Screening of Organic Contaminants in Urban Snow

Karin Björklund*, Ann-Margret Strömvall* and Per-Arne Malmqvist*

* Department of Civil and Environmental Engineering, Chalmers University of Technology, Sven Hultins Gata 8, 412 96 Göteborg, Sweden
(E-mail: *karin.bjorklund@chalmers.se; ann-margret.stromvall@chalmers.se; per-arne.malmqvist@chalmers.se*)

Abstract
Snowmelt is known to cause peak concentrations of pollutants which may adversely affect receiving water quality. High concentrations in snow have been shown for e.g. metals and suspended solids, whereas studies on organic pollutants are rarely reported. This study aims at investigating the occurrence of anthropogenic organic compounds in urban snow, and at identifying sources of the pollutants. Snow from sites in Gothenburg, Sweden, was sampled and a range of organic substances was analysed. The most frequently detected organic pollutants in urban snow were polycyclic aromatic hydrocarbons, high molecular-weight phthalates, 4-nonylphenol and 4-octylphenol. Brominated flame retardants and chlorinated paraffins were only sporadically detected. In several snow samples, the concentrations of specific PAHs, alkylphenols and phthalates were higher than reported stormwater concentrations and European water quality standards. Pollutant source identification and sustainable management of snow are important instruments for the mitigation of organic contaminants in the urban environment.

Keywords
Alkylphenols; PAHs; perfluorinated compounds; phthalates; sources; urban runoff

INTRODUCTION

In the beginning of 2010, southern Sweden experienced unusually cold weather and a winter with extraordinary amounts of snow. To manage these masses, many municipalities were forced to dump urban snow into watercourses and coastal waters. This led to a debate on the contamination of snow, and it was concluded that the occurrence of hazardous substances in snow is, to a large extent, unexplored.

Urban snow accumulates pollutants emitted from human activities and acts as a temporary storage of contaminants during sometimes month-long periods. Snowmelt generally occurs during a short time period, which often results in concentration peaks of contaminants in surface runoff and in receiving waters (Meyer and Wania 2008). The transport of pollutants from the snowpack shows distinct peak loads where the dissolved pollutants are released with the initial meltwater fractions and the particulate pollutants at the end of the snowmelt. Snowmelt studies show that levels of metals and suspended solids may be several orders of magnitude higher than pollutant loads in rain-induced runoff (Westerlund and Viklander 2006). Organic contaminants in urban snow have, however, not been studied to the same extent. The occurrence of e.g. perfluorinated compounds (Liu *et al.* 2009), and polycyclic aromatic hydrocarbons (PAHs) (Reinosdotter *et al.* 2006) in urban snow has occasionally been reported. Screening studies of snow, where several groups of organic contaminants are investigated, are even more infrequent. Screenings of organic substances have earlier been performed on e.g. river water (Dsikowitzky *et al.* 2004) and landfill leachate (Paxéus 2000). These studies illustrate the broad spectrum of organic contaminants emitted from urban areas.

This study aims at investigating the occurrence of anthropogenic organic compounds in urban snow in Gothenburg (Sweden). Initially, non-target screenings of semi-volatile organic compounds (SVOCs) were performed, and thereafter target analyses of specific compound groups. These target compounds were selected according to their potential to reach the urban stormwater system and their potential effects in the aquatic environment. The study also aims at identifying sources of the detected pollutants in urban runoff.

MATERIALS AND METHODS

Sampling

Sampling of snow was conducted during both 2009 and 2010. Snow sampled in February 2009 had fallen during one day, five days prior to sampling, and was used for the non-target screenings and for analyses of phthalates and alkylphenols. Samples were collected from three sites in Gothenburg: Kärra – suburban area, estimated annual average daily traffic (AADT) 500 vehicles; Gårda – motorway area, AADT 90 000; and Järnbrott – access road to residential and industrial area, AADT 59 000. The samples were collected from snow banks in direct proximity of the road. In 2010, Gothenburg experienced several snowfall occasions (Figure 1) and the sampled snow was used for target analyses of organic contaminants. Samples were collected from Kärra, Gårda and Järnbrott, and at the end of the snowmelt period, samples were also collected at three centrally located snow deposits (Gårdamotet, Heden and Vallhamra). An urban background sample was collected in the Delsjön forest area, circa two kilometres from the city centre. The snow was collected in solvent-rinsed glass containers with Teflon-lined caps or in stainless steel containers. The snow was thawed in room temperature and samples analysed by the accredited laboratories were sent in coolers as soon as possible.

