PAH AND PCB REMOVAL EFFICIENCIES IN UMEÅ (SWEDEN) AND ŠIAULIAI (LITHUANIA) MUNICIPAL WASTEWATER TREATMENT PLANTS

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Abstract. Dissolved concentrations of polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) in the influents and effluents of two municipal sewage treatment plants (STPs) were monitored over 16- (Umeå, Sweden) and 22- (Šiauliai, Lithuania) day sampling periods. Sampling was performed using a passive sampling technique (semipermeable membrane devices; SPMDs) for sequestration of the dissolved (readily bioavailable) fraction of persistent organic pollutants (POPs). Removal efficiencies for individual low molecular weight (LMW) PAH compounds varied from 84% to levels at which the compounds were not detected in effluents from Umeå. The corresponding efficiencies of the Šiauliai plant were 33–95%. Measurements revealed that dissolved concentrations of most of the PCBs and some of the high molecular weight (HMW) PAHs increased during the conventional wastewater treatment at both plants. The release of dissolved PAHs and PCBs in the effluents from municipal wastewater treatment plants might increase the environmental contamination by readily bioavailable pollutants in the recipient waters; the rivers Umeälven and Kulpè. SPMDs were found to be suitable sampling tools for long-term (weeks-month) integrative monitoring of trace concentrations of the dissolved fraction of hydrophobic pollutants in the wastewater process, since the sampling and clean-up steps were easy to perform.

Keywords: analysis, bioavailable fraction, diffusive sampling, effluent, PAHs, PCBs, SPMDs, wastewater treatment efficiency

1. Introduction

Increased water consumption around the world has led to increased concern about the outgoing wastewater quality from municipal sewage treatment plants (STPs) (Liu *et al*., 1997). Modern STP processes include mechanical, chemical and biological treatment steps. However, the STPs commonly receive wastewater from combined domestic, industrial and storm-water sources, and the wide range of different sources leads to numerous kinds of pollutants being found in the wastewater. The main objective of conventional STP is usually to reduce organic material contents and, if necessary, nutrients such as nitrogen and phosphorus. In some cases,

trace concentrations of toxic organic pollutants are also removed, as a side-effect of the intended treatment.

To date, most efforts to monitor the contents of effluents from European STPs (Pham and Proulx, 1997; Wang *et al.*, 2001; Aguayo *et al.*, 2004; European commission, 2005) have focused on traditional parameters that are easily, inexpensively analyzed and regulated by the European Urban Wastewater Directive (91/271/EEC), such as BOD (biochemical oxygen demand), COD (chemical oxygen demand), TSS (total suspended solids), nitrogen and phosphorus. Unfortunately, these routine chemical analyses cannot give a complete overview of either the wastewater treatment efficiency or the threat to the environment posed by contamination associated with the release of persistent and bioaccumulative pollutants, such as persistent organic pollutants (POPs). The new Water Framework Directive (2000/60/EC) has broadened the scope of the earlier directives and presented a list of 'priority substances'; 33 substances which pose a threat to the environmental quality of surface waters in Europe. The Directive will also impose Community-wide emission controls for these priority substances. The occurrence of POPs in the wastewater treatment process has been recognized worldwide and reported in many countries (Manoli and Samara, 1999; Stevens *et al.*, 2003). Data raising concerns about PAHs and PCBs in both raw and treated wastewater have been reported by several authors recently. For instance, Jiries (1999) found total PAH (sum of 16 compounds) concentrations of 1224 ng/L in the raw wastewater and 303 ng/L in the effluent of a plant in Karak (southern Jordan) (Jiries *et al.*, 2000). In addition, Pham found $\Sigma 13$ PCB concentrations of 4.3 and 1.4 ng/L and for average concentrations of PAHs from 1.5 to 0.4 μg/L in raw and treated wastewater, respectively, from the Montreal Urban community (Canada) (Pham and Proulx, 1997). The above data were obtained from tests of traditional organic solvent extracts of wastewater, which represent hydrophobic pollutants from both the aqueous phase and varying amounts originating from the particle-bound phase, depending on the filtration and extraction method. Using this approach, the toxic concentration of the pollutants present may be overestimated, since both the readily bioavailable fraction and some of the less-readily available, adsorbed fractions will be extracted, although only the former possess a immediate risk to aquatic organisms.

