Partitioning of polycyclic aromatic hydrocarbons, alkylphenols, bisphenol A and phthalates in landfill leachates and stormwater

Yuliya Kalmykova [a,*], Karin Björklund [b], Ann-Margret Strömvall [a], Lena Blom [c]

[a] Department of Civil and Environmental Engineering, Chalmers University of Technology, 412 96 Gothenburg, Sweden
[b] Department of Civil Engineering, The University of British Columbia, 6250 Applied Science Lane, Vancouver, B.C., Canada V6T 1Z4
[c] Water and Recycling Office of Gothenburg City, 404 24 Gothenburg, Sweden

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ABSTRACT

Partitioning of organic pollutants is essential to their fate, mobility and removal from water and soil. To study the partitioning behavior of selected alkylphenols, bisphenol A, phthalates and polycyclic aromatic hydrocarbons (PAHs), a method for separating the truly dissolved and colloidal phase of organic pollutants was developed, verified and applied to samples of landfill leachate and stormwater from urban areas and waste-sorting sites.

Alkylphenols, bisphenol A, phthalates and PAHs were detected in all the untreated samples (total concentrations), most of the filtered samples and frequently in the colloid-bound phase. Concentrations of alkylphenols and PAHs in urban stormwater were one order of magnitude lower than in the landfill leachates and stormwater from waste-sorting sites.

The difference between total, dissolved and colloid-bound concentrations in the water samples was not statistically significant for any phenols or phthalates, but for three of the PAHs; naphthalene (mostly dissolved), phenanthrene and fluoranthene (mostly particulate). These results indicate that in landfill leachates and stormwaters, organic pollutants are predominantly attached to colloids and/or truly dissolved in contrast to their expected strong sorption to particulate matter. Occurrence and concentrations of pollutants in dissolved and colloid-bound phases correlated negatively with the K_{OW}. However, even highly hydrophobic compounds were frequently detected in filtered samples, i.e. the dissolved phases, and it is suggested that the organic content in the colloids decreases the compounds' partition to particles. The results confirm that the K_{OW} values of specific organic pollutants well describe the compounds partition-binding process to dissolved organic carbon (DOC) colloids. Our findings call for a re-assessment of the organic pollutants' mobility and associated risks. This knowledge can also serve as a base for selecting efficient treatment methods for stormwater and landfill leachates.

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1. Introduction

In this study, partitioning of selected alkylphenols, bisphenol A, phthalates and polycyclic aromatic hydrocarbons (PAHs) in landfill leachate and stormwater is investigated. The substances have been selected based on their expected occurrence in urban water streams and potential negative effects on aquatic environments. Alkylphenols – including 4-

nonylphenol (4-NP), 4-*t*-octylphenol (4-*t*-OP) and 4-*t*-butylphenol (4-*t*-BP) – bisphenol A (BPA), phthalates and PAHs have previously been reported in wastewater, urban snow, runoff and landfill leachate (Björklund et al., 2009, 2011; Sánchez-Avila et al., 2009; Slack et al., 2005; Zgheib et al., 2011). The substances' ubiquitous occurrence in urban water streams is a reflection of their widespread use and production in society. The PAHs are emitted from combustion and petroleum sources whereas the phenols and phthalates are emitted mainly from consumer products and building materials (Neilson, 1998; Björklund, 2010). As several of the selected PAHs, phenols and phthalates show acute toxic, carcinogenic and estrogenic effects on aquatic organisms, their occurrence in leachate and stormwater may pose a threat to receiving water quality (ECB, 2010; Neilson, 1998; Servos, 1999; Staples et al., 1997a).

As many organic compounds are hydrophobic by definition, it is often assumed that they can be removed from the water phase through removal of particles. In addition to the aqueous and particulate phase, organic pollutants may also adsorb to colloids, sometimes referred to as "the third phase". Studies have shown that organic compounds may adhere to organic colloids in marine waters, groundwater and soil (Chin and Gschwend, 1992; Kan and Tomson, 1990; Shimizu et al., 1998; Villholth, 1999). Lee and Kuo (1999) have for synthetic solutions analytically quantified the effect of dissolved organic carbon (DOC) presence on the decreased partitioning of organic pollutants to the solid phase. As an example, for a DOC concentration of 25 mg/L, the relative decrease in the partition of organic pollutants to particulates was as high as 300% for a compound with log K_{OW} = 6 (Lee and Kuo, 1999).

Knowledge on partitioning of organic pollutants in urban waters will assist in estimation of their mobility, toxicity and associated risks (Ditoro et al., 1991). The bioavailability of organic pollutants has been shown to depend on their form; the fraction binding to DOC tends to show lower bioavailability to aquatic animals than the truly dissolved fraction (Haitzer et al., 1998). On the other hand, DOC colloids may act as a carrier for organic pollutants and enhance their transport, i.e. colloid-facilitated transport, in groundwater, soil and other porous media (Jaradat et al., 2009). Because colloid-bound pollutants acquire different charge, size and stoichiometry compared to truly dissolved compounds they may travel through particle and adsorption filters (Shimizu et al., 1998; Forstner et al., 2001; Kalmykova et al., 2010). Thus, treatment methods for landfill leachates and stormwater need to be selected based on knowledge of organic pollutants partitioning. The importance of investigating the partitioning of organic pollutants between the truly dissolved, colloidal and particulate phases has also been stressed in a review on surface water quality monitoring programs by Vignati et al. (2009).

Wastewaters such as stormwater and landfill leachate are different regarding the origin and accumulation of organic pollutants, and the specific chemical and physical composition of the waters. The pollutant content of landfill leachates reflects the historical use of chemicals in the society, whereas stormwater is a sink for chemicals currently in use. Stormwater composition varies with storm event and catchment area (Björklund et al., 2009), and landfill leachates vary in composition depending on the type of waste, year of deposition, and the age of the landfill (Kjeldsen et al., 2002). Anaerobic conditions, which may be expected in landfills, have been shown to prevent or retard the degradation of phthalates (Yuan et al., 2002; Staples et al., 1997b). However, studies by both Kjeldsen et al. (2002) and Baun et al. (2004) suggest that degradation of phthalates in landfills or leachates may occur. Alkylphenolic compounds may degrade under both aerobic and anaerobic conditions (Isobe et al., 2001; Chang et al., 2004). Alkylphenol ethoxylate degradation was assumed to have taken place in a Danish landfill, where Baun et al. (2004) could not detect any nonylphenol ethoxylate isomers but only the degradation product nonylphenol ethoxycarboxylate. Angelidaki et al. (2000) showed that PAHs were biodegraded under both aerobic and anaerobic conditions in landfilled sewage sludge. Anaerobic degradation of PAHs is, however, in most cases slower than aerobic degradation (Eriksson et al., 2003).