Non-target screening

The thawed samples (~ 1.5 l) were spiked with internal standard (phenanthrene-d_{10} and 2-fluorobiphenyl) and extracted by solid phase extraction using C_{18} discs (3M Empore). The contaminants were eluted with isopropanol and *n*-heptane. The extracts were dried with Na_2SO_4 and evaporated before analysis on an ion-trap GC-MS (CP-3200 and Saturn 2200, Varian), equipped with a fused silica (VF-5ms) capillary column (30 m × 0.25 mm × 0.25 µm). Chromatographic conditions were: 1 µl split/splitless injection (290°C), initial oven temperature 50°C, 5 min hold, programmed to 290°C at a rate of 5°C/min. Helium was used as carrier gas (1 ml/min), and the transfer line and trap temperatures were set to 250°C and 220°C, respectively. The mass spectrometer performed a full scan from m/z 40 to 450. Identification was based on a comparison of chromatographic retention times and mass spectra of analytes with those of reference compounds and spectral databases (NIST/EPA/NIH Mass Spectral Library 2005). For quantification of analytes, three-point calibration curves were used. Procedural blanks were treated identically with the snow samples, from extraction to analysis.

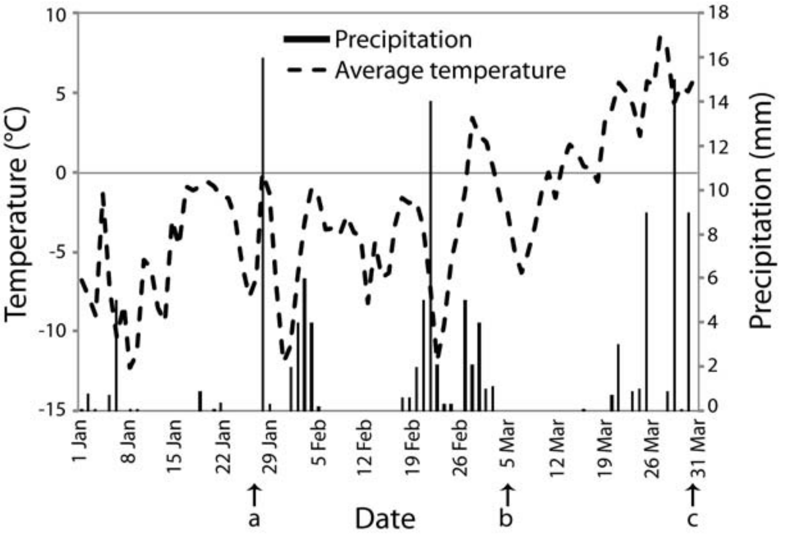

Figure 1. Weather data for Gothenburg during the winter 2010. The sampling occasions are marked with arrows: (a) road side snow in Järnbrott; (b) road side snow in Järnbrott, Gårda, Kärra; and (c) snow deposits in Gårdamotet, Heden, Vallhamra and background snow in Delsjön.

Target analysis of organic contaminants

Analyses of alkylphenols, brominated flame retardants, chlorinated paraffins, perfluorinated compounds, phthalates and PAHs were performed by accredited commercial laboratories. The methods for extraction, clean-up and analysis are summarized in Table 1.

RESULTS AND DISCUSSION

Non-target screening

The original aim of the non-target screening was to identify and quantify the most abundant pollutants in each snow sample. The identification was, however, obstructed by the large number of peaks in the total ion chromatograms, implying that the samples contain a multitude of different compounds, presumably of anthropogenic origin. All samples also showed an unresolved complex mixture (UCM), often used as an indication of petroleum-derived contamination, which prevented identification and quantification of peaks in the latter part of the chromatogram. We could, however, clearly identify specific phthalates (DiBP, DnBP, BBP and DEHP [full names of analysed substances are found in Table 1]), benzothiazole, several phenols (e.g. *tert*-butylphenol, butylated hydroxytoluene [BHT] and 2,4-di-*tert*-butylphenol) and PAHs in the snow samples. In addition, concentrations of SVOCs and total petroleum hydrocarbons (TPHs) were determined. The highest concentrations (6.0 mg TPHs/l and 8.0 mg SVOCs/l) were detected in the motorway area (Gårda), and the lowest concentrations (1.6 mg/l for both TPHs and SVOCs) were found in the residential area (Kärra).

Table 1. Chemical names, analytical methods and detection limits for the target compounds.