Several published studies have considered emissions of the primarily bioavailable hydrophobic pollutants, measured using semipermeable membrane devices (SPMDs), from STPs (Wang *et al.*, 2001; Wang *et al.*, 2003; Stuer-Lauridsen and Kjolholt, 2000; Miege *et al.*, 2004). Low removal rates of target PCB congeners, PAHs, organochlorine pesticides and substituted benzenes were found, and in some cases the concentrations were higher after than before the treatment process.

These studies concluded that SPMDs were suitable passive samplers for time-integrated sampling of the wastewater. Earlier, SPMDs were generally used as inexpensive and semi-quantitative or quantitative sampling tools for concentrating the dissolved fraction of a wide spectrum of organic pollutants from various environmental compartments, such as air, water and sediment pore water (Booij

and van Drooge, 2001; Bartkow *et al*., 2004; Sabaliunas *et al*., 1999; Vrana *et al*., 2001).

In our study the primarily bioavailable PAH and PCB concentrations in the influents and effluents of two different-sized STPs treating sewage with different characteristics were investigated. SPMDs were used in these investigations as good sampling approach since they are integrative tools that are simple to use, and highly efficient for monitoring the *in situ* bioconcentration of ultra-trace levels of PAHs and PCBs in wastewater processes. Environmental exposure (concentrations) to the dissolved fraction of PAHs and PCBs was calculated at both sampling sites from the effluent measurements. Removal efficiencies of POPs from two plants were evaluated.

2. Materials and Methods

2.1. SAMPLING SITES

Two sampling sites were chosen: Umeå (Sweden) and Šiauliai (Lithuania) municipal STPs. Both of the plants have provisions for conventional secondary treatment by an activated-sludge process. The Umeå STP is located on an island in the river Umeälven, which receives the treated wastewater. This treatment plant is located indoors, so the wastewater treatment processes occur under temperature-controlled conditions. The capacity of the treatment plant was around 116 000 PE (population equivalent) during the study. The plant received approximately 30 000 m^3/day of raw wastewater, average amount of TSS in the incoming sewage approximately 300 mg/L and the outgoing 10 mg/L. The removal rate of BOD_7 was, on average, 86% according to routine official analyses reported for the year 2001, which indicates good treatment efficiency (Bristav and Hedlund, 2002).

According to the Baltic Sea Joint Comprehensive Environmental Action Programme (JCP) of the Baltic Marine Environmental Protection Comission (HELCOM), Šiauliai STP (Lithuania) has been identified as a priority Hot Spot, since the discharged wastewater goes to the River Kulpė, one of the most polluted rivers in Lithuania. It is a small river consisting mainly of municipal and industrial wastewater discharges (Sabaliunas *et al*., 2000). The capacity of the treatment plant in the year 2001 was around 150 000 PE, and it received approximately 20 000 m^3/day of raw wastewater, the outgoing wastewater contained 20 mg/L of TSS. The wastewater treatment efficiency, in terms of BOD_7, was 72% (SIDA, 2001).

Samples were taken from both the influent and effluent at the Umeå treatment plant during March and at Šiauliai during October and November 2001. Duplicate samples were taken at the Šiauliai STP to study the sampling reproducibility. Average temperatures of the incoming and outgoing wastewater during the sampling periods in Umeå and Šiauliai were 13 °C and 15 °C, respectively. Both sampling sites for the influent wastewater were located just after the screens in the STPs,

where no treatment of the wastewater was performed. The effluents from the STPs were sampled just before they entered the river as cleaned, undiluted wastewater.

2.2. SAMPLING WITH SPMDS

Sampling was performed using passive sampling, standard size SPMD with Expos-Meter lipophilic from ExposMeter AB (Tavelsjö, Sweden), which sequester water-dissolved and vapor phase organic pollutants in water and air, respectively. Triolein enclosed in the membrane accumulates trace/ultra-trace levels of dissolved hydrophobic organic pollutants during the exposure period. The accumulated amount is related to the time-weighted average (TWA) concentration of the pollutants in the water. The amount of water extracted by a standard 1 mL triolein SPMD may approach 150–300 L for a thirty-day exposure to compounds with moderately high octanol/water partitioning coefficients (K_{ow}s) (Huckins *et al.*, 2002).

