Mass balance of micropollutants over the first year of operation in a stormwater biofilter: a field approach integrating water and soil

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Bilan de masse de micropolluants au cours de la première année de fonctionnement dans un ouvrage d'infiltration végétalisé : une approche in situ sur l'eau et le sol

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RÉSUMÉ

Le bilan de masse *in situ* d'une selection de micropolluants, comprenant le Cu, le Zn, le pyrène (Pyr), le phenanthrène (Phen), le bisphénol-A (BPA), l'octylphénol (OP), le nonylphénol (NP) et le bis(2-ethylhéxyl) phtalate (DEHP), a été évalué pour les premiers 13 mois de fonctionnement d'une noue filtrante traitant les eaux de ruissellement de voirie. Le bilan de masse prend en compte les flux polluants entrants et sortants, ainsi que l'évolution de la masse de polluant stockée dans le sol ; il a été évalué par couplage de mesures *in situ* de flux d'eau et d'analyses de micropolluants dans l'eau et dans le sol avec un modèle stochastique. Les incertitudes associées aux mesures expérimentales et à la méthode de calcul ont été évaluées. A partir des masses résultantes, l'abattement de flux annuel a été évalué et la masse cumulée dans le sol comparée à celle interceptée depuis les eaux de ruissellement. L'abattement en masse suit l'ordre Pyr>Phen>Zn>Cu>OP>BPA>NP>DEHP. Il n'a pas été possible de démontrer une dissipation des micropolluants organiques dans l'ouvrage. Pour certains polluants (Pyr, Phen), cela était dû à de grandes incertitudes dans la masse cumulée. Pour d'autres (BPA, OP, NP, DEHP), la masse cumulée dépassait de façon significative celle interceptée depuis les eaux de ruissellement, probablement du fait d'émissions depuis les matériaux de construction.

ABSTRACT

The in situ mass balance was evaluated over the first 13 months of operation of a biofiltration swale for several micropollutants, including Cu, Zn, pyrene (Pyr), phenanthrene (Phen), bisphenol-A (BPA), octylphenol (OP), nonylphenol (NP) and bis(2-ethylhexyl)phthalate (DEHP). The mass balance accounted for inlet and outlet loads and the evolution of pollutant mass in the soil and was established by coupling field measurements of water flows and concentrations of micropollutants in both water and soil with a stochastic model. Uncertainties associated with both experimental measurements and the calculation method were accounted for. Results were used to evaluate annual pollutant load reductions and to compare the accumulation of mass in the soil with that intercepted from stormwater. Annual pollutant load reductions followed the order Pyr>Phen>Zn>Cu>OP>BPA>NP>DEHP. Dissipation could not be demonstrated for any of the organic micropollutants. For some pollutants (Pyr, Phen), this was due to the high uncertainties in accumulated mass. For others (BPA, OP, NP, DEHP), the accumulated mass significantly exceeded that intercepted from stormwater, likely due to emissions from construction materials.

KEYWORDS

Biofiltration, fate mass balance, micropollutants, performance

1 INTRODUCTION

As techniques used for urban stormwater management evolve from approaches based on rapid drainage toward centralized management systems to source-management techniques, the technique of biofiltration, also referred to as bioretention, has gained in popularity, especially in contexts where pollution control is an important objective. A biofiltration device consists of a vegetated depression that receives and stores runoff water, which then filters through a filter media made of engineered soil, before being collected by an underdrain or exfiltrated to the surrounding soil (Davis et al., 2009). In this type of system, pollutants are expected to undergo various processes including sedimentation, physical filtration, sorption, biodegradation, volatilization and plant uptake.

A biofilter's pollution management performance may be considered on two levels: its ability to intercept pollution and, in the case of pollutants subject to biodegradation, volatilization and photodegradation, such as organic micropollutants, its ability to dissipate the intercepted pollution. The first type of performance has been evaluated in a field context for a variety of pollutants, including total suspended solids (TSS), nutrients, trace metals and polycyclic aromatic hydrocarbons (PAH) (Liu et al., 2014), while the potential for dissipation has mainly been studies at the laboratory (LeFevre et al., 2012) or mesocosm scale (Leroy et al., 2015). The two aspects are rarely considered together and few studies have accounted for the performance relative to emerging micropollutants common in the urban context, such as alkylphenols and phthalates.

The establishment of an in situ mass balance for a stormwater biofilter, accounting for inlet and outlet loads, as well as pollutant accumulation in the filter media, would allow for an integrated evaluation of both types of efficiency over a given period. The objective of the present work is to evaluate an integrated mass balance for several micropollutants—Cu, Zn, pyrene (Pyr), phenanthrene (Phen), bisphenol-A (BPA), octylphenol (OP), nonylphenol (NP) and bis(2-ethylhexyl)phthalate (DEHP)—over the first 13 months of operation of a biofiltration swale in order to (i) consider the integrated pollutant load reduction over the period and (ii) compare the intercepted mass with that accumulated in the soil. This is achieved by combining results of a field campaign with a stochastic modeling approach to integrate the data and account for uncertainties.