Substance group	Included compounds	Extraction, clean-up, analysis	Reported detection limit
Alkylphenols * (APs)	4-*tert*-octylphenol (4-OP); 4-nonylphenol (4-NP); 4-*t*-octylphenol and 4-nonylphenol mono-, di-, tri-, tetra-, penta-, and hexa-ethoxylates (OP1-6EO and NP1-6EO, respectively)	Liquid liquid (LL) extracted with *tert*-butyl methyl ether, cleaned with silica gel and derivatized using N-Methyl-N-(trimethyl-silyl) trifluoroacetamide. GC-MS	10 ng/l for OPs; 500 ng/l for NPs. Up to 0.8 µg/l for OPs and 2 µg/l for NPs in samples with large matrix effects
Brominated flame retardants* (BFRs)	tetrabrominated diphenyl ether (tetraBDE); pentaBDE; hexaBDE; heptaBDE; octaBDE; nonaBDE; decaBDE; tetrabromobisphenol-A (TBBA); decabromobiphenyl (DeBB); hexabromocyclododecane (HBCD)	Extracted with toluene, followed by several clean-up steps. GC-MS	0.0003–0.10 µg/l
Chlorinated paraffins (CPs)	C_{10}-C_{13} short-chain chlorinated paraffins (SCCP); C_{14}-C_{17} medium-chain chlorinated paraffins (MCCP)	Extracted with toluene. GC-NCI/MS	0.20–0.50 µg/l
Perfluorinated compounds* (PFCs)	perfluorooctane sulfonate (PFOS); perfluorooctanesulfonamide (PFOSA); perfluorooctanoic acid (PFOA)	Solid phase extraction, eluted with methanol. LC-MS/MS	1.5–2.0 ng/l
Phthalates*	dimethyl (DMP); diethyl (DEP); di-*n*-butyl (DnBP); di-*iso*-butyl (DiBP); di-*n*-octyl (DnOP); di-(2-ethylhexyl) (DEHP); butylbenzyl (BBP); di-*iso*-decyl (DIDP); di-*iso*-nonyl phthalate (DINP)	LL extracted with *n*-hexane. GC-MS	0.1–2.0 µg/l
Polycyclic aromatic hydrocarbons * (PAHs)	naphthalene (NAP); acenaphthylene (ACY); acenaphtene (ACE); fluorene (FL); phenanthrene (PHE); anthracene (ANT); fluorantene (FLR); pyrene (PYR); benzo[*a*]anthracene (BaA); chrysene (CHY); benzo[*b*]fluoranthene (BbF); benzo[*k*]fluoranthene (BkF); benzo[*a*]pyrene (BaP); dibenzo[*a*,*h*]anthracene (DBA); benzo[*ghi*]perylene (BPY); indene[1,2,3-*cd*]pyrene (INP)	Extracted with *n*-hexane. GC-MS	0.01 and 0.1 µg/l

* Analysis accredited by Swedac (The Swedish Board for Accreditation and Conformity Assessment). Measurement errors have been reported (no included in table).