The wastewater flow was turbulent at all sampling points. Water flows was higher in the incoming wastewater locations, but since the membranes were enclosed in a deployment canister the water flow velocity on the membrane surfaces were strongly reduced. The water flow does probably not influence the sampling efficiency differently between points. However, the buildup of sludge inside some canisters might have reduced the water exchange to the membranes, which reduce the uptake and, thus, the measured water concentration. Different types of biofouling at the sampling points might also have a slight influence on sampling rate, but the rate can not be estimated without the use of performance reference compounds.

Before and after the sampling all membranes were stored at −20°C, in sealed, acetone-washed, tin cans. To provide field controls (FC) for the exposure during deployment in the study areas, a single SPMD was exposed to air during the deployment and retrieved in the same manner as the membranes used for sampling at each site

2.3. MEMBRANE EXTRACTION, CLEAN-UP AND ANALYSIS

Each retrieved membrane was treated as detailed by Söderström *et al.* (Söderström and Bergqvist, 2003).We used aluminum foil to cover the glassware in order to reduce photodegradation of PAHs during the analysis steps. After the dialysis, three [2]H-labeled PAHs and ten ^{13}C-labelled PCBs (Cambridge Isotope Laboratories, Inc. U.S.A.) were added as internal standards (IS) to evaluate losses during clean-up. [2]H-labeled dibenzofuran (Promochem, Kungsbacka, Sweden) was added as a recovery standard (RS). Samples were analyzed using HRGC/LRMS. The native standard mixture contained 24 PAH compounds obtained from Promochem (SRM 2260, Kungsbacka, Sweden) and six PCB compounds obtained from Larodan Fine Chemicals AB (L180013, Malmö, Sweden).

A laboratory blank (LB) consisting of only the appropriate solvents and cleanup material used during analysis was used in both sample batches. The LB value was

10% or less of the sample values for all of the analyzed compounds. The analytical results were checked for possible contamination during handling by comparing them to the appropriate field controls (FC), values was accepted when FC amounts ≤30% of individual compounds in the samples, otherwise compounds were not quantified.

The method limit of detection (LOD) was calculated as the quantity of a pollutant that gave a response three times higher than the baseline noise of the pollutant chromatogram at the expected retention time. Recoveries for the surrogate internal standards were calculated and used to correct the final results. The recovery values were typically between 46–114% for PAHs and 60–120% for PCBs.

2.4. WATER-DISSOLVED CONCENTRATIONS AND REMOVAL EFFICIENCY

Depending on the physico-chemical properties of the pollutants of interest, environmental conditions and duration of the sampling, the analytes sequestered by the membrane may be in the linear uptake (integrative sampling), curvilinear or equilibrium partitioning phase of sampling. To evaluate readily bioavailable concentrations of PAHs in equilibrium and linear uptake sampling ranges a simplified equations 1 and 2 were used (Huckins *et al.*, 2002). PAHs with log $K_{ow} < 4.0$ (naphthalene 3.5, 1-methylnaphthalene 3.9 and biphenyl 3.9 (Mackay D. 1992)) was considered to be at equilibrium uptake phase after the two to three weeks sampling periods, water concentrations were calculated according to Equation 1:

$$C_w = \frac{N_s}{K_{sw} \cdot V_s} \tag{1}$$

where C_w is concentration of the pollutant in water (ng/L), N_s is the amount of the pollutant collected by one standard-sized ExposMeter lipophilic membrane (ng) and K_{sw} is SPMD-water partitioning coefficient (naphthalene 3793, 1-methylnaphthalene 8851, biphenyl 9594 $L \cdot L^{-1}$) and V_s is volume (4.95 cm^{-3}). For PAHs at linear uptake range Equation 2:

$$C_w = \frac{N_s}{R_s \cdot t} \tag{2}$$

where t is the sampling time (days), uptake rates R_s (L/day) at 10°C from Huckins (Huckins *et al.*, 2002; Huckins *et al.*, 1999) were used to calculate the *in situ* dissolved concentrations of compounds. Uptake rates for other PAH compounds that are no available data, were estimated from compounds with similar physical properties. The following, simplified expression was used for all studied PCBs:

$$C_w = \frac{N_s}{1.2649 \cdot t \cdot \exp(0.0764 \cdot T)} \tag{3}$$

where T is the average temperature of the wastewater during the sampling period. To evaluate the wastewater treatment efficiency in both plants the removal of primarily available detected PAH and PCBs was evaluated using eq. 4 (Pham and Proulx, 1997):

$$R = \frac{(C_{IN} - C_{OUT})}{C_{IN}} \times 100\% \tag{4}$$

where R is the removal rate (%), C_{IN} is the dissolved concentration of the compound in the wastewater influent (ng/L) and C_{OUT} is the dissolved concentration of the compound in the wastewater effluent (ng/L).

3. Results and Discussion

3.1. PAH RESULTS

Twenty-four individual PAHs were examined in the membrane extracts from both sampling sites: the 16 U.S. EPA-listed priority pollutant PAHs (pp PAHs); five methylated PAHs (1-methylnaphthalene, 2-methylnaphthalene, 2,6-dimethylnaphthalene, 2,3,5-trimethylnaphthalene, 1-methylphenanthrene); and three others—biphenyl, benzo(e)pyrene and perylene. The physico-chemical properties of individual PAHs influence their behavior in aqueous environments. PAHs with a molecular weight <200 g/mol, that have two or three aromatic rings and a log $K_{ow} < 5$ were defined as low molecular weight (LMW PAHs). The remainder, from fluoranthene to dibenzo(a,h)anthracene, were defined as high molecular weight (HMW) PAHs. Low molecular weight PAHs are partly dissolved and highly available for degradation processes (Lundstedt, 2003). In contrast, high molecular weight PAHs are less soluble and volatile, so they are mainly associated with particles and less available for degradation in the water.

3.1.1. *Results for Low Molecular Weight PAHs*
The results from Umeå STP (Figure la)   showed that LMW PAHs were detected and the sum of 11 analyzed LMW PAHs (all except fluorene), was 380 ng/L in the incoming wastewater. Contamination of the field control sample with fluorene, or a compound with the same m/z and retention time, was detected, and, thus results for this compound are not presented. The most abundant pollutants entering the treatment plant were methyl-PAHs, such as 2-methylnaphthalene (130 ng/L), 1-methylnaphthalene (85 ng/L), 2,6-dimethylnaphthalene (36 ng/L) and 2,3,5-trimethylnaphthalene (21 ng/L). The domination of the pollution profile by methylated low molecular weight PAHs is indicative of typical wastewater contamination by petroleum products.

Light PAHs in Umeå STP were removed from the wastewater effectively during the treatment process; removal efficiencies for individual PAH compounds varied
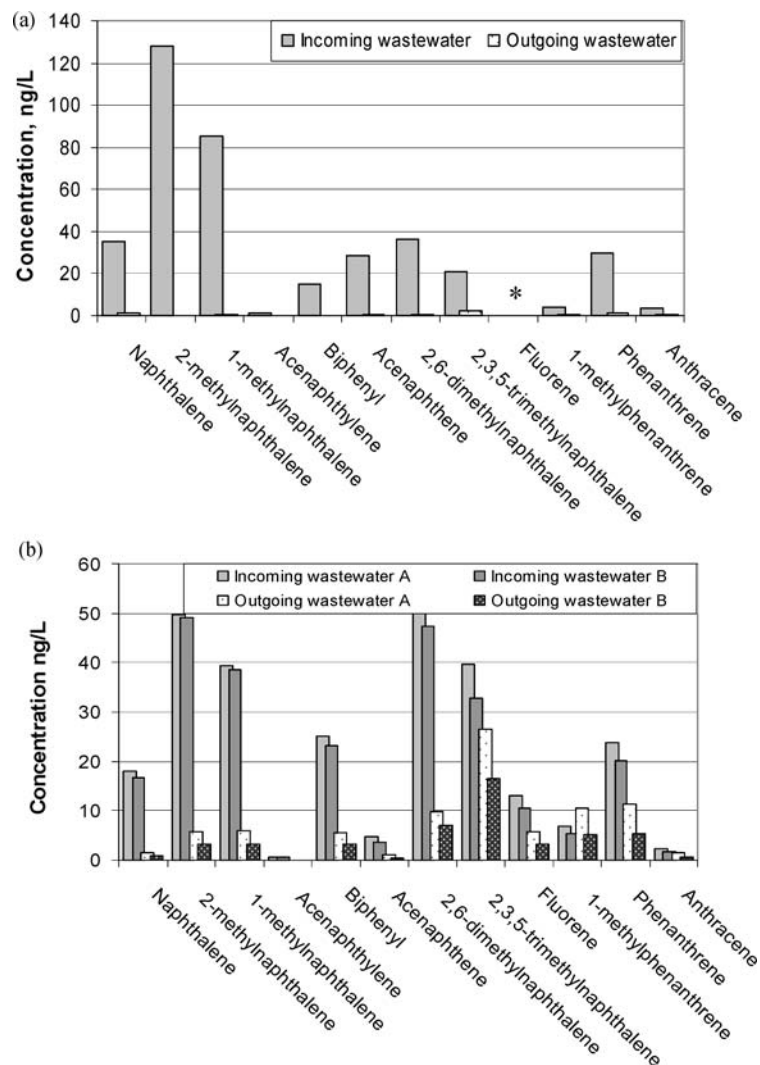
Figure 1. Concentrations of primarily bioavailable low molecular weight PAHs in the influents and effluents of the treatment process at (a) Umeå and (b) Šiauliai *Fluorene not analyzed.

