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Reduction of micropollutants and bacteria in a constructed wetland for combined sewer overflow treatment after 7 and 10 years of operation

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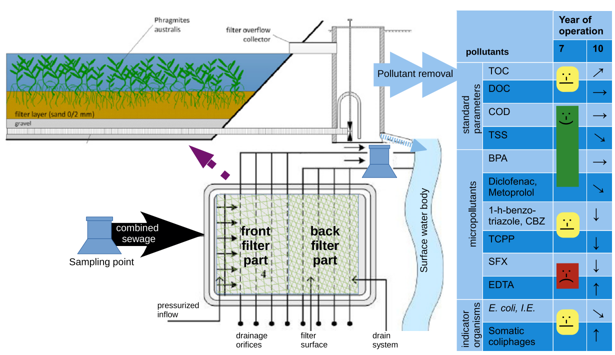
HIGHLIGHTS

- Investigations on CSOs treating wetlands 7 and 10 years after starting operation
- Focus on micropollutants and indicator bacteria
- (bio-)chemical pollutant long-term removal depends on renewable adsorption capacity.
- Ageing effects for non-biodegradable substances removal; seasonal effects for BPA
- Limited process variations for indicator microorganism: stable partial removal

GRAPHICAL ABSTRACT



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ABSTRACT

Repeated investigations on constructed wetlands for the treatment of combined sewer overflows, also named bioretention filters or retention soil filters, are necessary to provide information on their long-term performance. In this study, a sampling campaign was conducted on micropollutants, indicator microorganisms and standard parameters ten years after such filters were in operation and three years after the first investigation; it revealed that the filters lost capacity to remove chemical substances with no or only slow biological degradability. This was the case e.g. for phosphate (decrease from 29 to 11%), diclofenac (67 to 34%) and TCPP (34% to negative reduction). They continued to remove easily degradable parameters such as COD (stable around 75%) stably. The indicator microorganisms *Escherichia coli* (1.1/0.8 $\log_{10}$), intestinal enterococci (1.3/0.8 $\log_{10}$) and somatic coliphages (0.6/1.0 $\log_{10}$) showed comparably low process variations given the difficulties in sampling and analysing microbial parameters representatively as well as given natural variations in microbial behaviour and growth. Additionally, for bisphenol A, we found a temperature-related difference of removal efficiencies: while in the cold months (winter), the removal was only 53% on average, it increased to 90% in the warm months (summer).

As for the long-term prospective of retention soil filters, decision-makers need to identify the most important pollutants in a specific catchment area and adapt the filter design accordingly. If pollutants are targeted that lead to an exhausted filtration capacity, post treatment or the exchange of charged filter material is necessary. However, for easily biologically degradable substances, so far, there is no limit in their use.

Abbreviations: BPA, bisphenol A; CBZ, carbamazepine; CFU, colony forming unit; CSO, combined sewer overflow; EDTA, ethylenediaminetetraacetic acid; LOQ, limit of quantification; MPN, most probable number; RSF, retention soil filter; TCPP, tris (2 chloroisopropyl) phosphate.

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1. Introduction

Pollutants can bypass sewage treatment systems when combined sewer overflows (CSOs) result from heavy rainfall events. As an example, Launay et al. (2016) found 62 of 69 investigated micropollutants in CSOs in one catchment area. In terms of microbial impact, the loads discharged via CSOs into surface waters can be higher than that of continuously discharged effluent from sewage treatment plant with tertiary treatment from the same catchment (Tondera et al., 2016b; Schreiber et al., 2016). Since such pollutant loads created by CSOs are one reason threshold values of the EU Water Framework Directive and the Bathing Water Directive are exceeded, states and municipalities need to make greater efforts to reduce them.

Currently, the only constructions able and available to reduce both particulate as well as dissolved pollutants from unavoidable CSOs on a large scale are vertical flow constructed wetlands for combined sewer overflow treatment. Other options theoretically capable of reducing both micropollutants and microorganisms are ozonation (Tondera et al., 2015), horizontal flow wetlands (Pisoeiro et al., 2016) or ballasted flocculation (Gasperi et al., 2012), which were all tested in pilot scale studies, but have not yet proven to be reliable on a large scale. Additionally, ozonation cannot reduce the particle load and provides a risk of forming toxic degradation products.

Vertical flow constructed wetlands are often referred to as bioretention filters, or also as retention soil filters (RSFs), which is a literal translation from German since soil was used as filter material in the first sites. Nowadays, sand (grain size 0/2 mm) is used as filter material, with up to 20% of broken limestone to prevent acidification caused by microbial processes such as nitrification.

This wetland system works as follows: contaminated water released from the overflow of CSO storage tanks is filtered through a cropped sand layer, drained and discharged throttled into a surface water body. In between precipitation events the filter dries out and further aerobic degradation of the retained pollutants occurs.