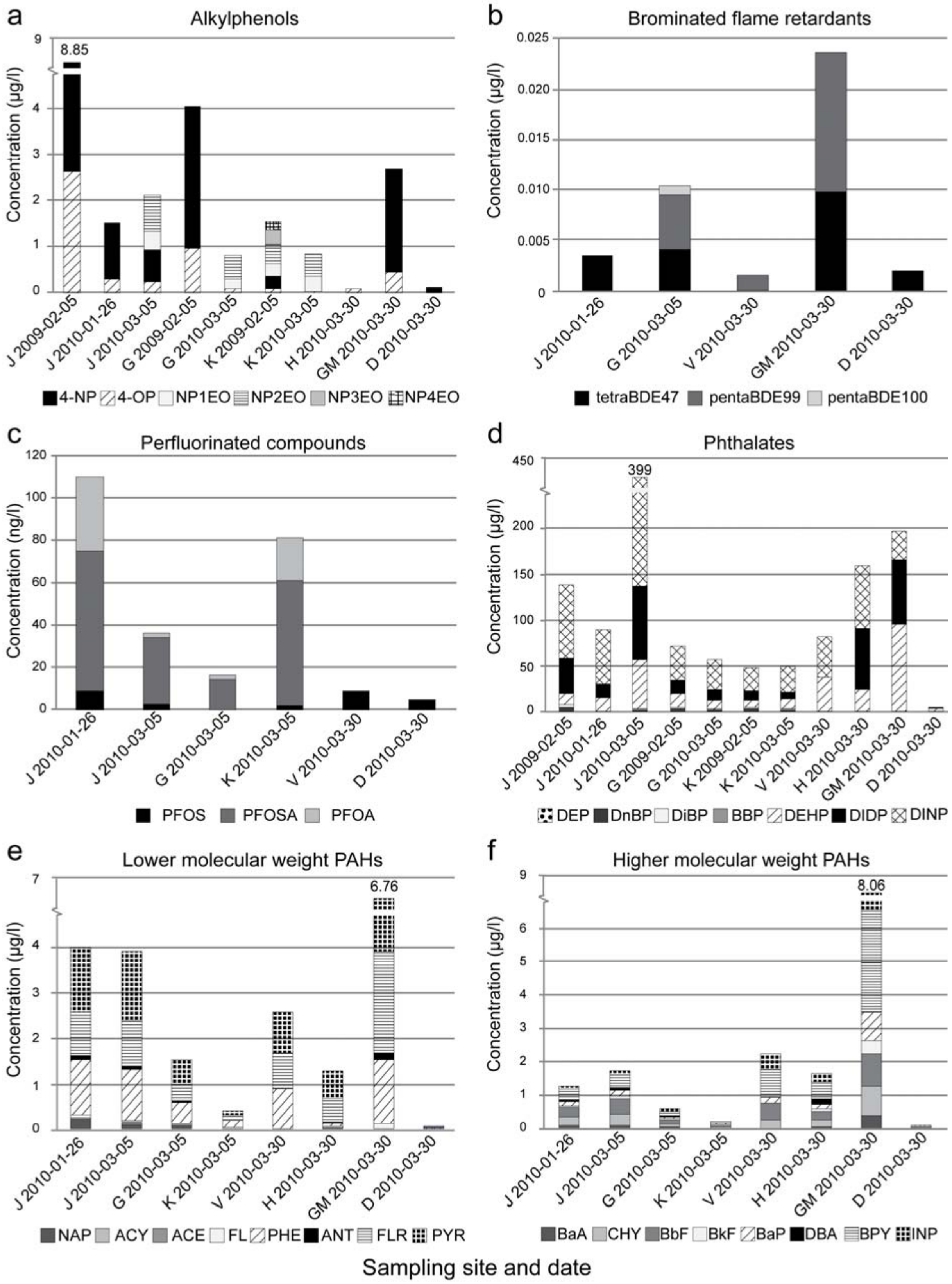

Figure 2. Detected concentrations of (a) alkylphenols; (b) brominated flame retardants; (c) perfluorinated compounds; (d) phthalates; (e) lower molecular-weight PAHs; and (f) higher molecular-weight PAHs. Sampling occasions where concentrations of all APs, PFCs and BFRs are below the detection limits are not shown in the graphs. Sampling sites: J – Järnbrott; G – Gårda; K – Kärra; V – Vallhamra; H – Heden; GM – Gårdamotet; D – Delsjön.

Alkylphenols

Alkylphenols were analysed in snow sampled in both 2009 and 2010. The highest maximum and median AP concentrations (Figure 2a) were observed for 4-NP (6.2 and 0.27 µg/l, respectively), whereas 4-OP showed the highest detection frequency (82%). Nonylphenols are used in larger amounts than octylphenols, and the trend with higher NP concentrations have been observed in environmental matrices world-wide (Ying *et al.* 2002). The octylphenol ethoxylates were below the d.l. in all samples, whereas NP1EO and NP2EO were detected in four of the samples, and NP3EO and NP4EO in one sample (Figure 2a). For the snow deposit samples (Heden, Vallhamra and Gårdamotet), the detection limits (d.l.) were elevated up to 2.0 µg/l for 4-NP and 12 µg/l for NP4EO due to large matrix effects, and all APs were below the d.l. in the Vallhamra sample. The varying detection limits render data analysis difficult and it has not been possible to identify clear trends. There is only weak concentration correlation between samples collected on the same day, and between samples from the same site. It can be concluded, though, that the 4-NP and 4-OP concentrations found in snow exceeded most reported stormwater values from Sweden (maximum 1.2 and 0.35 µg/l, respectively; *n* = 13 [Björklund *et al.* 2009]). In addition, the European environmental quality standard for surface water (EQS [Directive 2008/105/EC of the European Parliament and of the Council]) for 4-NP – maximum allowable concentration (MAC) 2.0 µg/l – was exceeded in three of the samples and the EQS for 4-OP – annual average (AA [considered protective against short-term pollution peaks]) 0.1 µg/l – was exceeded in five of the snow samples.

Nonylphenol is the precursor in the production of ethoxylates, and also a major degradation product of NPEOs, which are used in industrial applications of surfactants. Abiotic degradation of NPEOs is considered to be negligible in comparison to biodegradation (Langford *et al.* 2005), which is likely to be inhibited by the prevailing temperatures at the time of snow sampling. The high concentrations of 4-NP in snow, compared to NPEOs, and the expected low degradation of NPEOs into NP, suggest that 4-NP may be emitted directly to the urban environment. In a substance flow analysis vehicles, where nonylphenolic compounds are used in lubricants, fuel and car care products, air-entraining additives in concrete and tyre wear, were identified as the most important NP and NPEO sources in stormwater (Björklund in press). The ethoxylates may also be used in de-icing products and have been suggested as additives in diesel, but reports on the use of 4-NP in such products have not been found. In conclusion, we have not found an explanation of the relatively high 4-NP concentrations found in some of the snow samples. Octylphenol sources are of different character than nonylphenols sources since only 2% of the total use of OP is used for producing ethoxylates and the remaining part is used in the manufacturing of resins (DEFRA 2008). Circa 80% of the resins are used in the production of tyres, which suggests that the octylphenol in urban snow mainly originates from tyre rubber. Other applications of octylphenolic compounds include paints, polymers and cleaning products.

