gap, samples were collected over one year at a CSO outfall and at a wastewater treatment plant (WWTP) in Southwest Germany (with a capacity of 160,000 population equivalents). 69 organic micropollutants were considered, which are representative for various sources and pathways, as well as having different chemical and physical properties: 17 pharmaceutical and personal care products, 5 X-ray contrast media, 18 industrial chemicals, 7 urban pesticides, 6 PCBs and 16 EPA PAHs. Although CSO discharges represent 18 % of the total annual water discharge (CSO plus WWTP effluent discharges), CSO discharges contribute 30–95 % of the annual load for 26 pollutants (among them 16 compounds with EU environmental quality standards). In contrast, for 26 other pollutants less than 10 % of the annual load was contributed by CSO discharges. In order to reduce the discharged micropollutant loads into the receiving water body effectively, it is necessary to combine different measures regarding both CSO and WWTP management strategies.

KEYWORDS

Annual pollutant loads, combined sewer overflows, micropollutants, urban catchment

1 INTRODUCTION

The occurrence of organic micropollutants in the aquatic environment is an important contemporary issue. Principal sources of micropollutants in urban surface waters are wastewater treatment plant (WWTP) discharges, stormwater runoff and combined sewer overflows (CSO) (Phillips et al., 2012). Many recent studies showed that inputs from CSO can represent an important source of organic micropollutants which can affect the surface water quality (Buerge et al, 2006; Phillips and Chalmers, 2009; Musolff et al., 2010; Gasperi et al., 2012, Launay et al., 2013 and Becouze-Lareure et al., 2015). However, only few studies deal with pollutant loads discharged by CSO and their relative contribution to annual emitted loads (Cladière et al., 2011; Phillips et al., 2012). It represents a key issue when dealing with management decisions in order to reduce micropollutant emissions into surface waters. To improve the understanding of impacts of CSO on water quality, the present study focused on micropollutant loads discharged by CSO events into urban receiving water. Sample and data collection for a wide range of organic pollutants over one year allowed estimating the relative contribution of CSO and WWTP to annual emitted loads.

2 MATERIAL AND METHODS

2.1 Study site and sampling strategy

This study was conducted in an urban catchment (area 35 km²) in the south-west of the city of Stuttgart, Germany. The urbanised areas are mainly drained by a combined sewer system and the coefficient of imperviousness of the catchment is 50 %. Upstream of the WWTP discharge, the river Körsch receives discharges from 37 CSO structures. Among them, 16 are combined with storage tanks. Based on rainfall-runoff simulations, the volumes of discharge from each CSO during storm events were calculated. The volume discharged from the last CSO outfall upstream of the WWTP represented 34 % of the total overflow discharge and therefore was selected for monitoring campaigns. Flow-weighted composite samples were collected over 12 months during both dry and wet weather conditions at the WWTP influent (n = 143), final effluent (n = 41) and grab samples were collected in the receiving water (n = 55). From July to October 2014, 6 events were collected at the CSO outfall upstream of the WWTP. Online flow measurements in the CSO discharge pipe allowed to collect flow-weight samples by an automatic sampler (n = 25). Main characteristics of CSO events sampled are given in table 1.

Table 1. Characteristics of CSO events sampled

Dates	28/07	02/08	26/08	12/09	21/09	17/10
Rainfall [mm]	23	14	46	30,7	19.9	21.7
Volume [m³]	76,190	4,258	40,412	32,801	12,554	35,256
Duration [h]	14	2.66	11.5	17.75	8.5	8

69 organic micropollutants were chosen for chemical analysis, which are representative for various sources and pathways (municipal and industrial wastewater, urban runoff), as well as having different chemical and physical properties: 17 pharmaceutical and personal care products, 5 X-ray contrast media, 18 industrial chemicals, 7 urban pesticides, 6 PCBs and 16 EPA PAHs.

2.2 Analysis

Analyses were carried out by the laboratory of Stuttgart's urban drainage service. Each sample was analysed for pH, electrical conductivity, total suspended solids (TSS), chemical oxygen demand (COD), NH_4^+-N, NO_3^--N, NO_2^--N, total phosphorous and phosphate (PO_4^{3-}), in accordance with standard European methods. Regarding micropollutant analysis, after adding the internal standards, the samples (non-filtered and filtered samples using cellulose nitrate membranes 0.45 µm) were extracted with dichloromethane. Before analysis, extracts were spiked with internal standards for quantification. The analysis was performed by gas chromatography coupled to mass spectrometry (GC-MS) for PAHs, PCBs, flame retardants, synthetic musks and phenolic xenoestrogens. For pharmaceuticals, urban pesticides, X-ray contrast media, artificial sweeteners and corrosion inhibitors, analysis was performed by liquid chromatography coupled to a tandem mass spectrometry (LC-MS/MS). All results shown in this paper are based on original non-filtered samples.