from 84% to levels at which the compounds were not detected in the outgoing wastewater. The highly turbulent flow of the wastewater and their high availability for biodegradation in the treatment process could be the reasons for the high removal rate of the dissolved LMW PAHs in this plant.

Unlike the Swedish plant, the Lithuanian STP had very low removal rates for some of the investigated LMW PAHs, for example 2,3,5-trimethylnaphthalene, 1-methylphenanthrene, phenanthrene and anthracene (Figure 1b). The concentrations of these compounds were slightly decreased after the treatment process

(R = 33–61%), but the concentrations of some compounds appeared to be higher in the effluent than the influent (e.g. there was a calculated increase in the concentration of 1-methylphenanthrene of approximately 50%). The methyl-PAHs, 2,6-dimethylnaphthalene (54 ng/L), 2-methylnaphthalene (50 ng/L), 2,3,5-trimethylnaphthalene (40 ng/L), 1-methylnaphthalene (39 ng/L), were also the most abundant pollutants detected in the incoming wastewater of the Lithuanian plant (Figure 1b).

The relative percent difference (RPD, Equation 5) can be used to investigate the variability of individual samples from the same sampling point (duplicates) (US EPA, 2005), and thus was used to describe the precision between pairs of observations:

$$RPD = (|SR - DSR|/((SR + DSR)/2) \times 100\% \qquad (5)$$

where SR is the sample result for a particular analyte and DSR is the duplicate sample result for that analyte.

The RPD of duplicate samples for individual LMW PAHs varied from 1–35% for the influent and 11–50 (in some cases up to 76)% for the effluent wastewater at the Šiauliai treatment plant. The highest variation was seen for individual compounds detected close to their determination limit. Repeatability results for SPMDs are commonly ≤25%. The Lithuanian plant treatment process seems not to remove LMW PAHs as efficiently as the Umeå plant; the sum of 12 studied LMW PAHs (fluorene included) decreased R = 76% by average in the effluent, from the original concentration of 270 ng/L in the influent.

3.1.2. *Results for High Molecular Weight PAHs*

The physico-chemical properties of the 12 HMW PAHs suggest that their behavior during the treatment process should differ from that of the LMW PAHs, since the high lipophilicity of HMW PAHs increases their adsorption to the small particles in the wastewater during the treatment process. Thus, pollutants present in the wastewater might re-partition between the water phase and particle phase during the course of the chemical and biological processes involved in the wastewater treatment.

The dissolved fraction analysis of HMW PAHs suggested that the concentration increased, or at least there was no significant removal, of four- and some of the five-ringed PAHs (benzo(a) anthracene, chrysene, fluoranthene, pyrene, benzo(b) fluoranthene, benzo(k)fluoranthene, benzo(e)pyrene, benzo(a)pyrene, perylene) during the treatment process at both sampling sites (Figure 2(a&b)). Five- and six-ringed PAHs were present at low (sometimes less than 0.5 ng/L) concentrations which were close to the detection limits and, thus, no trends can be discerned for the low level compounds.