The aim of this study is to investigate the partitioning of selected alkylphenols, bisphenol A, phthalates and PAHs in landfill leachate and stormwater samples. The objectives are to: i) determine concentrations of pollutants in the total, dissolved and colloidal fractions; ii) investigate whether occurrence in different fractions can be explained by the pollutants' physico-chemical properties and/or by certain properties of the investigated waters. The hypotheses are that: i) the PAHs, 4-NP, 4-*t*-OP and the high molecular weight (HMW) phthalates are adsorbed to particles in the aquatic environment (Ying et al., 2003; Cousins et al., 2003; Mackay and Callcott, 1998); ii) the <C_6 phthalates, the alkylphenol ethxoylates, BPA, 4-*t*-BP and 4-*t*-pentylphenol (4-*t*-PP) are found partly dissolved in water due to their higher water solubility and iii) organic compounds may show higher than expected water solubility in landfill leachates and stormwater due to the presence of DOC.

2. Experimental

2.1. Sampling

Leachate samples were collected at four landfills in the Gothenburg region, Sweden. The Brudaremossen landfill was in use 1936–1976 and contains a mixture of household and industrial waste with a high organic content. At this site four samples were collected: BR1 – raw leachate, BR2 – leachate after oil separator, BR3 – leachate after oil separator and sedimentation pond, and BR4 – a separate untreated raw leachate flow. At the Sörmossenlandfill, in use 1938–1978 with mainly high organic content industrial waste, a raw leachate sample (SR) was collected. One sample (FL) of raw leachate was collected from the most recently constructed Fläskebo landfill (2003), containing industrial waste and contaminated soil with an organic content below 5%. The Tagene landfill has been in use since 1974 and contains residues from municipal solid waste incineration (raw leachate sample TA1). In one section of the landfill, sewage sludge was deposited until the 1980s (raw leachate sample TA2).

Stormwater was collected at Högsbo and Skräppekärr waste-sorting sites (HB and SK), which handle predominantly industrial and bulky waste. The waste is left uncovered, sometimes for extended periods, before being sorted and transported to a final destination. Stormwater was also collected from urban areas: sewers draining a motorway with an annual average daily traffic of ∼85 000 vehicles at Gårda (G1), and from the Järnbrott area with a major road, housing and small-scale industrial surfaces (J2).

All waters were grab sampled and collected in dark solvent rinsed glass bottles, kept at +4 °C and directly sent to the laboratory for chemical analysis.

For all landfill leachates and stormwater studied in this project, data on other chemical parameters than organic pollutants are presented in Supplementary Table S1. These data show that several of the general measured parameters, concentrations are in the same order for all the different wastewaters. The most important differences are that the landfill leachates contain much higher concentrations of main elements such as for example Ca, Fe and Mn, nutrients and Cl. The stormwater samples, on the other hand, show much higher concentrations of toxic trace metals as Cd, Cr, Cu, Pb, Ni and Zn. The result in Table S1 also indicate that landfill leachates have higher concentrations of total organic carbon (TOC), often correlated to high DOC concentration, whereas the stormwater have much higher concentrations of solid particles measured as total suspended solids (TSS).

2.2. Separation of fractions

Included in the study are 16 specific PAHs, 4-t-octyl- and 4-nonylphenol and their ethoxylates, three phenols and eight phthalates, see Tables 1 and 2 and Supplementary Table S2. The total, particle bound, colloid bound and truly dissolved concentrations were determined for all compounds, following a procedure modified and developed from the determination of colloidally associated PAHs in water using C_{18} solid-phase extraction (SPE) disks (Brown and Peake, 2003). The determination of organic pollutants was performed in steps as follows:

- Total concentrations were determined on unfiltered samples using solvent liquid–liquid extraction (LLE).

Table 1 – Analyzed phenol and phthalate (µg/l) total concentrations, number of samples (n) and samples with concentrations above the detection limit (>d.l.) in the current study, compared with water quality standards and results from screenings of runoff and landfill leachate.

Compound		This study[a]		Water quality standards and guidelines[b]	Runoff, waste sorting sites and urban, Sweden[g]		Landfill leachates, Sweden	
		Range (median)	n (>d.l.)	AA/MAC/LT[c]	Range (median)	n (>d.l.)	Range (median)	n (>d.l.)
4-Nonylphenol	4-NP	<0.1–7.3 (0.26)	13 (12)	0.3/2.0/1[d]	<0.10–4.5 (0.20)	16 (10)	<0.2–5.7 (2.5)[i]	12 (7)
Nonylphenol monoethoxylate	NP1EO	<0.1–3.6 (<0.1)	13 (4)	–[e]	<0.20–14 (<0.20)	11 (4)	<0.2–2.4 (0.23)	12 (6)
4-tert-Octylphenol	4-t-OP	<0.01–1.3 (0.23)	13 (10)	0.1/NA[f]/–	<0.20–0.35 (<0.20)	8 (4)	n.a.	
Octylphenol monoethoxylate	OP1EO	<0.01–5.33 (0.031)	13 (10)	–	<0.20–1.4 (<0.20)	11 (4)	<0.2–15 (0.033)	12 (8)
4-tert-Butylphenol	4-t-BP	0.027–8.1 (0.97)	13 (13)	–	n.a.[h]		n.a.	
4-tert-Pentylphenol	4-t-PP	<0.1–0.66 (0.062)	13 (10)	–	n.a.		n.a.	
Bisphenol A	BPA	<0.01–107 (0.55)	13 (10)	–	n.a.		n.a.	
Dimethyl phthalate	DMP	<d.l.	11 (0)	–	<0.10–60 (<0.10)	18 (5)	n.a.	
Diethyl phthalate	DEP	<0.10–22 (<0.10)	11 (4)	–/–/19	<0.10–49 (0.20)	18 (11)	<1–5 (<1)[j]	20 (*)[k]
Di-n-butyl phthalate	DnBP	<0.10–1.8 (0.30)	11 (9)	–	<0.10–63 (0.30)	18 (7)	<1–4 (<1)	20 (*)
Butyl benzyl phthalate	BBP	<0.48–0.76 (0.27)	11 (8)	–	<0.10–12 (<0.10)	18 (3)	<1–1(<1)	20 (*)
Di-(2-ethylhexyl) phthalate	DEHP	<1.0–23 (1.8)	11 (10)	1.3/NA/16	<1.0–47 (<1.0)	18 (7)	n.a.	
Di-n-octyl phthalate	DnOP	<0.10–0.28 (<0.10)	11 (2)	–	<0.10–0.16 (<0.10)	17 (1)	n.a.	
Diisononyl phthalate	DINP	<1.0–70 (<1.0)	11 (4)	–	<1.0–85 (1.2)	13 (9)	n.a.	
Diisodecyl phthalate	DIDP	<d.l.	11 (0)	–	<1.0–17 (0.58)	13 (9)	n.a.	

a All analysis data for each specific sample are available in Table S2 in the Supplementary Information.
b Environmental quality standards in the field of water policy (2008) and Water Quality Guidelines for the Protection of Aquatic Life (CCME, 2011).
c AA: annual average; and MAC: maximum allowable concentration for inland surface waters; LT: freshwater, long term exposure.
d Nonylphenol and its ethoxylates.
e – = No available standard or guideline.
f NA = not applicable.
g Junestedt et al. (2003) and Björklund et al. (2009).
h n.a. = Not analyzed.
i Cerne et al. (2007).
j Öman and Junestedt (2008).
k * = Not reported. i.e. the number of samples above the d.l. was not reported. The detection limit was 1 µg/l as indicated by the minimum values (i.e. <1).