2 METHODS

2.1 Study site

The study site includes a biofiltration swale (BFS), which drains water from the RD 212, a suburban highway with 11,000 vehicles/day/direction traffic, located in Compans, France in the Paris region. The system is both drained and lined and was constructed in March 2016. Water was collected from a drain beneath the filter media (a sandy loam) at a depth of 50 cm. Inlet quality was evaluated using a reference catchment on the same road and with identical traffic, located 68 m from the BFS catchment (Figure 1). Flow was measured in the BFS drain and RR manhole using tipping bucket flow meters; overflow from the BFS was measured using a V-notch weir. Turbidity was also measured continuously in the RR manhole.

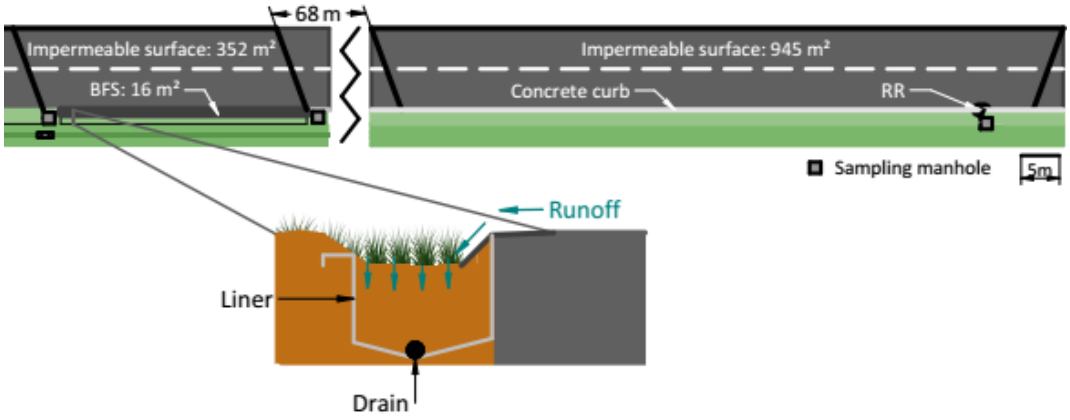

Figure 1: The Compans study site

2.2 Sampling and analysis

Water was collected at each point in proportion to water flow in order to measure the event mean concentration (EMC). 15 rain events were sampled between May 2016 (two months after the

construction of the BFS) and July 2017. While simultaneous sampling of all points was attempted, due to technical difficulties, not all data is paired. Samples were collected within 24 h of each rain event and sent to partner laboratories within 24 h of collection, where they were immediately filtered to distinguish between dissolved and particulate phases for a wide range of micropollutants. Campagnes and water quality results are discussed thoroughly elsewhere (Flanagan et al., 2018); the integrated mass balance was evaluated for a selection of these pollutants (Cu, Zn, Pyr, Phen, BPA, OP, NP and DEHP).

In order to evaluate the mass of pollutants accumulated in the filter media, an initial sample was taken before installation and a sampling campaign was undertaken 13 months after the beginning of operation. In this campaign, seven composite filter media cores were sampled in the BFS from zones with similar Cu, Pb and Zn surface contamination, as determined from a cartography produced by soil contamination at 90 points using X-ray fluorescence spectrometry.

2.3 Mass balance and uncertainty calculations

Five mass balance terms were evaluated using a stochastic approach: the mass entering the biofilter (M_{in}), the mass exiting the biofilter through the drain (M_{drain}), the mass overflowing from the biofilter (M_{over}), the initial mass in the soil ($M_{soil,i}$) and the final mass in the soil ($M_{soil,f}$). The evaluation of each mass balance term was done stochastically, varying uncertain values randomly within a supposed distribution, and repeating the calculations 10,000 times.

The masses associated with water flows were evaluated according to Eq. 1. Measured event volumes and event mean concentrations (EMC) were used when available, stochastically accounting for uncertainties in the measurements. When measured values were not available, volumes were reconstituted using simple hydrologic models, stochastically varying the parameters of the model, while inlet and drain concentrations were reconstructed stochastically from observed distributions and the overflow concentration was derived from the inlet concentration accounting for dilution.

$$M = \sum V_{ev} C_{ev} \qquad \textit{(Eq. 1)}$$

Where M is the load over the full period, V_{ev} is the flow volume for a given event and C_{ev} is the event mean concentration for the event.

Soil masses was also evaluated stochastically, accounting for uncertainties associated with soil sample representativity, soil concentration analysis, density and the composition of the soil.

The evaluated masses were then used to calculate an integrated mass reduction over the 13-month period (E_{int}, Eq. 2), as well as to compare the intercepted mass ($M_{int}=M_{in}-M_{drain}-M_{out}$) with that accumulated in the soil ($\Delta M_{soil}=M_{soil,f}-M_{soil,i}$).

$$E_{int} = 1 - \frac{M_{drain}+M_{over}}{M_{in}} \qquad \textit{(Eq. 2)}$$

Where E_{int} is the proportion of pollutant mass intercepted by the system, M_{drain} is pollutant load in the outflow from the drain, M_{over} is the pollutant load in the system overflow and M_{in} is the pollutant load from road runoff.