3 RESULTS AND DISCUSSION

3.1 Micropollutant loads discharged by six CSO events sampled

The micropollutant loads discharged during six CSO events are given in Table 2. The total volume discharged at the CSO outfall sampled as well as event duration ranged from 4,258 m³ to 76,190 m³ and from 2.66 h to 17.75 h respectively, which explains the high variability of micropollutant loads discharged by CSO. The highest discharged loads were found for caffeine (695 g), artificial sweeteners acesulfame and sucralose (174 g and 115 g respectively) and plasticiser DEHP (113 g). For 38 organic micropollutants (55 % of substances analysed), loads discharged by CSO events were greater than WWTP effluent daily loads. These results were found for 16 EPA-PAHs, urban pesticides, synthetic musks, organophosphorus flame retardants, caffeine and ibuprofen. For these compounds, the ratios of CSO loads to WWTP loads ranged from 1.1 to 420 (for caffeine).

Table 2. Loads (g/event) of selected organic micropollutants for the six CSO events sampled and WWTP effluent daily loads (g/d)

Substance	28/07	02/08	26/08	12/09	21/09	17/10	Median	WWTP effluent (Mean – n=9)
Caffeine	340	61	410	695	171	215	278	1.7
Carbamazepine	2.3	0.3	1.8	5.2	1.9	1	1.9	8.4
Diclofenac	4.7	0.7	3.7	8.8	3	2.5	3.4	26.1
Ibuprofen	44	5.8	46	74	18	23	34	0.74
Mecoprop	17.8	0.8	1.7	4.8	1.7	3.5	2.6	0.52
Diuron	28.6	0.5	4.3	8.5	1.1	5.5	4.9	0.68
Isoproturon	8.2	0.2	2.0	5.9	0.4	3.2	2.6	0.42
Acesulfam	110	15	110	174	45	28	78	225
DEHP	113	17	48	101	54.5	63	58.7	4.7
Fluoranthene	7.8	0.58	3.9	9.5	3.5	1.7	3.7	0.05
4-Nonylphenol	6.9	2.2	5.3	20.7	4.7	15.8	6.1	3.3

3.2 Annual emitted loads for the year 2014

Annual loads discharged into receiving water by all CSO outfalls from the entire urban catchment were assessed, assuming that stormwater is completely mixed with raw sewage in the sewer system. Annual loads discharged by the WWTP were also calculated based on sampling campaigns performed during dry and wet weather conditions on 2014. As figure 1 illustrates, although CSO discharges represented 18 % of the total annual water discharge (CSO plus WWTP effluent discharges), CSO discharges contributed 30–95 % of the annual load for 26 micropollutants (among them 16 compounds * with EU environmental quality standards). In contrast, for 26 other pollutants less than 10 % of the annual load was contributed by CSO discharges.