Table 2 – Analyzed specific PAH total concentrations (μg/l), number of samples (n) and samples with concentrations above the detection limit (> d.l.) in the current study, compared with water quality standards and results from screenings of runoff and landfill leachate.

Compound		This study[a]		Water quality standards and guidelines[b]	Runoff waste sorting sites, Sweden[f]		Landfill leachates, Sweden[h]	
		Range (median)	n(>d.l.)	AA/MAC/LT[c]	Range (median)	n (>d.l.)	Range (median)	n (>d.l.)
Naphthalene	NAP	0.02–3.8 (0.05)	11 (11)	2.4/NA[d]/1.1	<0.34–0.80 (<0.34)	6 (2)	<0.05–8.0 (<0.05)	22 (*)[i]
Acenaphthylene	ACY	<0.01–0.09 (<0.01)	11 (3)	–[e]	<0.25	6 (0)	<0.05–1.8	20 (*)
Acenaphthene	ACE	<0.01–1.1 (<0.01)	11 (5)	–/–/5.8	<0.02–0.10 (<0.02)	6 (2)	<0.05–1.3 (0.01)	20 (*)
Fluoranthene	FL	<0.01–1.5 (0.02)	11 (7)	0.1/1/3	<0.03–0.60 (0.33)	6 (5)	<0.01–0.87 (0.01)	20 (*)
Phenanthrene	PHE	<0.01–1.4 (0.11)	11 (9)	–/–/0.4	<0.03–10 (1.1)	6 (5)	<0.05–2.7 (0.05)	20 (*)
Anthracene	ANT	<0.01–0.50 (<0.01)	11 (4)	0.1/0.4/0.012	<0.013–0.70 (0.26)	6 (5)	<0.01–0.3 (<0.01)	20 (*)
Fluorene	FLU	<0.01–1.1 (0.01)	11 (7)	0.1/1/0.04	<0.02–8.7 (0.95)	6 (4)	<0.05–2.5 (<0.05)	20 (*)
Pyrene	PYR	<0.01–0.91 (0.05)	11 (8)	–/–/0.025	<0.037–5.8 (0.90)	6 (5)	<0.01–0.5 (0.015)	20 (*)
Benzo[a]anthracene	BaA	<0.01–0.32 (0.02)	11 (6)	–/–/0.018	<0.007–1.6 (0.20)	6 (5)	<0.01–0.07 (<0.01)	20 (*)
Chrysene	CHY	<0.01–0.38 (0.03)	11 (7)	–	<0.016–1.9 (0.45)	6 (5)	<0.01–0.08 (<0.01)	20 (*)
Benzo[b,k]fluoranthene	BbkF	<0.01–0.41 (0.03)	11 (6)	0.03/NA/–	<0.016–1.4 (0.15)[g]	6 (5)	<0.01–0.2 (<0.01)	20 (*)
Benzo[a]pyrene	BaP	<0.01–0.20 (<0.01)	11 (3)	0.05/0.1/0.015	<0.026–1.5 (0.08)	6 (4)	<0.01–0.07 (<0.01)	20 (*)
Benzo[g,h,i]perylene	BPY	<0.01–0.12 (<0.01)	11 (3)	$\sum$ = 0.002/NA/–	<0.004–1.0 (0.04)	6 (4)	<0.03–0.09 (<0.03)	20 (*)
Indeno[1,2,3-cd]pyrene	INP	<0.01–0.09 (<0.01)	11 (4)		<0.006–1.0 (0.05)	6 (4)	<0.01–0.03 (<0.01)	20 (*)
Dibenzo[a,h]anthracene	DBA	<0.01–0.19 (<0.01)	11 (2)	–	<0.012–0.10 (<0.012)	6 (2)	<0.01–0.02 (<0.01)	20 (*)

a All analysis data for each specific sample are available in Table S2 in the Supplementary Information.
b Environmental quality standards in the field of water policy (2008) and Water Quality Guidelines for the Protection of Aquatic Life (CCME, 2011).
c AA: annual average; and MAC: maximum allowable concentration for inland surface waters; LT: freshwater, long term exposure.
d NA = not applicable.
e – = No available standard or guideline.
f Junestedt et al. (2003).
g BbF included, BkF excluded.
h Öman and Junestedt (2008).
i * = Not reported.

- Dissolved concentrations, comprising truly dissolved and colloid-bound organic pollutants, were determined on samples passed through 0.7 μm glass-fiber filters, followed by LLE.
- Particle-bound concentrations were calculated through mass balances by subtracting dissolved concentrations from total concentrations.
- Truly dissolved concentrations of PAHs and phthalates were determined using the extract adsorbed onto C_{18} SPE disks (pre-filtered samples) and the colloid-bound concentrations were calculated through mass balances by subtracting truly dissolved concentrations measured on the SPE disks from dissolved concentrations.
- For the phenolic compounds the filtrates passing through the C_{18} SPE disks (pre-filtered sample) were measured to determine colloid-bound concentrations while the truly dissolved concentrations were calculated through mass balances (subtracting colloid-bound concentrations measured on the SPE-filtrates from dissolved concentrations).

The procedure for the separation of the colloid-bound fraction described above has been verified as follows: 100 μg NPs Technical mixture was added to 1 l of tap water, both with and without addition of 100 mg DOC as humic colloids (prepared according to Florence, 1982), to serve as a model-colloid phase. All filtrations, extractions and chemical analyses were

conducted as described below. The C_{18} material used in the SPE disks in this study has already been verified for separation of colloid-bound PAHs and other colloid-bound hydrophobic organic compounds in freshwater (Brown and Peake, 2003).

2.3. Extraction and chemical analysis

For the quality assurance, blank samples were extracted for all the procedures; internal standards were used and for phenolic compounds also duplicate sampling, extractions and chemical analysis were carried out. The dissolved and colloid-bound concentrations were determined for pre-filtered (0.7 μm glass fiber, GF/F Whatman) samples.

