Assessment of organic pollutants with respect to occurrence and fate in combined sewer systems and possible impacts on receiving waters

Évaluation des polluants organiques : présence et évolution dans les réseaux unitaires et impacts possibles sur les milieux récepteurs

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RESUME

Des polluants organiques sélectionnés sont classés sur la base de recherches bibliographiques intensives. Deux paramètres d'eaux usées (DCO et ammonium) et six polluants organiques sélectionnés (HAP, DEHP, E2, EE2, ATED, ANT) sont spécifiés. En conséquence, cet article présente des concentrations représentatives de flux par temps sec, de ruissellements de surface et d'effluents d'usines de traitement des eaux usées dans les systèmes d'assainissement combinés.

La seconde partie de l'article présente une première estimation des rejets d'un système combiné et des impacts éventuels sur les eaux réceptrices en termes de (1) charges de rejets annuels calculés par le biais de simulations de charges de pollution dans un bassin versant théorique et (2) de concentrations calculées dans les rejets de surverses de systèmes combinés et les concentrations dans les eaux de réception.

ABSTRACT

Selected organic pollutants are classified based on an intensive literature survey. Two wastewater parameters (COD and ammonium) and six selected organic pollutants (polycyclic aromatic hydrocarbons (PAH), diethylhexylphthalate (DEHP), estradiol (E2), ethinylestradiol (EE2), ethylenediamine tetraacetic acid (EDTA) and nitrilo triaceticacid (NTA)) are specified. As a result, for the first time representative concentrations in dry weather flow, surface runoff and effluent of wastewater treatment plants (WWTPs) in combined sewer systems (CSS) are stated.

The second part of the paper presents a first estimation of main emission out of a combined sewer system and possible receiving water impacts in terms of (1) annual discharged loads calculated by pollution load simulations in a hypothetical catchment and (2) concentrations calculated in combined sewer overflows (CSO) discharges and resulting receiving water concentrations.

KEYWORDS

Assessment, CSO, impacts on receiving waters, organic pollutants, pollution load simulation.

1 INTRODUCTION

The recently released European Community Water Framework Directive (WFD) considers specific organic pollutants (e.g. pesticides, PAHs, DEHP) as priority substances, for classifying the chemical status of receiving waters (EC, 2000). In Germany emissions out of the urban drainage system have already been identified to be a significant source of receiving water pollution for some of these organic pollutants (e.g. PAHs) (Irmer, 2006). As a consequence, it has become necessary to monitor a wide range of organic pollutants in addition to conventional water quality parameters, such as chemical oxygen demand (COD).

An issue having received very limited attention are emissions from combined sewer systems that combine the dry weather flow with surface runoff and discharge the combined sewer overflows (CSOs) nearly untreated into the receiving waters.

Moreover, the occurrence and fate of organic pollutants in the combined sewer system and possible impacts on receiving waters are poorly understood, particularly because there is a deficit of analytical data. A number of studies have reported data on organic pollutant concentrations in WWTP effluents. In contrast inflows of WWTPs and CSOs have not been investigated so far. Therefore main objectives of this study are to give a first assessment of organic pollutants in the combined sewer system by

- summarizing reported concentrations of selected pollutants in dry weather flow, surface runoff and effluent of WWTPs,
- classifying pollutants with respect to occurrence and fate in combined sewer systems and
- identify important emission points and possible receiving water impacts in terms of annual discharged loads and receiving water concentrations as well.

2 METHODS

2.1 Literature survey

In a first step reports evaluating occurrence of selected pollutants in the combined sewer system were analyzed. Concentrations of selected water quality parameters and specific organic pollutants were compiled from over 300 reported studies (for details see Welker, 2004, 2005). The selection of the described substances was based on toxicity, regulatory significance and data availability.