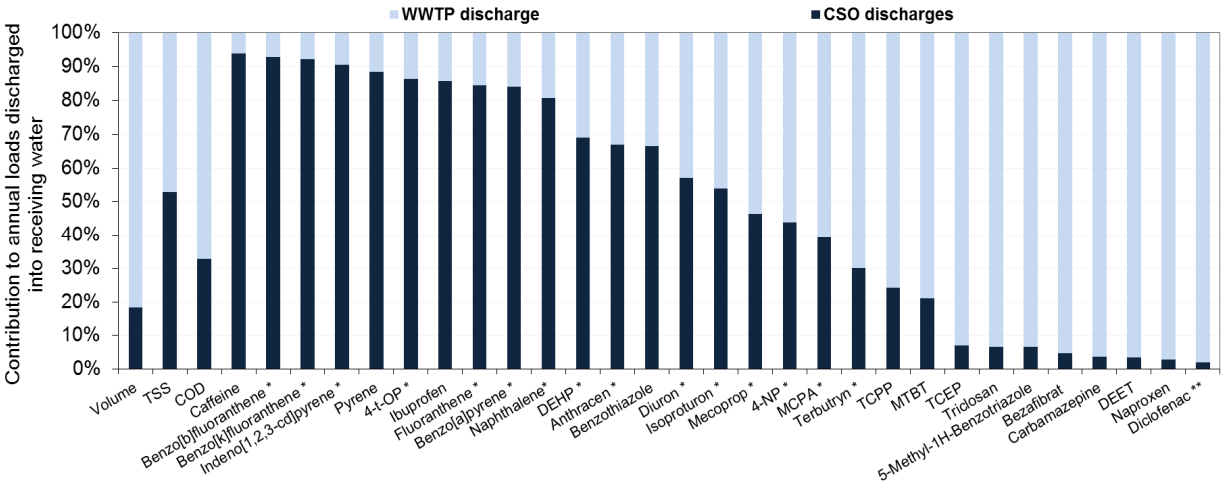

Figure 1. Contribution of CSO events and WWTP to annual micropollutant loads discharged into receiving water

As shown in figure 2, CSO events represented the main source of micropollutants to receiving water for substances with very high WWTP removal efficiency (> 85 %), like caffeine, ibuprofen, and PAHs. Our findings are similar with results from two other previous studies (Weyrauch et al., 2010; Phillips et al., 2012).

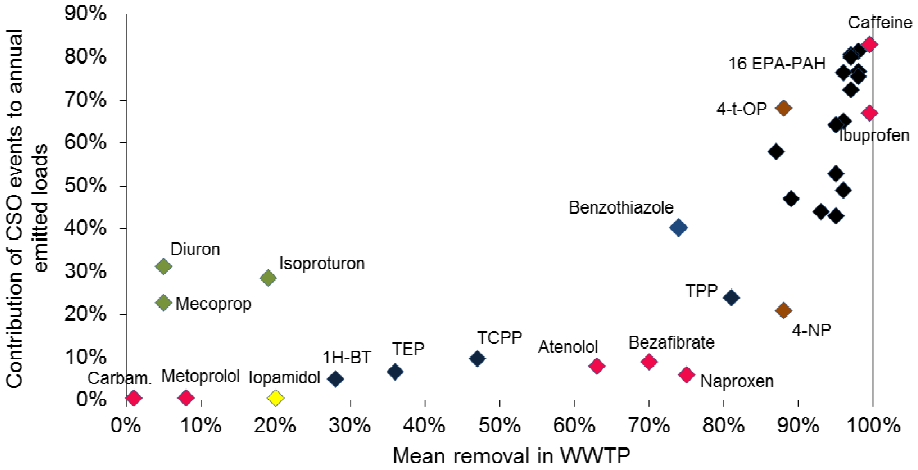

Figure 2. Mean removal in WWTP and relative contribution of CSO events to annual loads

Furthermore, although removal efficiency of urban pesticides was low (from 5 % to 20 %), the relative contribution of CSO events to emitted annual loads was quite important (from 20 % to 32 %). This can be explained by the fact that these compounds are transported mainly during storm events into sewer system due to catchment surface runoff (Becouze-Lareure et al., 2015). Consequently, loads discharged from WWTP during dry weather are lower than for other substances which occurrence in sewers is not weather-dependent and don't play a major role for the relative contribution of WWTP to annual loads.

4 CONCLUSIONS

The relative contribution of CSO to annual loads of micropollutants discharged into urban receiving water showed a very high variability depending on the substance considered. Although CSO discharges represent 18 % of the total annual water discharge (CSO plus WWTP effluent discharges), CSO discharges contributed 30–95 % of the annual load for 26 pollutants (among them 16 compounds with EU environmental quality standards). CSO represented a relevant source of discharged loads for caffeine, ibuprofen, 16 EPA-PAH, phenolic xenoestrogens and urban pesticides. In contrast, WWTP was the main source of pharmaceuticals like carbamazepine, diclofenac, bezafibrate, naproxen and X-ray contrast media in surface water. In order to reduce the discharged micropollutant loads in the receiving water body effectively, it is necessary to combine different measures regarding both CSO and WWTP management strategies. Several scenarios will be developed and the efficiency of combined measures will be evaluated with the help of integrated modelling.

ACKNOWLEDGEMENT

This work was supported by the Ministry of the Environment, Climate Protection and the Energy Sector Baden-Württemberg, Germany. The authors acknowledge the technical staff of the WWTP for the cooperation during the study. The authors would also like to thank the technical and laboratory staff of the Stuttgart Municipality for their valuable contributions to this work.

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