2.3.1. PAH and phthalates
For the analysis of total and dissolved concentrations, separatory funnel LLE was performed on 1000 ml samples following the US EPA standardized method 3510C with some modifications. One ml of a surrogate spiking solution, corresponding to 2 μg pyrene-d_{10} (98 Atom % D, Sigma–Aldrich) and 1 μg benzyl benzoate (NEAT, Supelco), was added to the original sample bottle and mixed thoroughly. A mixture of 1:1 cyclohexane and ethylacetate (both HiPerSolv CHROMANORM for HPLC, VWR) was used for extraction. The extracts were concentrated by a rotary evaporator to ~10 ml. For determining the truly dissolved concentrations in pre-filtered

samples, one ml of the surrogate spiking solution, was added to the original sample bottles and mixed thoroughly. The samples were extracted through conditioned C_{18} SPE disks (3M Empore) following the procedure in the US EPA standardized method 3535 with a few modifications, and the 1:1 cyclo-hexane and ethylacetate mixture was used as elution solvent. All extracts (both from LLE and SPE) were dried by filtrations through a column bed of anhydrous sodium sulfate (99+%, ChemPur), and reduced to 1.0 ml by N_2 and analyzed by GC—MS following standardized procedures.

2.3.2.　Phenolic compounds

Total and dissolved, i.e. pre-filtered through 0.7 μm glass fiber (GF/F Whatman), concentrations for phenolic compounds were determined by an LLE procedure and GC—MS stan-dardized procedure at a commercial laboratory. For the analysis of colloid-bound compounds, 1000 ml of pre-filtered (0.7 μm glass fiber, GF/F Whatman) samples were extracted at the Chalmers laboratory using the C_{18} SPE disks and following the same procedure as described above, but no surrogate solution was added in this step. The remaining filtrates were collected and sent to the commercial labora-tory, where the samples were acidified by concentrated H_2SO_4, internal standards added and the samples LLE three times with 10 ml dichloromethane. The extracts were concentrated by rotary evaporation followed by drying through Na_2SO_4, and evaporation to 1.0 ml by N_2. The extracts were analyzed by GC—MS following a standardized procedure.

In several of the samples, higher concentrations were detected in the filtered compared to total samples, and in the colloidal phase compared to the filtered samples. This may be explained by the sensitivity of the analytical method. The less dirty samples, i.e. where particles have been removed, show a lower matrix effect, which leads to higher method sensi-tivity and lower concentrations can be detected. The phenomena may also be explained by inhomogeneous samples as it is difficult to achieve equal distribution of organic compounds when samples are portioned into three subsamples before chemical analysis.

2.4.　Calculations of K_{DOC}

In addition to sorption onto particles in water samples, the organic pollutants can also partition to the colloid-sized parti-cles (<0.1 μm). To get an understanding of how the organic pollutants are distributed in water samples, three-phase partition models can be established (Liu et al., 2005; Eadie et al., 1990, 1992). In these models the organic compounds are distributed among the particulate, colloid and dissolved pha-ses, and the distribution is controlled by partition to the organic carbon content of both particles and colloids. The partition coefficients determined using empirical data can be correlated to the contaminants octanol/water coefficient, K_{OW}, and the contaminants water solubility. The partition coefficient, K_{DOC}, quantifies the ratio of DOC-bound concentrations of pollutants, C_{DOC}, to the free dissolved concentration of the pollutant, C_D, and has in this study been calculated as:

$$K_{DOC} = C_{DOC}/[DOC]C_D; \quad \text{in ml/g DOC}$$

3.　Results and discussion

3.1.　Occurrence and concentrations of organic pollutants in total samples

The results for each specific sample are presented in Supplementary Table S2.

Among the phenols, BPA was detected at the highest total concentrations, followed by 4-t-BP and 4-NP (Table 1). The detection frequency (d.f.) of the phenols ranged between 75 and 100% (BPA < 4-NP = 4-t-PP < 4-t-OP < 4-t-BP). Among the alkylphenol ethoxylates, only OP1EO was detected in a majority of samples (d.f. 75%), whereas the other ethoxylates showed a d.f. of 33% or less. Ethoxylates other than OP1EO were found only in the four stormwater samples (HB, SK, G1, J2). However, NP1EO—NP6EO may be more abundant than measured in this study as the detection limit is 100 ng/l, in contrast to 10 ng/l for other investigated phenols. As a comparison, a majority of the detected concentrations of octylphenolic compounds fall under 100 ng/l.

The DINP showed the highest detected concentration among the phthalates, followed by DEHP and DEP (Table 1). The highest detection frequency was found for DEHP > DBP > BBP > DEP = DINP > DnOP. The DEHP, DINP and DIDP may potentially be present in other samples as they were measured with a detection limit of 1.0 μg/l, in contrast to 0.1 μg/l for the rest of the phthalates.

Naphthalene (NAP) showed the highest detection frequency and concentrations among the PAHs, followed by fluorene (FL), phenanthrene (PHE), fluoranthene (FLU) and acenaphthene (ACE) (Table 2). The low molecular weight (LMW) PAHs (MW ≤ 200) were detected at maximum concentrations one magnitude higher than the heavier PAHs (MW ≥ 228), with the exception of acenaphthylene (ACY), which was found at concentrations closer to the heavier PAHs (Table 2). However, the median concentrations of all analyzed PAHs were very similar and considerably lower than the maximum concentrations. Phenanthrene (PHE) showed the highest median concentration at 0.11 μg/l and seven of the 15 PAHs showed a d.f. of less than 50% (Table 2).

The median alkylphenol and phthalate concentrations measured in this study are similar to concentrations reported for landfill leachates and stormwater in Sweden (Table 1). On the contrary, the Swedish screenings of runoff from waste-sorting surfaces (Table 2) show up to one order of magnitude higher median concentrations for PHE, FLU, PYR, BaA, CHY, BbkF and BaP and much higher detection frequencies for all the high molecular weight (HMW) PAHs (Junestedt et al., 2003). Phenol concentrations reported for leachates from Japan are lower than levels found in this study: 0.05—0.07 μg/l for 4-NP and 0.04—0.31 μg/l for 4-t-OP (Asakura et al., 2004); <0.003—0.014 μg/l (median <0.003) for 4-t-PP and <0.003—64 μg/l (median 0.135 μg/l) for 4-t-BP (Kurata et al., 2008). The detected concentrations of BPA in the current study are lower or in the same magnitude as concentrations reported e.g. by Kurata et al. (2008) from 38 landfills in Japan (0.009—3600 μg/l, median 1.7 μg/l) and Yamamoto et al. (2001) from ten sites in Japan (1.3—17,200 μg/l, median 269 μg/l). Similar to this study, Asakura et al. (2004) detected higher

concentrations of DEHP (9.6—49 µg/l) than other investigated phthalates (BBP 0.7—7.8 µg/l; DBP 3—15 µg/l; DEP 1—8.4 µg/l) in landfill leachates from Japan. In landfill leachates ($n = 7$) from Portugal, Herbert et al. (2006) found NAP, ACE and FL levels that were up to 16, 46 and 14 times higher, respectively, than corresponding concentrations in this study. In contrast, concentrations of PAHs in leachate from operating Finnish landfills ($n = 10$), reported by Marttinen et al. (2003), were generally lower than concentration found in the current study and no PAHs were detected in leachate from closed landfills.