Brominated flame retardants

The most used BFR in Sweden is currently TBBP-A, followed by decaBDE and HBCD (KemI 2010). The use of decaBDE is, unlike penta- and octaBDE, not regulated in Sweden. It is therefore somewhat surprising that the BFRs detected in urban snow (Figure 2b) were pentaBDE-99 (*n* = 3; 0.0015–0.014 µg/l), pentaBDE-100 (*n* = 1; 0.001 µg/l), and tetraBDE-47 (*n* = 4; 0.0019–0.0097 µg/l), the latter being the most occurring congener in pentaBDE. The commercial mixtures of pentaBDE include tetra-, penta- and hexa congeners; octaBDE includes hexa-, hepta-, octa- and nonaBDE; whereas decaBDE only includes small amounts of nonaBDE (de Wit 2002). A phase-out of the production and use of pentaBDE in Sweden started in 1999 (KemI 2003). Since 2004 the use is restricted within the European Union (Directive 2003/11/EC) and pentaBDE is currently found only in imported goods. The main application of pentaBDE is in polyurethane foam found in e.g. furniture and car headrests and ceilings, and to minor extent in textiles, building materials and packaging. Historically, tetraBDE-47 has been the most occurring congener in environmental matrices from areas generally affected by pollution (de Wit 2002). Studies from the past decade indicate, however, that decaBDE-209 is currently the most occurring congener in the Swedish environment (ter Schure *et al.* 2004). This congener was, however, not detected in urban snow. It should be noted that the analytical detection limit was higher for decaBDE (up to 0.10 μg/l) than for tetra- and pentaBDE (up to 0.010 μg/l).

Background concentrations of Σpolybrominated diphenyl ethers (PBDEs [varying number of congeners included]) in rain and bulk precipitation in Sweden have been detected in the low nanogram-per-litre-range (ter Schure *et al.* 2004). The PBDE concentrations in urban snow are similar to Swedish precipitation levels, which suggest that the PBDEs in snow may originate from atmospheric deposition. No other studies on the occurrence of BFRs in urban snow or stormwater have been found. The detection limits of HBCD and TBBP-A (up to 0.10 μg/l) in the current study are presumably too high compared to expected environmental concentrations (Remberger *et al.* 2002; 2004), and the substances were below the d.l. in all snow samples.

Chlorinated paraffins

The use of CPs has decreased by 80% in Sweden since 1994, but a substance flow analysis revealed that the largest emissions into the environment derive from large stocks of CPs in the urban area (Fridén and McLachlan 2007). Short-chained and medium-chained CPs were detected in two snow deposit samples (Heden and Gårdamotet) at 0.33 and 32 μg/l, respectively. The analytical detection limits of SCCPs and MCCPs were 0.20–5.0 μg/l (the higher value in one sample with large matrix effects), whereas many reported concentrations found in natural waters are in the nanogram-per-litre-range (Bayen *et al.* 2006).

Perfluorinated compounds

Perfluorinated alkylsulfonates (PFASs) and perfluorinated carboxylates (PFCAs) are two major PFC classes of environmental concern (KemI 2006; Kim and Kannan 2007; Fromme *et al.* 2009). The most studied PFAS is PFOS, which is considered to be the final degradation product of many commercially used sulfonated fluorochemicals, including PFOSA, and is shown to be exceptionally persistent in the environment. The most studied caboxylate, PFOA, is also persistent and have shown carcinogenicity and reproductive toxicity.

PFOA and PFOSA were detected in four and PFOS in five out of eight snow samples (Figure 2c). PFOS and PFOA are the most frequently detected PFCs in many environmental studies and concentrations found in this study (<d.l.–9.0 and <d.l.–36 ng/l, respectively) are lower or in the same range as those reported in stormwater in Japan (Murakami *et al.* 2009; Zushi and Masunaga 2009); rain and snow-induced runoff in the US (Kim and Kannan 2007); and comparably lower than concentrations found in fresh snow in China (Liu *et al.* 2009). The median concentrations of PFOS and PFOA in urban snow (0.81 and 2.3 ng/l, respectively) are similar to concentrations detected in precipitation in Sweden (median 2.2 and 3.1 ng/l, respectively [Woldegiorgis *et al.* 2006]). On the contrary, the PFOSA concentrations in snow (maximum 66 ng/l) are, when detected, one magnitude higher than most reported concentrations in stormwater and rain (Woldegiorgis *et al.* 2006; Kim and Kannan 2007;

Murakami *et al.* 2009). In addition, PFOSA concentrations being occasionally several times higher than PFOS and PFOA concentrations have not been observed in the referred studies.