Summarizing reported field data provides a preliminary estimate of representative concentrations in dry weather flow, surface runoff and effluent WWTP (Table 1). These values were used as input for the simulation studies described below.

value	COD [mg/l]	NH4-N [mg/l]	PAH [µg/l]	DEHP [µg/l]	EE2 [ng/l]	E2 [ng/l]	EDTA [µg/l]	NTA [µg/l]
dry weather flow	441	22	0.7	23	6	15	59	150
surface runoff	107	1.0	2.3	7.3	<0.02*	<0.02*	<0.05*	<0.05*
WWTP (dw)	66	2.0	0.3	4.6	3	0.75	59	7.5
WWTP (ww) *2	31	0.7	0.8	3.1	0.9	0.21	17	2.1

dw: dry weather; ww: wet weather; COD: chemical oxygen demand; NH_4-N: ammonium; PAH: polycyclic aromatic hydrocarbons; DEHP: diethylhexylphthalate; EE2: ethinylestradiol; E2: estradiol; EDTA: ethy-lenediamine tetraacetic acid; NTA: nitrilo triaceticacid; * calculation with 50 % of the lowest published detection limit; *2: calculated by a balance

Table 1. pollutant concentrations in important flow types of the combined sewer system

2.2 Pollution load simulations

A hypothetical catchment (187 ha) with 9,900 inhabitants, which was previously used as a representative catchment for various theoretical studies in Germany (Leinweber, 2002) was chosen for this study. The catchment has a total paved area of 98 ha and is drained by a combined sewer system including two CSOs and two storm water tanks. The overflow structures and the tanks are designed according to the German guideline A 128 (ATV, 1992).

For pollution simulation the software KOSMO (Kontinuierliche Schmutzfrachtmodellierung) has been used (Schmitt, 1993). KOSMO includes models for all relevant processes e.g. surface pollution is calculated with an exponential accumulation equation. Surface runoff and corresponding sewer flow is computed with a hydrological method (for details see Welker, 2006).

2.3 Calculation of receiving water concentrations

The pollution load simulation provides information about flow characteristics of every CSO structure and the corresponding substance concentrations, including average concentrations, maximum and minimum values. Average CSO concentrations out of the simulation were used in order to calculate receiving water levels. The next step is to assume representative flow ratios. Investigations at existing sites show, that the ratio of discharged CSO flow to receiving water flow ranges widely from < 1 to 30 (Wolf and Borchardt, 1990). In this study CSO flow (313 l/s) is discharged into two hypothetical receiving waters with flows of 50 l/s and 500 l/s which results in realistic flow ratios between 6.3 and 0.6.

Assuming concentrations in the receiving water form literature data, the mixed concentration after discharge of CSO can be calculated by simple mixing equations. These theoretical concentrations are then compared either with receiving water monitoring data or with regulation demands and various toxicity data. Using this procedure, preliminary predictions of organic pollutant concentrations in receiving waters after CSO discharge are possible.

3 RESULTS AND DISCUSSION

3.1 Classification of selected organic pollutants in the combined sewer system

An important result of the literature survey considering nearly 100 substances was the selection of representative compounds. These substances can be seen as example compounds, which have typical substance properties and show a representative behaviour in the combined sewer system. Other not mentioned pollutants (e.g. pharmaceuticals) are either described elsewhere (Welker, 2006) or can be arranged in a specified substance group with comparably properties. Table 2 gives information about the main describing variables of selected pollutants.

	PAH*	DEHP	EE2	E2	EDTA	NTA
general remarks						
characterization	group of hydrocarbons	phthalate	synthetic hormone	natural hormone	complexing agent	complexing agent
citation in water reg.	yes	yes	no	no	yes	yes

citation in WFD	priority sustances	priority substance	no	no	no	no
remarks	moderate data base; high toxic potential	ubiqui-tous; inconsis-tent toxic data	high endocrine potential	high endocrine potential	unclear ecotoxic evaluation	unclear ecotoxic evalua-tion
substance properties						
transport rel. to particles	high	high	moderate	low	low	low
degradation potential	low	moderate	moderate	high	low	high
elimination WWTP	high (s)	high (d,s)	high (d)	high (d)	low (-)	high (d)
occurrence in css						
origin	runoff/ dwf	runoff/ dwf	dwf	dwf	dwf	dwf
literature	1-3	4-6	7-9	7-9	10-12	10-12

* group of substances: variation between single compounds; reg.: regulations; rel.: related; WFD: European Water Framework Directive; WWTP: waste water treatment plant; dwf: dry weather flow; d.: degradation; s.: sorption; css: combined sewer system

literature: 1: Makepeace et al. 1995; 2: Marsalak and Schröter, 1988; 3: Haritopoulou, 1996; 4: Helmreich, 2001; 5: Litz, 1998; 6: Kollotzek et al., 1998; 7: Birkett and Lester, 2003; 8: Spengler et al., 2001; 9: ATV-DVWK IG-5.4, 2002; 10: LUA NRW, 2003; 11: CSTEE, 2003; 12: Alder et al., 1997

Table 2. Classification of selected organic pollutants with respects to general remarks, citation in water regulations, important substance properties and occurrence in the combined sewer system

3.2 Pollution load simulations for a combined sewer system

Annual discharge loads of both main emission matrices of the hypothetical catchment (WWTP effluent and CSO) are calculated in order to point out important emission points (Fig.1).