It is believed that less hydrophobic substances tend to elute easily from the landfill material and rapidly move into the leachate compared to more hydrophobic substances (Asakura et al., 2004). However, no correlation between the phenol and phthalate occurrence or concentrations in total samples and the compounds' hydrophobicity (log K_{OW}) was found in this study. Therefore, other factors, such as occurrence of the compounds in the landfilled material and on urban surfaces, transport processes and their degradation patterns are likely to be more important. In contrast, occurrence and concentrations of PAHs followed the substances' water solubility and log K_{OW} reasonably well (d.f. NAP [log $K_{OW} = 3.3$] > FL [log $K_{OW} = 4.2$] > PHE [log $K_{OW} = 4.5$] > FLU [log $K_{OW} = 4.9$] > ACE [log $K_{OW} = 4.0$]).

Bauer et al. (1998) found that the presence of organic macromolecules in leachate may enhance the migration of hydrophobic substances such as many HMW phthalates and PAHs. The moderately hydrophobic 4-t-OP and the more hydrophilic BPA were positively correlated to total organic carbon (TOC) levels, which may be an indication of their adherence to TOC (Spearman's $\rho = 0.69$ and 0.63, respectively, significant at the 0.05 level). Similarly, Urase and Miyashita (2003) also observed a slight positive correlation between BPA and TOC leachate concentrations. No correlation to TOC was found for other compounds; also monitoring of stormwater quality in Sweden showed no correlation between phthalates and alkylphenols and TOC or DOC (Björklund et al., 2009). These results suggest that TOC cannot be used as predictors of levels of phthalate, PAH and most phenols in landfill leachates and stormwater.

According to a fate modelling performed by Sato et al. (2011), the fate of PAHs in landfills is dominated by the presence of organic carbon. The HMW PAHs attach strongly to organic carbon and will therefore have a long residence time in the landfill. Our results confirm this hypothesis as both the detection frequency and the concentrations of the HMW PAHs was much lower than those for the LMW PAHs, suggesting that the former are adsorbed to the organic material in the landfills. Volatile PAHs, such as NAP, may evaporate from the landfill material and, because of lower hydrophobicity, are more likely to be eluted with leachate. This assumption was confirmed in this study, as NAP was the only PAH that was detected in all the samples.

The highest phenol concentrations were generally found in TA2, leachate from landfilled sewage sludge, and in BR1, the old landfill with mixed waste. Both sites were in operation 1940—1980. Abundance of phenols in both solid waste and wastewater sludge from this period illustrates their widespread use before legal restrictions in the 1990s (e.g. Regulations 793/93, 689/2008 and Directive 2003/53/EG). The source of

phenolic compounds in BR1 and TA2 is most likely consumer products. This assumption is confirmed by the fact that leachate from an industrial landfill (SR) operated during the same period contains considerably less phenols. Concentrations of phenols in leachate from the ash landfill (TA1) were low as the organic content of the waste has been combusted and only smaller amounts of sludge are used to stabilize the ash. In FL, the newer industrial landfill, phenol concentrations were below or close to the detection limit, therefore no partitioning analysis of this sample was performed. The observed detection frequencies and concentrations may also be a result of phenol degradation patterns. The anaerobic conditions of covered landfills are assumed to slow down or prevent the degradation of BPA (Yuan et al., 2002; ECB, 2010), which may explain the high BPA concentrations in BR1 and TA2, as both landfills were covered in the 1980s. Biodegradation may explain the low occurrence of especially longer ethoxylates (NP4—6EOs) in landfill leachates in the current study, as was seen in the study of Baun et al. (2004). In contrast, stormwater from the waste-sorting sites (HB and SK) is still exposed to materials possibly containing alkylphenol ethoxylates. Urban stormwater is constantly exposed to emissions from vehicles and urban surfaces containing alkylphenol ethoxylates such as concrete and plastic materials, lubricants, tyre rubber and car care products. Hence, runoff samples are believed to contain higher concentrations of alkylphenol ethoxylates than leachate samples, which was also the case in this study.

In urban runoff, observed concentrations of DINP (70 and 21 µg/l) were one magnitude higher than in the leachate samples (1.5 and 2.1 µg/L). The opposite relationship was true for DEHP, for which concentrations up to 23 µg/l were found in the leachate from an old landfill (BR1), whereas measured concentrations in urban stormwater were 2—3 µg/l. These results validate the hypothesis that stormwater quality reflects the recent use of chemicals whereas landfill leachate reflects the historical use. Historically, DEHP is one of the most extensively used phthalates worldwide. However, following restrictions on the marketing and use of DEHP (e.g. Directives 2003/36/EC, 2004/93/EC and 2005/84/EC), the higher molecular weight phthalates, including for example DINP and DIDP, has increasingly taken over the market. Urban runoff has been shown to be an important sink for phthalates, which are emitted from PVC applications on vehicles, roofing and cladding materials, and sealants (Björklund, 2010). At BR landfill, all types of waste have been deposited and it is likely that PVC is an important phthalate source at this site, as the major use of phthalates is as PVC plasticizers (ECPI, 2010).

The high DEHP levels detected at BR1 suggests that after more than 30 years of possible degradation, DEHP is still emitted at high levels from this site. Similarly, PAHs are still emitted at high levels from this landfill, confirming their slow degradation process. PAH levels in other samples were considerably lower.