In Germany, RSFs have been used for more than two decades. On a national scale, there are no data available about how many RSFs exist, but in the State of North Rhine-Westphalia, there are at least 137 sites in operation in combined sewer system and 42 in separate sewer system.[1] The construction has been adapted by researchers in France and Italy (Meyer et al., 2013; Masi et al., 2016; Pálfy et al., 2017). Evaluating their performance over time can provide important information for improving their design and overall value to the receiving waterbodies.

To date, only few investigations exist on the reduction of bacteria and bacteriophages in large scale applications. Waldhoff (2008) conducted a comprehensive investigation into the reduction rates of *Escherichia coli* (*E. coli*; median reduction of 1.2 $\log_{10}$) and intestinal enterococci (I.E., median reduction of 0.9 $\log_{10}$) in pilot and large scale systems. Ruppelt et al. (2018a), finding similar reductions of the indicator bacteria in pilot scale systems, also investigated those of somatic coliphages (1.0 to 1.2 $\log_{10}$).

Reduction of faecal indicator bacteria ranges between 1 and 2 $\log_{10}$ with filtration, straining processes and adsorption serving as the main removal mechanisms (Ruppelt et al., 2018a). Merkel and Schaule (2010) investigated the performance of four established but sparsely-loaded RSFs in North Rhine-Westphalia, which had been in operation for 3.5 to 4 years. The reduction of *E. coli* varied between a median of 1.4 $\log_{10}$ to almost 3 $\log_{10}$ for the different facilities. The observed retention time was up to eight days, with reduction performance improving during longer lasting events.

Event-based long-term monitoring of an RSF by Christoffels et al. (2014) over 14 months showed that it had a higher mean reduction capacity than other investigated systems, of at least 3 $\log_{10}$ for faecal indicator bacteria, faecal pathogenic bacteria and somatic coliphages in CSOs. Additionally, higher maximum reductions were observed of 4.7 $\log_{10}$ for *E. coli* and of 4.9 $\log_{10}$ for enterococci for the same plant. During the monitoring period, the first flush (mixed sample out of up to 300 L with time-delayed effluent sampling start) from 25 of 33 events could be analysed for several bacteria and somatic coliphages with different dry phases between the events depending on the local weather conditions in the course of the year (Mertens et al., 2013). Filter material taken from the large scale filter investigated in this study was also used for a pilot scale study testing RSFs for the post-treatment of a sewage treatment plant (Brunsch et al., 2018). However, the continuous intermittent charge of the filter creates conditions of a post-treatment step which is different to a stochastically loaded RSF in combined sewer systems.

Most studies are based on relatively short sampling periods of one year and the samples were collected under difficult environmental conditions, which is also true for the even fewer investigations on micropollutants. Scheurer et al. (2015) reported a reduction for diclofenac of 81 ± 21% based on sampling inflow and outflow during five events. Christoffels et al. (2014) chose a different way of evaluating the events since diclofenac could only be quantified in 68% of 343 single samples from 33 events over 14 months, but in only 9% of the outflow samples (quantification limit: 100 ng L^{-1}). Hence, the maximum concentration of diclofenac in the outflow was 65% lower than that in the inflow. Up to twelve subsamples (depending on the inflow and outflow duration) taken by an automatic sampler during one event allowed a higher temporal resolution for micropollutants than for microbes. Some investigated pharmaceuticals such as carbamazepine could not be removed sufficiently in the study. Other investigated substances were industrial chemicals, e.g. bisphenol A.

Pálfy et al. (2017) investigated the reduction of PAHs in the first vertical flow constructed wetland for CSO treatment in France, but did not find conclusive results due to the low number of sampled events and the use of new filter material, which might have released pollutants initially attached to it after operation had begun.

The RSF presented in this study was evaluated by Tondera et al. (2013) in a first survey monitoring, where data from a sampling campaign after seven years of operation were presented. Since there are still few repeated long-term observations on the reduction of micropollutants and indicator organisms through RSFs (Mertens et al., 2013, Ruppelt et al., 2018a), the filter was subject to another sampling campaign almost three years after the first one. The results should provide information on how this system performs over a long period of time. Additionally, this paper uses a method for evaluating data which were gathered under highly varying field conditions common in large scale facilities, especially those fed with stormwater driven overflows: the evaluation of the measurement results was extended by a statistical approach with a Monte Carlo simulation used in Tondera et al. (2016b).

The presented subsequent investigation of a RSF seven and ten years after starting operation is to our best knowledge the first of its kind for a large scale site published and gives first indications of how the reduction for micropollutants and microbial indicator parameters develops in sites with ageing filter material.

2. Methods

2.1. Site description and sampling regime

During a first one year sampling phase between July 2011 and June 2012 (Phase I), we took samples on a large scale site at Bergheim (conurbation of Cologne). Since first flushes of the combined sewage exceeded the capacity of the wastewater treatment plant, they were captured in two storage tanks (Fig. 1); thus, the inflow to the filter does not show significant first flush effects (Tondera et al., 2013). Eight events were sampled in Phase I, as presented in Tondera et al.

[1] www.elwasweb.nrw.de (9.05.2018)