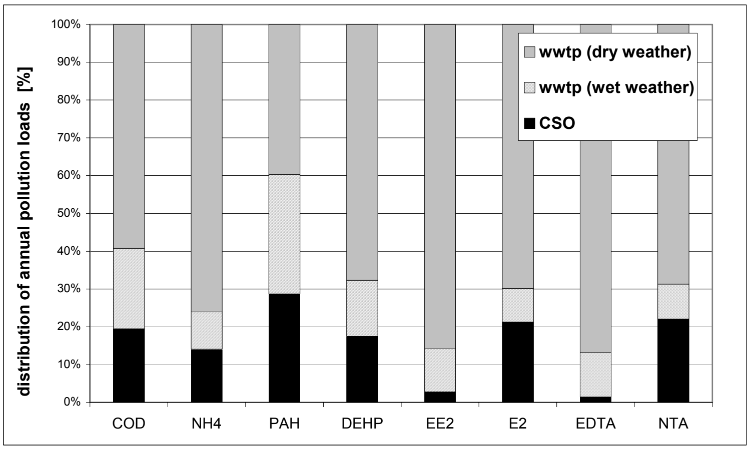


Figure 1: Percentage of annual pollution loads of COD, NH4, PAH, DEHP, EE2, E2, EDTA, and NTA calculated by pollution load simulation

The results show that discharges from the WWTP are generally significant in combined sewer systems. The importance of emissions from CSOs differs significantly from substance to substance.

Substances that predominately originate from surface runoffs also occur in high quantities in CSO emissions. This was quantified by the example of PAH (ca. 30% of the pollution loads result from CSO). Furthermore, the relevance of the effluent of WWTPs during wet weather is demonstrated by the example of primarily surface runoff derived substances. This effect was already confirmed by experimental data (Schäfer, 1999).

A completely different result is obtained for dry weather flow derived substances, such as EE2. In contrast to PAH the percentage of CSO loads is negligible (ca. 3%). Main emissions come from the WWTP effluent, predominately during dry weather due to the relatively low elimination rate in the WWTP (ca. 50%). Therefore the origin of a substance (surface runoff or dry weather flow) is an important factor considering quantities and distribution of pollution loads in a combined sewer system. However, the example of E2 shows that the origin is not the only factor. Similar to EE2, it predominately originates from dry weather flow. In contrast to EE2, annual loads from CSOs are not negligible (over 20%). This effect is caused by the very high elimination rate (up to 99%) in the WWTP.

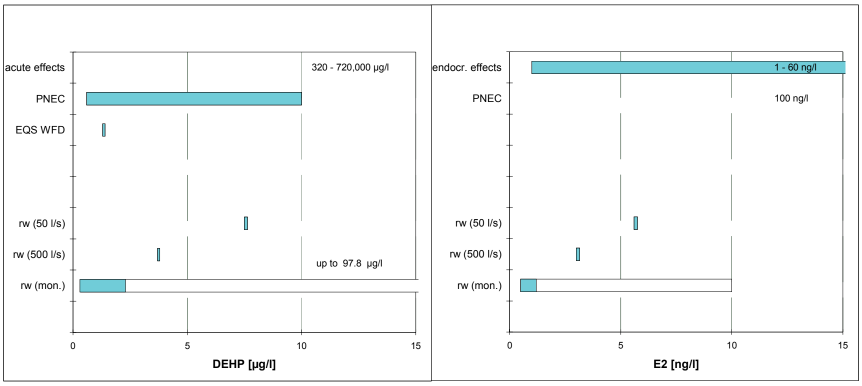
The estimation of the distribution of the emissions from a combined sewer system therefore requires information about the predominant origin (surface runoff, dry weather flow) and the elimination rate in the WWTP.

3.3 Concentration based estimation of possible impacts of CSOs on receiving waters

In addition to annual loads, being most relevant for substances with long-term effects, for short-term effects the pollutant concentrations are more important.