Combustion of organic materials, including e.g. coal, petroleum products, wood and waste products, is the largest contributor to PAHs in the environment (Mostert et al., 2010). In the urban environment, vehicle exhausts, rubber tires and asphalt materials are probable emission sources (Norin and Strömvall, 2004). The PAHs may reach the landfill through atmospheric deposition and via disposed material, such as
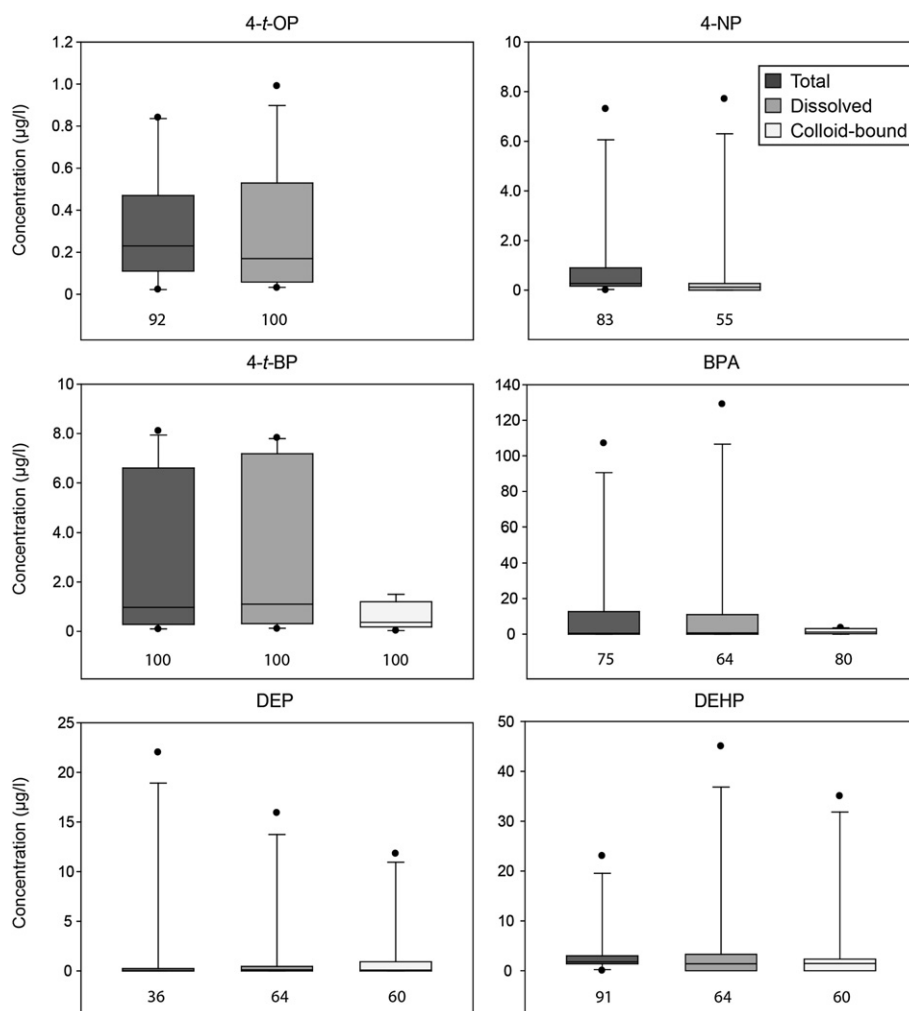
Fig. 1 – The minimum and maximum, the lower, median and upper quartile, and possible outliers of total, dissolved and colloid-bound concentrations of 4-NP, 4-t-OP, 4-t-BP, BPA, DEP and DEHP in landfill leachate and stormwater samples. The number below the box denotes the detection frequency, expressed as a percentage.

tires, asphalt, used oils and creosote treated wood. Naphthalene, the most abundant PAH in the current study, is a significant component of creosote tar and reclaimed asphalt material. The PAHs are found as mixtures of the different molecules, it is therefore difficult to identify specific sources of an identified PAH pattern (Mostert et al., 2010). Though, as a general rule, LMW PAHs are a result of low temperature processes such as wood burning, whereas high temperature processes such as vehicle emissions give rise to HMW PAHs. This rule, however, does not explain why high concentrations of NAP are found in the leachate samples. The fact that NAP is the most water soluble PAH is probably the governing factor for the observed results.

3.2. Comparison with water quality guidelines

As seen in Table 1, detected levels in the wastewater samples repeatedly exceeded the European environmental quality standards for DEHP ($n = 9$ exceeding AA), octylphenol ($n = 9$) and nonylphenol ($n = 5$) in surface waters, and the Canadian freshwater quality guideline for NP and its ethoxylates ($n = 4$).

The European water quality standards for BbkF and BPY + INP were exceeded in all samples where the substances were detected ($n = 6$ and 4, respectively, see Table 2). Similarly, the Canadian water quality guidelines were repeatedly exceeded for several of the PAHs (Table 2). The infringements of the quality standards imply that negative effects on aquatic life from these anthropogenic stressors cannot be excluded. The studied landfill leachates are conveyed to the sewage treatment plant, and the urban runoff is treated in sedimentation facilities prior to discharge into receiving waters. However, the efficiency of the sedimentation facilities for the removal of studied pollutants is unknown and should be investigated. Treatment by filtering is currently being installed at the waste-sorting sites by the recommendation of the authors.

3.3. Distribution between the total, dissolved and colloid-bound phases

For method verification purposes, the distribution of phenols between the total, dissolved and colloid-bound phases has
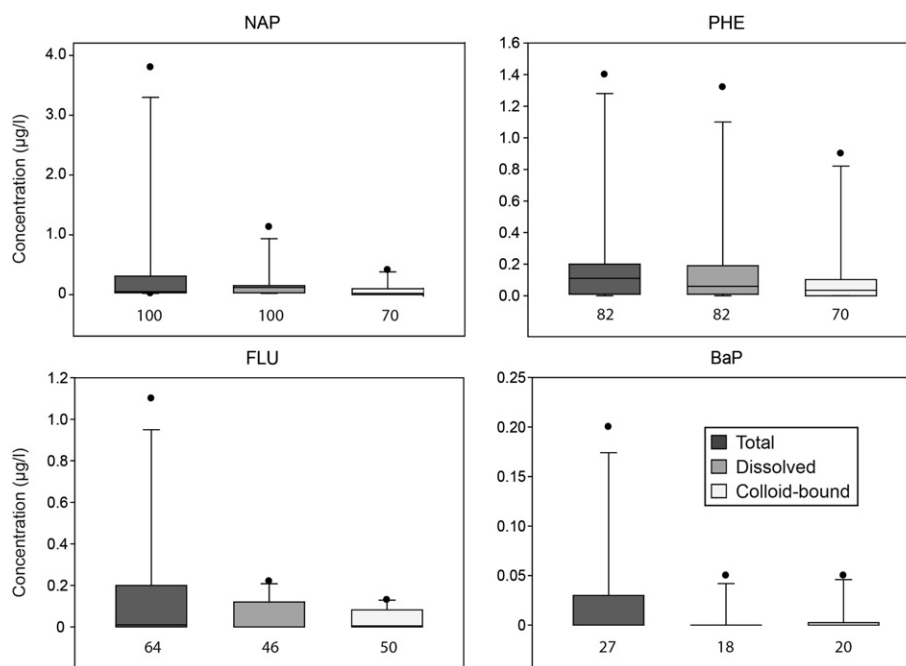
Fig. 2 − The minimum and maximum, the lower, median and upper quartile, and possible outliers of total, dissolved and colloid-bound concentrations of NAP, PHE, FLU and BaP in landfill leachate and stormwater samples. The number below the box denotes the detection frequency, expressed as a percentage.

been studied in spiked samples of tap water with and without addition of humic colloids (DOC). In the total and filtered samples without DOC addition, 44 and 17 µg L^{-1} of 4-NP was detected, while no 4-NP was detected in the SPE-treated sample. In the total and filtered samples with DOC addition, 77 and 32 µg L^{-1} of 4-NP was detected and 24 µg L^{-1} 4-NP was detected in the SPE-treated sample. Results for the SPE-treated samples confirmed that SPE disks remove truly dissolved compounds while the colloid-bound compounds are transported through the disks. The fact that a concentration twice as high was detected in the total sample with addition of DOC as compared to the samples without DOC addition indicates that 4-NP becomes more water-born in the samples with DOC, as 4-NP adheres to DOC instead of the glass equipment surfaces. As the technical NPs standard was used, not all the added NPs isomers were measured. However, 4-t-BP and 4-t-PP were also detected and results were similar to those of 4-NP (not presented).