Results from the Umeå STP plant indicated that the most abundant PAHs in the influent and effluent wastewater were four-ringed PAHs, particularly pyrene
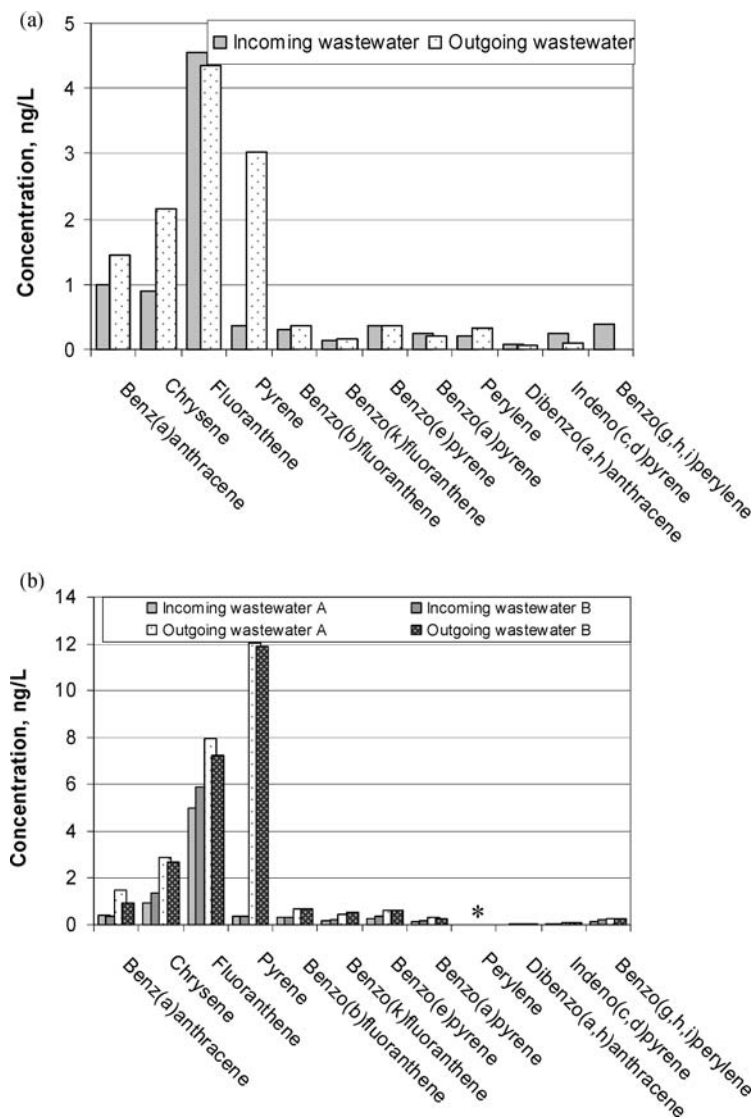
Figure 2. Concentrations of primarily bioavailable high molecular weight PAH in the influents and effluents of the treatment process at (a) Umeå and (b) Šiauliai *Perylene was not detected in the samples.

(Figure 2a). The concentration of dissolved fluoranthene remained unchanged during the entire wastewater treatment process, which means that the treatment did not effectively remove the dissolved phase of this compound. The dissolved concentration of pyrene in the effluent was 10 times higher than the concentration in the influent water.

Removal rates for 5- and 6 ringed HMW PAHs (except benzo(b) and (k)fluoranthene, perylene, benzo(e)pyrene and perylene) varied from 18–60%, but most of the studied HMW PAHs were not removed at all, or there may even have been an increase in their dissolved water concentration during the treatment process. The sum of the 12 studied HMW PAHs increased from 8 to 12 ng/L.

The sum concentration of 12 studied HMW PAHs from Šiauliai plant increased from 8 ng/1 in the influent to 27 ng/1 in the effluent. The most abundant compounds detected in the samples were four-ringed PAHs, as in the Swedish case. The effluent had 50–150% higher dissolved concentrations of four-ringed PAHs compared to the influent, before the treatment, except pyrene where the level increased 32-fold. The treatment process did not appear to remove the dissolved fraction of HMW PAHs efficiently.

