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► **To cite this version:**

Alexandra Müller, Helene Österlund, Kerstin Nordqvist, Sarah Lindfors, Maria Viklander. Organic micropollutants in stormwater runoff from three urban catchments in Sweden. Novatech 2023, Graie, Jul 2023, Lyon, France. hal-04186335

HAL Id: hal-04186335

<https://hal.science/hal-04186335>

Submitted on 8 Sep 2023

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Organic micropollutants in stormwater runoff from three urban catchments in Sweden

Micropolluants organiques dans les eaux de ruissellement de trois bassins versants urbains en Suède

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RÉSUMÉ

Il est reconnu que le ruissellement urbain contribue à la détérioration de la qualité des eaux de surface et des recherches antérieures ont souligné la nécessité de se concentrer sur les micropolluants organiques. Cette étude présente des mesures de la qualité des eaux pluviales en ce qui concerne les hydrocarbures aromatiques polycycliques, les huiles, les phtalates, les alkylphénols, les composés organostanniques (OTC), les substances polyfluoroalkyles (PFAS) et les polychlorobiphényles (PCB) de trois bassins versants suédois (deux parcs industriels et un parking). Les résultats ont montré que tous les groupes de micropolluants organiques étudiés, à l'exception des PFAS et des PCB, étaient présents dans les eaux pluviales. Les niveaux d'huile étaient similaires dans le flux de base par rapport au ruissellement, tandis que d'autres substances avaient des concentrations plus élevées pendant les événements de ruissellement par rapport au flux de base. Parmi les phtalates analysés, le DEHP et le DINP sont ceux qui ont été détectés à des concentrations supérieures aux limites de déclaration analytique, le DINP étant le plus abondant. Parmi les OTCs, le monobutylétain était le plus abondant, il a été détecté dans tous les échantillons et présent dans les concentrations les plus élevées. En ce qui concerne les différents types de ruissellement étudiés (pluie, fonte des neiges et pluie sur la neige), aucune différence claire n'a pu être identifiée par l'ensemble des données disponibles.

ABSTRACT

Urban runoff is recognised to contribute to the deterioration of surface water quality and previous research pointed out a need to focus on organic micropollutants. This study presents measurements of stormwater quality with respect to polycyclic aromatic hydrocarbons, oil, phthalates, alkylphenols, organotin compounds (OTC), polyfluoroalkyl substances (PFAS) and polychlorinated biphenyls (PCBs) from three Swedish catchments (two industrial parks and one parking lot). The results showed that all the studied groups of organic micropollutants except for PFAS and PCBs were present in the stormwater. The oil levels were similar in base flow compared to runoff, while other substances had higher concentrations during runoff events compared to base flow. Among the analysed phthalates, DEHP and DINP were those that were detected in concentrations above the analytical reporting limits, of which DINP was the most abundant, with a maximum concentration of 140 µg/L. Among the OTCs, monobutyltin was the most abundant, which was detected in all samples and present in the highest concentrations (up to 270 ng/L). Regarding the different types of runoff studied (rain, snowmelt and rain on snow) no clear differences could be identified by the available body of data.

KEYWORDS

Diffuse pollution, Snowmelt quality, Stormwater quality, Urban runoff

1 INTRODUCTION

Urban runoff is widely known to contribute to the degradation of surface water quality, and previous research efforts point to an increased need for research on the presence of organic micropollutants in stormwater and snowmelt runoff from different catchment types (e.g., Zgheib et al., 2012; Müller et al., 2020; Wicke et al., 2021). Some of these micropollutants have documented adverse effects on aquatic life and are, thus, listed as priority pollutants, e.g., in the EU Priority Pollutant Directive for surface water quality (Directive 2013/39/EU), including substances such as polycyclic aromatic hydrocarbons (PAHs), alkylphenols: nonylphenols (NPs) and octylphenols (OPs) and the phthalate di-(2-ethylhexyl) phthalate (DEHP). The reporting of certain organic micropollutants in stormwater from various catchments is uncommon in the existing literature, e.g., considering substances such as diisononyl phthalate (DINP) and the organotin compounds (OTC). In the present study, the following groups of organic micropollutants were monitored in stormwater runoff from three catchments: PAHs, oil, phthalates, NPs and OPs, OTCs, polyfluoroalkyl substances (PFAS) and polychlorinated biphenyls (PCBs).