3.3.1. Phenols

The concentrations of phenols in the dissolved and colloid-bound fractions generally follow the order found in total samples: 4-t-BP > BPA > 4-t-PP > 4-t-OP > 4-NP (Fig. 1). Occurrence in the dissolved and the colloidal phases generally decreased with increasing log K_{OW}, in contrast to total samples, where no correlation was found to log K_{OW}. The least hydrophobic phenols, 4-t-BP and BPA, showed high detection frequencies in both the dissolved (100 and 64%, respectively) and the colloidal phase (100 and 80%, respectively). Concentrations of the moderately soluble 4-t-PP and 4-t-BP were mainly unchanged after filtration (see Supplementary Table S2), whereas the colloidal concentrations were lower

than total and dissolved concentrations (Wilcoxon signed rank test, statistically significant at the 0.05 level). Therefore, most of the 4-t-PP and 4-t-BP is likely to be present as truly dissolved while a smaller fraction is colloid-bound. However, the difference in medians was not statistically significant for BPA between any of the fractions, which indicates that BPA is predominantly adsorbed to colloids (as suggested, DOC). Urase and Miyashita (2003) also found that a major fraction of BPA in landfill leachates remained in the filtered samples.
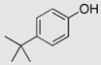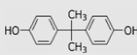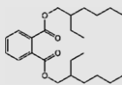
The alkylphenol ethoxylates were infrequently detected in the dissolved and colloidal phase which suggests them being mostly bound to particles >0.7 µm; no Wilcoxon signed rank test was performed for these compounds. The least water soluble of the investigated phenols, 4-t-OP and 4-NP, showed only a slight decrease in median concentrations (not statistically significant) after filtration, but were not detected in the colloidal phase. Due to their hydrophobic nature, it is unlikely that 4-t-OP and 4-NP are truly dissolved and therefore adsorbed to the SPE-disk material. Instead, they may be adsorbed to the particles which are smaller than 0.7 µm but larger than the SPE-disk pores (0.006 µm) and are therefore sterically hindered and retained by the disk.

3.3.2. Phthalates

The detection frequencies of phthalates in the dissolved fraction were generally lower than in the total samples, but at similar levels as in the colloid-bound fraction (Fig. 1). The median concentrations of DEP, DnBP, BBP and DEHP decrease only slightly after filtration and SPE-treatment. Statistically significant differences (Wilcoxon signed rank test) between the medians in different fractions were not found for any of the phthalates (not shown).

Table 3 — Partition coefficients to DOC colloids, log K_{DOC} and K_{DOC}, compared with the octanol/water partition coefficients, log K_{OW}, and water solubility for selected phenolic, phthalic and polycyclic aromatic pollutants in landfill leachates and stormwater samples.

Chemical name/CAS no	Formula	MW g/mol	Water solubility[a] mg/l	Vapour pressure[a] pa	Log K_{OW}[a]	Log K_{DOC}[b] ml/g	K_{DOC}[b] ml/g	K_{DOC} literature[c] ml/g
Phenols								
4-Tert-butylphenol (4-t-BP) 98-54-4		150	610	5.0×10^{-1}	3.3	3.7	4.7×10^3	—[d]
Bisphenol A (BPA) 80-05-7		228	300	5.3×10^{-6}	3.4	4.5	32×10^3	8.9×10^3
Phthalates								
Diethyl phthalate (DEP) 84-66-2		222	590	6.5×10^{-2}	2.5	4.6	31×10^3	—
Di(2-ethylhexyl) phthalate (DEHP)117-81-7		391	2.5×10^{-3}	2.5×10^{-5}	7.7	5.9	740×10^3	—
Polycyclic aromatic hydrocarbons								
Naphthalene (NAP) 91-20-3		128	32	12	3.3	4.3	22×10^3	2.6×10^3—19×10^3
Phenanthrene (PHE) 85-01-8		178	1.2	9.1×10^{-2}	4.5	4.9	87×10^3	19×10^3—20×10^3
Fluoranthene (FLU) 76774-50-0		202	2.0×10^{-1}	6.7×10^{-4}	4.9	5.0	100×10^3	94×10^3—99×10^3

a ECB (2008, 2010), Cousins et al. (2003) and Neilson (1998).
b Calculated from the analysis data in this study, for original data see supporting material.
c Liu et al. (2005), Brown and Peake (2003) and Lee and Kuo (1999).
d No data available.

The HMW phthalates are highly hydrophobic and are expected to adsorb strongly to particles, hence can be removed by filtration and sedimentation. However, despite its hydrophobic character (log K_{OW} = 7.7), DEHP was detected in the dissolved and the SPE-treated samples as frequently as was DEP (Fig. 1). The same trend was seen for BBP (log K_{OW} = 4.7, not shown). This indicates that the partition coefficient to the particulate organic content K_{OC} in the water samples of DEHP and BBP are decreased by components of the sample matrix, for example DOC as suggested by Lee and Kuo (1999). Similarly, Zheng et al. (2007) showed that most of DBP and DEHP in landfill leachate were soluble (0.7 μm) and highly associated with DOC.

3.3.3. Polycyclic aromatic hydrocarbons

The detection frequencies of PAHs in the filtered samples were similar to the total samples, slightly lower in the colloid-bound fraction and correlated well with the PAH water solubility; the highest detection frequencies were observed for the most soluble PAHs (Fig. 2). The heaviest PAHs — BaP, DBA, BPY and IND — were detected in less than 27% of the filtered samples and in the colloid-bound phase. Contrary to what can be expected from the high log K_{OW} values, no statistically significant difference (Wilcoxon signed rank test) in concentrations between phases was found for the HMW BaP (log K_{OW} 6.1). This suggests that the partition to the organic content in particles (K_{OC}) is decreased by the presence of DOC.