Results of mixing calculations of two organic compounds (DEHP and E2) in receiving waters are presented below and compared with different regulatory and toxicity values. It was assumed that effluents mix completely and rapidly and that concentrations remain relatively constant, which means, that there is an absence of removal by sorption, biodegradation, or photolysis in the receiving water body.

Generally, toxicity values are expressed as acute effects (e.g. LC_{50}: Lethal Concentration) or chronic effects (e.g. NOEC: No Effect Environmental Concentration) and vary widely for a given substance for different target organisms. In order to obtain one toxicity value often the lowest effect concentration is used. After dividing it with a safety factor (often 1,000) a so called PNEC: Predicted No Effect Concentration is obtained, which was already defined for some of the investigated organic pollutants (see Figure 2).



rw (mon.): measured receiving water data; PNEC: Predicted No Effect Concentration; EQS WFD: Environment Quality Standards Water Framework Directive (EC, 2006)

Figure 2. Comparison of receiving water concentrations (monitored and calculated after CSO discharge in receiving waters with flows of 50 l/s and 500 l/s) with toxicity data and different regulatory limits for DEHP and estradiol (E2)

DEHP-concentrations, which were measured in receiving waters (rw mon.) are frequently in the same range as the limits of EQS-levels recently proposed in the EC-Water Framework Directive (EC, 2006). Moreover, they are clearly below DEHP levels that cause acute toxic effects (320 - 720,000 µg/l) (Kollotzek et al., 1998).

CSO discharges can significantly increase receiving water concentrations. The calculated concentrations (rw (50 l/s); rw (500 l/s)) are as high as or exceed toxicity values (PNEC, EQS WFD). Data on acute effect concentrations are in every case higher than the calculated receiving water concentrations.

Generally DEHP is difficult to evaluate since data for receiving waters are limited and extremely heterogeneous. Some authors attribute the occurrence of DEHP in receiving waters exclusively to atmospheric deposition (LUA NRW, 2001) although our calculations demonstrate that CSO is potentially a significant source for DEHP in surface waters. More data is needed to quantitatively assess the impact of the different sources.

The calculated concentrations in receiving water for estradiol (E2) increase significantly after CSO discharge under different river flow conditions. Anyhow, these concentrations are far beyond PNEC-levels. In contrast, they reach levels that may elicit endocrine effects.

In summary, this analysis indicates CSO discharges as significantly influencing the quality of receiving waters with respect to specific organic pollutants. For some pollutants the estimated concentrations exceed toxicity or regulatory thresholds. Applying these conclusions to specific cases, requires verification of the underlying assumptions of this study (complete mixing and no degradation). Nevertheless the presented method is a useful “screening-tool” for estimating possible impacts of CSOs on receiving waters.

4 CONCLUSIONS

Results of the pollution load simulation show, that emissions from CSOs are significant for both pollutants from surface runoff and dry weather flow. This finding applies also to dry weather flow derived pollutants that are removed efficiently in WWTPs, such as estradiol. This conclusion is confirmed by results from the receiving water calculations. They indicate that concentrations of some organic compounds increase significantly after CSO discharge even exceeding toxicity and regulatory thresholds in some cases. In summary, an estimation on the distribution of the emissions from a combined sewer system requires information about the predominately origin (surface runoff, dry weather flow) and the elimination rate in the WWTP.

In addition to the presented pollution load simulations, it will be interesting to observe possible changes in the load distribution when conditions are modified in the catchment (Welker, 2005). Further studies on pollution load balances will therefore consider additional organic pollutants with variations in the catchment structure, which nowadays presumably occur. In this field the integration of separate sewer system and the implementation of new treatment facilities (e.g. constructed wetlands) are possible scenarios.

Due to different legal regulations (e.g. Water Framework Directive) more and more organic compounds in receiving waters have to be considered. Emissions out of the combined sewer system have already been identified to be a significant source of receiving water pollution for some organic pollutants in Germany (Irmer, 2006).

Looking at existing measurement programs in combined sewer systems a wide concentration range of constituents and a significant lack of data is obvious. A main conclusion of this is the necessity for a broader date base on the amount of substances, especially organics, in the matrices of the combined sewer system and for the CSOs in particular. Beside of the introduced substances further compounds (e.g. surfactants, pharmaceuticals, herbicides, flame retardants) should be taken into account with respect of occurrence and behaviour in the combined sewer system.

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