3.2. PCB RESULTS

Ten analyzed polychlorinated biphenyl congeners (PCB_{10} 28, 47, 52, 101, 105, 118, 138, 153, 156 and 180) were quantified and evaluated in the samples. The readily bioavailable concentrations of PCB_{10} increased from 0.3 to 1 ng/L in Umeå and from 9 to 34 ng/L in Šiauliai STPs during the treatment process, and the sum of 7 PCBs (PCB_7, excluding PCBs 47,105,156) increased from 0.3 to 1 ng/L and from 1 to 6 ng/L, respectively. The most abundant PCBs in the incoming wastewater in Šiauliai were tri- and tetra-chlorinated congeners. High concentrations of these lightly chlorinated biphenyls might suggest that not weathered PCBs are leaching into the wastewater system.

In comparison, the Swedish plant's influent wastewater had relatively equal concentrations of tri-and tetra-, compared to penta- and hexa-chlorinated biphenyls (such as PCBs 118, 138 and 153). This indicates that a leakage from old PCB pollution sources could be the source of contamination of the Swedish wastewater, or specific uses of highly chlorinated PCBs. The dissolved concentrations of PCBs 156 and 180 were close to detection levels in both sample batches, making it difficult to evaluate the treatment efficiency of these highly hydrophobic PCBs.

The increase in PCB_{10} suggests that the treatment processes are not efficiently removing PCBs during the wastewater treatment process at either plant.

4. Conclusions

Readily biologically available (dissolved) PAH and PCB concentrations were studied in two different STPs. The results demonstrated that the dissolved concentration of four-ringed PAHs, some of the five-ringed PAHs and most of the studied PCBs were higher in the effluent, after the treatment process, than in the influent. In the effluent from the treatment plants, the studied priority pollutants might pose higher toxic risks than the particle-bound pollutants. Thus, the apparent increase in

the concentrations of these compounds in the dissolved fraction during the treatment process is a concern for the treatment method and maybe also for the outside environment.

Removal efficiencies for individual LMW PAH compounds varied from 84% to virtually 100% for LMW PAHs in the Umeå STP and from 33–95% in the Šiauliai STP. The reason why the Umeå STP had a more efficient treatment process with respect to the dissolved fraction of LMW PAHs, compared to the Lithuanian plant cannot be explained by the data collected in this study. In both plants methyl-PAHs were the most abundant compounds detected in the samples of incoming wastewaters.

The effluents of both treatment plants had higher concentrations of HMW PAHs (such as chrysene, pyrene, benzo(b)fluoranthene and benzo(k)fluoranthene) than the incoming water. The largest increase was seen for pyrene, which increased 10-fold in the Umeå STP and 32-fold in the effluent from the Šiauliai STP. Fluoranthene remained at the same level (Umeå STP), whereas removal rates for other HMW PAHs varying from 5–60% for individual compounds at the Umeå plant. In the Lithuanian plant, studied HMW PAH concentrations were unchanged or only slightly increased after the wastewater treatment process. The low removal efficiency and the increase in bioavailable HMW PAHs confirmed that the conventional wastewater treatment process is not designed for the removal of this group of compounds and the effluents from the plants introduce bioavailable HMW PAHs to the recipient water bodies.

PCB levels detected in the influents and effluents at both plants were not high, but the increase in bioavailable concentrations during the treatment process confirmed the inability of the wastewater treatment process to remove bioavailable PCBs.

SPMDs were shown to be inexpensive and labor saving sampling tools, suitable for monitoring the influents and effluents of STPs in different locations. The influence of biofouling was not possible to determine in this study. Further studies should examine the factors that influence repartitioning between the water and particle phases of dissolved HMW PAHs and PCBs, the increase in the dissolved concentrations of these compounds and the evaporation of LMW PAHs into the air. The membrane sampling could utilize performance reference compounds spiked sampling membranes for a better understanding of biofouling and water turbulence influence of the data. We need a better understanding of the wastewater treatment process efficiency regarding bioavailable PAHs and PCBs.

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