2 METHODS

2.1 Study sites

Three different catchment areas in Umeå, Sweden, were considered in this study. Umeå is located in the northeast part of Sweden (63°49'30"N 20°15'50"E) and has a municipal population of about 130 000 inhabitants. The catchments studied included two industrial parks (IP1 and IP2) of different sizes and one parking lot (PL). The study catchments and the runoff event characteristics were described in greater detail in a previous publication (Lindfors et al., 2020). IP1 is an industrial park of about 15 ha, while IP2 has a catchment area of about 12 ha. On IP1, oil separators are connected to the stormwater sewers to a larger extent than on IP2. Both IP1 and IP2 has constant base flow in the stormwater sewers. Finally, the study site PL includes exclusively the surface runoff from a parking lot near a hospital, with the area of 0.45 ha. PL does not have base flow in the stormwater sewer between runoff events. The road adjacent to the PL had an annual daily traffic density of 1630 vehicles/day, and the traffic density at the parking lot was assumed to be approximately six and thirteen times higher than at IP1 and IP2, respectively, based on this observation and the catchment sizes (Lindfors et al., 2020).

2.2 Runoff sampling

The samples were collected from nine runoff events, of which three were rain, three were snowmelt, and three were rain on snow, between April 2016 – November 2019. Samples were taken directly from the stormwater sewers. For this, automatic samplers (Teledyne ISCO 6712) were used. Each automatic sampler holds 8 glass bottles. The samplers as well as an Area-Velocity flow meter and logger (Teledyne ISCO 2150) were installed in manholes close to the outlets or connections to adjacent catchment areas of each specific site. At sites IP1 and IP2, the base flow was sampled prior to each runoff event, in order to determine differences between base flow and runoff quality. The sample frequency of each sampling occasion was flow proportional, with 5-8 samples per storm event. Because of joint sampling with another project, the two last events (both rain on snow) were taken as time proportional samples every 10 minutes. The collected samples were thereafter mixed to one composite sample per event. The samples were analysed with respect to 16 PAHs, oil index (C10-C40), the following 13 phthalates: Dimethyl phthalate, diethyl phthalate, di-n-propyl phthalate, di-n-butyl phthalate, diisobutyl phthalate, di-n-pentyl phthalate, di-n-octyl phthalate, di-(2-ethylhexyl) phthalate, butylbenzyl phthalate, dicyclohexyl phthalate, diisodecyl phthalate, diisononyl phthalate, and di-n-hexylphthalate, 4-nonylphenols, 4-tert-octylphenol, and OTCs: monobutyltin (MBT), dibutyltin (DBT), and, tributyltin (TBT), as well as PFAS and PCBs (one event). All samples for chemical analyses were submitted to an accredited laboratory (ALS Scandinavia AB).

3 RESULTS AND DISCUSSION

The concentration ranges (min-max) for selected substances in base flow and runoff from the nine sampling events at the different sites is presented in Tab. 1. PFAS and PCBs were analysed in BF and RO from the first event but was not detected in concentrations above the analytical reporting limits in any of the samples and was therefore excluded from the following sampling campaign. The PAH levels were in general relatively low, e.g., compared to the data presented in Wicke et al., (2021) (mean concentration: 1.7 µg/L for all catchment types), and were highest at site PL that also had the highest traffic densities among the study sites. Moreover, RO concentrations were higher compared to BF at IP1 and IP2, suggesting that the surface runoff in these catchments contributed PAHs to the stormwater.

Table 1 Concentration ranges (min-max) of $\Sigma 16$ PAHs, oil index, di-(2-ethylhexyl) phthalate (DEHP), diisononylphthalate (DINP), 4-nonylphenols (4-NP), 4-tert- octylphenol (4-t-OP) in $\mu\text{g/L}$, and monobutyltin (MBT), dibutyltin (DBT), and tributyltin (TBT) in ng/L in base flow (BF) and runoff (RO) from the three study sites: IP1, IP2 and PL.