3 RESULTS AND DISCUSSION

3.1 Integrated mass reduction

The integrated mass balance calculations showed mass reductions of between 27-72% for the pollutants studied, with the highest reductions observed for PAH molecules Pyr and Phen, followed by heavy metals Zn and Cu, then TSS, OP, BPA, NP and DEHP (Table 1). Mass reductions of all pollutants were higher than the reduction of water volume, which was only 20%, with 35% overflow and 45% exiting through the drain. Despite the fact that drained water accounted for a greater volume than overflow, overflow accounted for a greater mass for most pollutants than drained water, due to the much lower concentrations observed in the drain than in the untreated road runoff. For example, because PAH concentration reductions generally exceeded 90%, only 7% of Pyr and Phen mass exited through the drain. On the contrary, DEHP concentrations were not significantly reduced and 51% of pollutant mass exited through the drain. The high overflow from this system indicates that increasing surface storage could improve performance.

3.2 Comparison of intercepted mass and mass accumulated in the soil

When comparing intercepted mass (M_{int}) with the mass accumulated in the soil (ΔM_{soil}), no significant dissipation could be observed for any of the pollutants studied. Among the conservative trace metals

studied, M_{int} was within (case of Cu, Figure 2) or slightly below (case of Zn) the 95% confidence interval for ΔM_{soil}. The higher mass of Zn found in the soil may be due to a galvanized barrier between the swale and the road, which did not have contact with water on the reference catchment.

Parameter	E_{int} (%)	M_{over}/M_{in} (%)	M_{drain}/M_{in} (%)
Water	20 (15, 24)	35 (31, 37)	45 (41, 52)
TSS	67 (56, 74)	20 (13, 34)	13 (9, 16)
Cu	63 (54, 69)	23 (17, 33)	14 (10, 20)
Zn	65 (51, 73)	21 (15, 34)	13 (8, 24)
Pyr	72 (60, 79)	21 (14, 34)	7 (4, 13)
Phen	70 (58, 77)	23 (16, 37)	7 (4, 11)
BPA	45 (38, 51)	29 (23, 36)	26 (21, 33)
OP	53 (40, 62)	26 (20, 34)	20 (14, 33)
NP	36 (15, 48)	28 (22, 36)	35 (24, 57)
DEHP	27 (3, 44)	21 (13, 38)	51 (33, 77)

Table 1: Median and 95% confidence interval of mass reduction E_{int}, proportions of inlet mass accounted for in overflow (M_{over}/M_{in}) and in drained water ($M_{,drain}/M_{in}$)

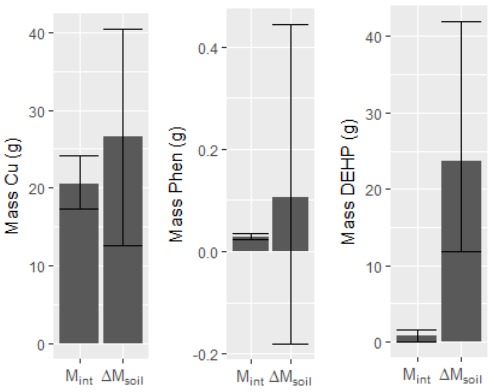

Figure 2: Comparison of intercepted mass to accumulation in the soil for Cu, Phen and DEHP.

Among organic micropollutants, two different issues prevented the observation of dissipation. For PAH molecules, the accumulation was low compared to the initial mass in the soil, leading to very large relative uncertainties in ΔM_{soil}, such that the accumulation was not significantly different from zero and dissipation could not be demonstrated even if it occurred. In the case of BPA, OP, NP and DEHP, M_{int} was much smaller than ΔM_{soil}, indicating that an additional source of these pollutants were present. Indeed, leaching tests on synthetic materials (liner, drain, drain filter fabric, asphalt) used for BFS construction demonstrated that sources of each of these pollutants were present. These emissions may also partially explain the lower mass reductions observed for these pollutants.

4 CONCLUSIONS

The present study demonstrates the utility of a stochastic approach, which may be coupled with experimental measurements in order to evaluate integrated mass balances at the annual scale. A significant mass reduction was observed for all of the considered pollutants (between 27-72%) in the studied biofiltration system, although large overflow volumes limited the performance. In addition, the study showed some of the methodological difficulties associated with evaluating dissipation in the field. Uncertainties in the accumulated soil mass were very high for all pollutants, underlining the importance of accounting for uncertainties when attempting to interpret soil sampling campaigns. The potential of synthetic materials commonly used in biofiltration devices to emit some organic micropollutants, polluting the soil, was also demonstrated. These emissions pose a problem for demonstrating dissipation of these pollutants in situ, but also contribute to contamination of the urban environment and may limit the ability of the system to intercept pollutants over the long term.

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