Kim and Kannan (2007) found high concentrations of PFCs in runoff from parking lots and areas with high traffic intensity, and studies by Zushi and Masunaga (2009) revealed positive correlation between high PFCs concentrations in stormwater and transportation-related land use. The PFCs are added to several transportation-related products, such as car care products, water and dirt repellant car-seat textiles, upholstery and windshield washer fluids (KemI 2006; Murakami *et al.* 2009). Other potential sources in snow and runoff include impregnated snack and food wrappings, and water- and dirt-resistant products for shoes and clothes, which is the major application area for PFCs in Sweden (KemI 2006). The high PFOSA concentrations, compared to PFOS and PFOA, may be an indication of PFOSA point sources. It is a major metabolite of N-alkylperfluorosulfonamides, which have been used in paper and packaging applications and surface treatment of e.g. carpets and textiles (Fromme *et al.* 2009). Specific point sources of PFOSA have, however, not been identified in the sampling area.

Phthalates

Phthalates were analysed in snow sampled in both 2009 and 2010. The most frequently occurring phthalates in urban snow were DnBP, DiBP, DEHP, DIDP and DINP (detection frequency 55, 55, 100, 91 and 91%, respectively [Figure 2d]). The DEHP concentrations in snow (2.3–96 µg/l) all exceeded the EQS of 1.3 µg/l in surface waters. The highest phthalate concentrations were found in snow sampled in Järnbrott in March 2010, with maximum concentrations at 260 µg/l for DINP and 81 µg/l for DIDP. Compared to studies on urban stormwater from Sweden and Austria (Björklund *et al.* 2009; Clara *et al.* 2010), the snow samples show higher maximum concentrations of all phthalates, except DMP and DEP.

The contribution of DINP to total phthalate concentrations exceeded 40% in all samples, except the samples from Gårdamotet and Delsjön. Björklund *et al.* (2009) and Clara *et al.* (2010) showed similar results; high detected concentrations of DIDP, DINP and DEHP compared to most low molecular-weight phthalates. Within the European Union, the use of DEHP is currently restricted due to the substance’s toxic effects and DIDP and DINP have replaced DEHP in many applications (KemI 2010). Björklund *et al.* (2009) showed strong correlation between the relative phthalate distribution in stormwater sediment and the prevalent phthalate use in Sweden, which is dominated by DINP and DIDP. The current study further strengthens the theory that DIDP and DINP are the phthalates occurring at the highest concentrations in urban matrices in Sweden.

A substance flow analysis of phthalates revealed that traffic is an important source to these substances in stormwater (Björklund in press). Phthalates may be emitted from applications of soft PVC, which is the main use for the high molecular-weight phthalates, car undercoating, adhesives, sealants, rubber, and paints. Other phthalate sources include coated roofing and cladding, tarpaulins, cables, shoe and textile wear. Coated roofing and cladding materials are believed to be minor sources of phthalates in the road side snow, since all samples were collected from sites where the snow has no contact with buildings.

Polycyclic aromatic hydrocarbons

The PAHs could be detected in all road side snow samples (Figure 2e and f); the highest total concentrations were found in Järnbrott (5.6 and 5.2 µg/l), followed by Gårda (2.1 µg/l). The Gårdamotet snow deposit showed, however, the highest total PAH concentrations of all samples at 15 µg/l, which is at least 2.5 times higher than the concentrations found in the other samples. A study on stormwater quality in Gårda showed total PAH concentrations that are comparable to the concentrations found in snow in this area (Pettersson *et al.* 2005). The snow concentrations found in the current study are generally within the range of other reported snow studies (Viskari *et al.* 1997; Lindgren 1998). The European EQS for BaP in surface waters – MAC 0.1 µg/l – was, however, exceeded in five samples; the EQS for BbF+BkF – AA 0.03 µg/l – was exceeded in all samples, except the background sample; and the EQS for BPY+INP – AA 0.002 µg/l – was exceeded in all samples.

The PAHs generally detected at the highest concentrations, and also the most frequently occurring, were PYR, PHE, FLR and BPY (Figure 2e and f). The maximum detected concentration was found for BPY (3.1 µg/l) in the Gårdamotet deposit sample, whereas PYR generally showed higher concentrations in other samples. Among the US EPA classified carcinogenic PAHs, the most frequently occurring, and also the substances detected at the highest concentrations, were BbF, INP and CHY. The highest concentration was found for INP (1.5 µg/l) in Gårdamotet, whereas BbF generally showed higher concentrations in other samples. Similar trends in occurrence of PAHs have been observed in other studies of road side snow (Viskari *et al.* 1997; Reinosdotter *et al.* 2006).