Substance	IP1 BF	IP1 RO	IP2 BF	IP2 RO	PL
$\Sigma 16$ PAHs, $\mu\text{g/L}$	<0.095 – 0.34	0.043 – 0.62	<0.095 – 0.041	0.11 – 0.62	0.065 – 2.8
Oil index, $\mu\text{g/L}$	<50 – 11 000	220 - 3200	<50 - 220	<50 - 1900	66 - 3500
DEHP, $\mu\text{g/L}$	<1.0 – 4.7	2.2 – 7.7	<1.0	<1.0 – 7.9	<1.0 - 34
DINP, $\mu\text{g/L}$	<1.0 – 15	4.5 - 50	<1.0	3.8 - 27	1.6 - 140
4-NP, $\mu\text{g/L}$	<0.10 – 1.7	0.23 – 3.8	<0.10 – 0.22	<0.16 – 0.53	<0.220 – 1.3
4-t-OP, $\mu\text{g/L}$	<0.010 – 0.14	0.017 – 0.085	<0.010 – 0.015	0.013 – 0.071	0.04 – 0.95
MBT, ng/L	1.8 - 22	4.14 - 32	1.1 - 20	3.1 - 60	7.1 - 270
DBT, ng/L	<1.0 – 33	<1.0 - 11	<1.0 – 8.0	<1.0 – 65	<1.0 – 31
TBT, ng/L	<1.0 – 1.0	<1.0 – 22	<1.0	<1.0 – 8.3	<1.0 - 24