For the most water soluble PAH, NAP, total and dissolved concentrations were not significantly different, while the colloid-bound concentrations were lower (statistically significant at the 0.05 level) than the total concentrations. This suggests a presence of a significant fraction of truly dissolved NAP. For PHE, both the dissolved and the colloid-bound concentrations were lower (statistically significant at the 0.05 level) than the total concentrations, which indicates it
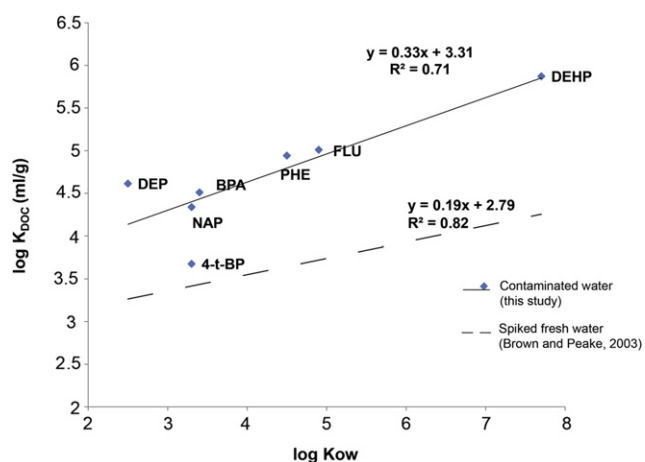
Fig. 3 – Linear regression analysis of log K_{DOC} (ml/g) and log K_{OW} for selected phenolic, polycyclic aromatic and phthalic pollutants in contaminated water (landfill leachates and stormwater), compared with data from determination of PAHs sorption on colloids in spiked fresh water.

being predominantly bound to particles. No Wilcoxon signed rank test was performed for the remaining PAHs due to low detection frequencies.

3.3.4. Importance of particles, DOC and K_{OW} for the partitioning of the organic pollutants

The hypothesis i) that the PAHs, 4-NP, 4-t-OP and the HMW phthalates are adsorbed to particles, was in part confirmed for selected samples and only for PHE for all the samples. Particulate concentrations of HMW PAHs, DEHP and DnOP were higher than dissolved concentrations for BR1 and BR2. In stormwater samples, 4-NP and nonylphenol ethoxylates' particulate concentrations were higher than dissolved concentrations. The abundance of organic pollutants, including those of high hydrophobicity, in the dissolved and colloid-bound fractions has confirmed the hypothesis of increased solubility of organic compounds in presence of colloids, such as DOC. Indeed, the dissolved and colloid-bound fractions were shown to be equally or more important than the particulate fraction for organic pollutants partitioning in urban waters. In such a way, the mobility of organic pollutants is increased, as illustrated by the example of the samples BR1, BR2 and BR3 taken at different stages of leachate treatment (oil separator and sedimentation pond). The fact that concentrations of organic pollutants do not decrease significantly between the treatment steps implies that sedimentation facilities currently being applied for treatment of stormwater and landfill leachates are not effective in removing organic pollutants, hence different techniques have to be developed.

As no correlation was found for occurrence and concentration of alkylphenols and phthalates with K_{OW} in total samples, the composition of landfilled waste and urban sources are likely to be the governing factors. Bisphenol A showed moderate correlation with the TOC concentration. In

addition, degradation patterns could explain occurrence of several of alkylphenols and phthalates. In contrast, occurrence and concentrations of PAH in total samples followed their water solubility reasonably well. Generally for most of compounds, a moderate correlation of −0.60 was found for the filtered and colloid-bound phases (Spearman rank test, significant at the 0.01 level, 2-tailed test). This potentially enables prediction of pollutants transport based on their physico-chemical properties.

3.3.5. Correlations of K_{DOC} and K_{OW}

Binding of organic pollutants in sediments is primarily controlled by physico-chemical properties like K_{OW} and the organic carbon content of sediments (Ditoro et al., 1991; Shea, 1988). The hydrophobic partition is the dominant binding mechanism to organic carbon in sediments; it is therefore of interest to evaluate if the same relation is valid for organic contaminants binding to the organic carbon in colloids. The determined log K_{DOC} for selected phenols, phthalates and PAHs, calculated from the analysis data of landfill leachates and stormwater, are presented in Table 3 and plotted against log K_{OW} in Fig. 3.

The results (Fig. 3) show a linear relationship between log K_{DOC} and log K_{OW} for the selected PAHs, phenolic and phthalic compounds. Hence, the suggested binding process of these contaminants to the DOC is partitioning. The compounds with the lowest water solubility, e.g. 4-t-BP and DEP, showed the lowest correlation (Table 3). Depending on the higher polarity these compounds may interact with colloids through other mechanisms than hydrophobic partition or sorption to organic macromolecules. The results confirm that the K_{OW} values for organic pollutants could be used to describe the organic pollutants binding process to DOC colloids. In Fig. 3, the results from this study are compared with results from a study of specific PAHs binding to humic acid colloids in spiked freshwater samples (Brown and Peake, 2003); the organic pollutants are to a higher degree sorbed to the DOC colloids in the landfill leachates and stormwater. This may be explained by complex matrix effects of other pollutants' occurrence in contaminated water as stormwater and landfill leachates, but also of effects from the much higher variation in DOC concentrations (2–230 mg/l) used in our study. The DOC in the landfill leachates and stormwater may also have a different chemical composition, e.g. more asphalt- and petroleum-related compounds, than the humic acid added to the freshwater samples.

4. Conclusions

Alkylphenols, Bisphenol A, phthalates and PAHs were detected in all the untreated and most of the filtered samples. 4-tert-Pentylphenol was only detected in leachate samples whereas DINP was found only in urban stormwater. Concentrations of phenols and PAHs in urban stormwater were one order of magnitude lower than in landfill leachates and stormwater from waste-sorting sites. The phthalate concentrations were found to be in the same range for all types of waters apart from DINP, for which urban stormwater levels were one order of magnitude higher than the rest of the samples. The median

concentrations for most of the pollutants, 4-t-OP and OP1EO being the only exceptions, were in the same range as earlier reported concentrations for sewage in Sweden. The European environmental quality standards for surface water were repeatedly exceeded for octylphenol, nonylphenol, DEHP, BbkF and BPY + IND. However, all the studied waters are treated either locally or at the sewage treatment plant before discharge into receiving waters.

A method for separation of colloid-bound organic pollutants has been developed, verified and applied to the samples. Most of the studied compounds were frequently detected in the dissolved (<0.7 μm) and the colloid-bound phase, their occurrence generally decreasing with increasing log K_{OW}. A Wilcoxon signed rank test was performed to evaluate the hypothesis that there is a difference in medians between total, dissolved and colloid-bound concentrations. For most tested compounds, the total concentration is higher than the concentration in the dissolved samples and similarly dissolved > colloidal. However, the difference in concentrations between total and dissolved phase is not significant at the 0.05 level for any phenols or phthalates, but for three of the tested PAHs; naphthalene (mostly dissolved), phenanthrene and fluoranthene (mostly particle bound). These results indicate that organic compounds are predominantly truly dissolved and/or attached to particles <0.7 μm in landfill leachates and stormwater. It is suggested that DOC decreases the apparent K_{OW} of the compounds. The linear relationship between log K_{DOC} and log K_{OW} for the selected phenolic, PAHs and phthalic compounds confirmed that the binding process of these contaminants to the DOC is partitioning.

Acknowledgements

Appendix A. Supplementary material

Supplementary material associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.watres.2012.11.054.

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