Ravindra *et al.* (2008) have reviewed PAH source studies and summarize that PYR, FLR and PHE – the most abundant PAHs in the current study – are suggested as source markers for both incineration and vehicular emissions, including both diesel and petrol. Other suggested markers include oil combustion and wood burning as possible sources to the PAHs observed in our study. In addition, PAH profiles for petrol and diesel, lubricating oil, bitumen and asphalt, and tyre rubber were compared with the PAH content in urban snow. Moderate to strong correlations were seen between road side snow and tyre rubber (r ~ 0.7–0.9 [Lindgren 1998; Edeskär 2004]) and lubricating oil (r ~ 0.6 [Wang *et al.* 2000]).

CONCLUSIONS

- The snow samples contain a large amount of analytes with varying properties. Further fractionation and clean-up are therefore necessary steps to successfully identify specific pollutants in a non-target screening of SVOCs.
- The most frequently detected pollutants in urban snow were PAHs, high molecular-weight phthalates, 4-nonylphenol and 4-oktylphenol. Brominated flame retardants and chlorinated paraffins were only sporadically detected in urban snow.
- Strong correlation was generally observed for phthalate and PAH concentrations between sampling occasions, implying that the substance composition is similar in most samples. Within other substance groups and between substance groups, the correlation was generally moderate to weak. The snow was grab sampled and pollutant concentrations from different years and sites may not be comparable due to different climatic conditions, land use, snow-handling activities and changes in traffic intensity. In addition, the generation of pollutants in road side snow and snow deposits is not comparable.
- Due to the high snow concentrations of APs, DEHP, and PAHs, compared to water quality standards (no joint European EQS for other substances), urban snow should be deposited at sites were the meltwater is controlled and treated prior to discharge into natural waters.

ACKNOWLEDGEMENTS

This project was financially supported by Kretsloppskontoret Gothenburg and The Swedish Transport Administration. Lars-Ove Sörman, Chalmers, is gratefully acknowledged for his help during sampling.

REFERENCES

Bayen, S., Obbard, J.P. and Thomas, G.O. 2006 Chlorinated paraffins: A review of analysis and environmental occurrence. *Environ. Int.* **32**(7), 915-929.

Björklund, K. in press Substance Flow Analyses of Phthalates and Nonylphenols in Stormwater. *Accepted for publication in Water Sci. Technol.*

Björklund, K., Palm Cousins, A., Strömvall, A-M. and Malmqvist, P-A. 2009 Phthalates and nonylphenols in urban runoff: Occurrence, distribution and area emission factors. *Sci. Total Environ.* **407**(16), 4665-4672.

Clara, M., Windhofer, G., Hartl, W., Braun, K., Simon, M., Gans, O., Scheffknecht, C. and Chovanec, A. 2010 Occurrence of phthalates in surface runoff, untreated and treated wastewater and fate during wastewater treatment. *Chemosphere* **78**(9), 1078-1084.

de Wit, C.A. 2002 An overview of brominated flame retardants in the environment. *Chemosphere* **46**(5), 583-624.

DEFRA 2008 *4-tert-Octylphenol: Risk Reduction Strategy and Analysis of Advantages and Drawbacks*, Department for Environment, Food and Rural Affairs, London, UK.

Dsikowitzky, L., Schwarzbauer, J., Kronimus, A. and Littke, R. 2004 The anthropogenic contribution to the organic load of the Lippe River (Germany). Part I: qualitative characterisation of low-molecular weight organic compounds. *Chemosphere* **57**(10), 1275-1288.

Edeskär, T. 2004 *Technical and Environmental Properties of Tyre Shreds Focusing on Ground Engineering Applications*. Technical report 2004:05, Department of Soil Mechanics and Foundation Engineering, Luleå University of Technology, Luleå.

Fridén, U. and McLachlan, M. 2007 *Substansflödesanalys av klorparaffiner i Stockholms stad 2004 (Substance flow analysis of chlorinated paraffins in Stockholm 2004)*. ISSN: 1653-9168, Stockholm.

Fromme, H., Tittlemier, S.A., Völkel, W., Wilhelm, M. and Twardella, D. 2009 Perfluorinated compounds - Exposure assessment for the general population in western countries. *Int. J. Hyg. Envir. Heal.* **212**(3), 239-270.

KemI 2003 *Bromerade flamskyddsmedel – förutsättningar för ett nationellt förbud (Investigating the prerequisites for a national ban on brominated flame retardants)*. Report 4/03, Swedish Chemicals Agency, Stockholm, Sweden.

KemI 2006 *Perfluorinated substances and their uses in Sweden*. Report 7/06, Swedish Chemicals Agency (KemI), Sundbyberg, Sweden.