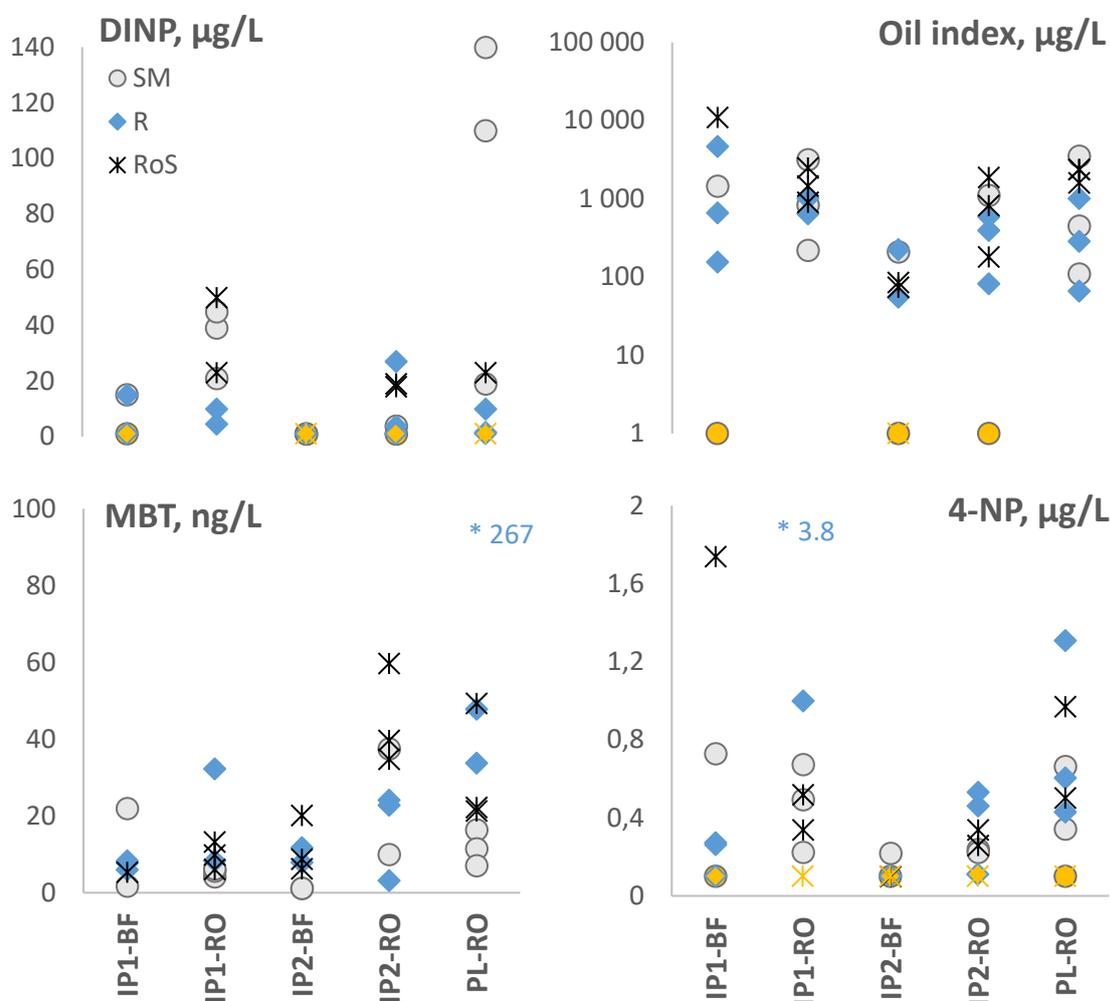


Figure 1: Concentrations of diisononylphthalate (DINP), oil index (C10-C40), monobutyltin (MBT) and 4-nonylphenols (4-NP) in base flow (BF) and runoff (RO) from the three study sites: IP1, IP2 and PL. The symbol colour represents the different kinds of runoff events sampled: grey represent snowmelt (SM), blue represent rain (R), and black represent rain on snow (RoS). The yellow-coloured symbols represent samples that were below the analytical reporting limit.

The oil index varied greatly between the samples, as can be seen both in Tab. 1 and Fig. 1 and was not higher in the runoff samples from IP1 and IP2 compared to the baseflow, indicating that other sources in the sewer network contributed oil to the stormwater. The potential oil sources connected to the storm sewers of IP1 and IP2 include oil separators and a bus washing facility. Among the phthalates, DEHP and DINP were those that were detected in concentrations above the analytical reporting limits, of which DINP was the most abundant. The levels of DEHP and DINP are comparable to those reported in runoff from commercial areas by Wicke et al. (2021) (mean concentrations: 27 µg/L DIDP+DINP, and 2.2 µg/L DEHP, respectively). The highest concentrations of DINP were obtained from the PL site during two snowmelt events (Fig. 1). Moreover, the BF from the two industrial sites (PL1 and PL2) had lower concentrations of DINP compared to the runoff, indicating that runoff contributed DINP to the stormwater. NPs and OPs were frequently detected in the stormwater, in similar concentrations as previous studies. For instance, Zgheib et al. (2012) reported a median concentration of 0.75 µg/L NPs in stormwater in Paris. Among the OTCs, MBT was the most abundant, which was detected in all samples (Fig. 1) and present in the highest concentrations. This is in line with the results from Zgheib et al. (2012), who also found MBT to be most frequently detected OTC, but in somewhat higher concentrations (median 101 ng/L MBT) compared to the present study. From the results reported herein, and presented visually in Fig. 1, there are no clear distinctions between the levels of the studied micropollutants between the different types of runoff (induced by rain, snowmelt, or rain on snow). Such inferences would require a larger body of data and, thus, constitute a suggestion for future research. Finally, it is challenging to predict the environmental effects of the discharge of stormwater into surface water bodies, especially considering dilution effects and the poor understanding of mixture effects of simultaneous exposure to various micropollutants (Masoner et al., 2019).

4 CONCLUSIONS

This study contributed to the knowledge on the occurrence and concentrations of several groups of organic micropollutants in stormwater. PFAS and PCBs were not quantified in any of the samples from the one baseflow and runoff event where it was analysed, while PAHs, phthalates (DINP and DEHP), alkylphenols (NPs and OPs) and OTCs occurred in concentrations similar to those presented in previous studies. In the industrial catchments, IP1 and IP2, the runoff concentrations of PAHs, phthalates and alkylphenols were in general higher compared to the base flow, indicating that the surface runoff in these catchments contributed those substances. No clear differences were visible between the levels of OTCs in runoff and baseflow, and the baseflow levels of oil were higher than those of the runoff, indicating that the other sources than surface runoff contributed to the oil in the stormwater, e.g., oil separators connected to the storm sewers. Regarding the different types of runoff studied, i.e., rain, snowmelt and rain on snow, no clear differences could be identified by the available body of data.

ACKNOWLEDGEMENTS

The study was carried out within the research cluster Stormwater&Sewers and competence centre DRIZZLE funded by Svenskt Vatten Utveckling and Swedish Governmental Agency for Innovation Systems - Vinnova (Grant no. 2016-05176), respectively. Financial support was provided by Formas through the project Undeclared Sources (Grant no. 942-2016-73). VAKIN are acknowledged for providing access and information about their sewers and our colleagues at LTU for assistance with sampling.

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