KemI 2010 KemI-stat (statistical information based on data in the Swedish Chemicals Agency´s products register and pesticides register). http://apps.kemi.se/kemistat/start.aspx (accessed May 2010).

Kim, S-K. and Kannan, K. 2007 Perfluorinated Acids in Air, Rain, Snow, Surface Runoff, and Lakes: Relative Importance of Pathways to Contamination of Urban Lakes. *Environ. Sci. Technol.* **41**(24), 8328-8334.

Langford, K.H., Scrimshaw, M.D., Birkett, J.W. and Lester, J.N. 2005 Degradation of nonylphenolic surfactants in activated sludge batch tests. *Water Res.* **39**(5), 870-876.

Lindgren, Å. 1998 *Road construction materials as a source of pollutants*. PhD Thesis, Division of Traffic Engineering, Luleå University of Technology, Luleå.

Liu, W., Jin, Y., Quan, X., Sasaki, K., Saito, N., Nakayama, S.F., Sato, I. and Tsuda, S. 2009 Perfluorosulfonates and perfluorocarboxylates in snow and rain in Dalian, China. *Environ. Int.* **35**(4), 737-742.

Meyer, T. and Wania, F. 2008 Organic contaminant amplification during snowmelt. *Water Res.* **42**(8-9), 1847-1865.

Murakami, M., Shinohara, H. and Takada, H. 2009 Evaluation of wastewater and street runoff as sources of perfluorinated surfactants (PFSs). *Chemosphere* **74**(4), 487-493.

Paxéus, N. 2000 Organic compounds in municipal landfill leachates. *Water Sci. Technol.* **42**(7-8), 323-333.

Pettersson, T.J.R., Strömvall, A-M. and Ahlman, S. 2005 Underground sedimentation systems for treatment of highway runoff in dense city areas. *Proc. of the 10th Int. Conf. on Urban Drainage*, 21-26 August 2005, Copenhagen, Denmark.

Ravindra, K., Sokhi, R. and Van Grieken, R. 2008 Atmospheric polycyclic aromatic hydrocarbons: Source attribution, emission factors and regulation. *Atmos. Environ.* **42**(13), 2895-2921.

Reinosdotter, K., Viklander, M. and Malmqvist, P-A. 2006 Polycyclic aromatic hydrocarbons and metals in snow along a highway. *Water Sci. Technol.* **54**, 195-203.

Remberger, M., Sternbeck, J., Palm, A., Kaj, L., Strömberg, K. and Brorström-Lundén, E. 2004 The environmental occurrence of hexabromocyclododecane in Sweden. *Chemosphere* **54**(1), 9-21.

Remberger, M., Sternbeck, J. and Strömberg, K. 2002 *Screening av triclosan och vissa bromerade fenoliska ämnen i Sverige (Screening of triclosan and certain brominated phenolic compounds in Sweden).* Report B1477, IVL Svenska miljöinstitutet, Stockholm, Sweden.

ter Schure, A.F.H., Agrell, C., Bokenstrand, A., Sveder, J., Larsson, P. and Zegers, B.N. 2004 Polybrominated diphenyl ethers at a solid waste incineration plant II: atmospheric deposition. *Atmos. Environ.* **38**(30), 5149-5155.

Wang, J., Jia, C.R., Wong, C.K. and Wong, P.K. 2000 Characterization of Polycyclic Aromatic Hydrocarbons Created in Lubricating Oils. *Water Air Soil Pollut.* **120**(3), 381-396.

Westerlund, C. and Viklander, M. 2006 Particles and associated metals in road runoff during snowmelt and rainfall. *Sci. Total Environ.* **362**(1-3), 143-156.

Viskari, E.L., Rekilä, R., Roy, S., Lehto, O., Ruuskanen, J. and Kärenlampi, L. 1997 Airborne pollutants along a roadside: Assessment using snow analyses and moss bags. *Environ. Pollut.* **97**(1-2), 153-160.

Woldegiorgis, A., Andersson, J., Remberger, M., Kaj, L., Ekheden, Y., Blom, L., Brorström-Lundén, Borgen, A., Dye, C. and Schlabach, M. 2006 *Results from the Swedish national screening programme 2005 – Perfluorinated Alkylated Substances*. Report B1698, IVL Svenska miljöinstitutet, Stockholm, Sweden.

Ying, G-G., Williams, B. and Kookana, R. 2002 Environmental fate of alkylphenols and alkylphenol ethoxylates – a review. *Environ. Int.* **28**(3), 215-226.

Zushi, Y. and Masunaga, S. 2009 Identifying the nonpoint source of perfluorinated compounds using a geographic information system based approach. *Environ. Toxicol. Chem.* **28**(4), 691-700.