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## Seasonal soil/snow-air exchange of semivolatile organic pollutants at a coastal arctic site (Tromsø, 69°N)



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### HIGHLIGHTS

- Soil/snow-air exchange was evaluated at Tromsø for 98 semi-volatile organic pollutants.
- PCBs and HCHs gas-phase concentrations showed a temperature dependent seasonality.
- Fugacity in air was correlated with fugacity in soil/snow for PAHs, PCBs and OCPs.
- Net volatilization of PAHs and HCBs during the warmer months
- PCBs and HCHs in air and soil/snow were close to equilibrium.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Soils are a major reservoir of semivolatile organic pollutants such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs), and exert a control on their atmospheric occurrence. We present here an assessment of the atmospheric occurrence and seasonality of soil/snow-air partitioning and exchange of PCBs, PAHs, hexachlorobenzene (HCB), and hexachlorocyclohexanes (HCHs) in the arctic city Tromsø, northern Norway. The fugacities of the organic pollutants in soils and snow were determined using a soil fugacity sampler by equilibrating the air concentrations with those in the surface soil/snow. The concentrations in soils did not show a significant seasonality. Conversely, the ambient air concentrations and the soil (or snow) fugacity showed a clear seasonality for PCBs, HCH, HCB and some PAHs, related to temperature. Fugacities in soil/snow were correlated with those in the ambient gas phase, suggesting a close seasonal air-soil/snow coupling. Generally, there was a net deposition or close to equilibrium conditions during the winter, which contrasts with the net volatilization observed during the warmer periods. The chemicals with lower octanol-air partition coefficients showed a larger tendency for being volatilized and thus remobilized from this coastal arctic environment. Conversely, the more hydrophobic compounds were close to air-soil/snow equilibrium or showed a net deposition.

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### 1. Introduction

Semi-volatile organic compounds (SVOCs), such as organochlorine pesticides (OCPs), polycyclic aromatic hydrocarbons (PAHs) and

polychlorinated biphenyls (PCBs), are ubiquitous pollutants of global environmental concern (UNEP, 2001). The effectiveness of important global regulations aimed to control emissions of some legacy SVOCs, such as the Stockholm Convention on persistent organic pollutants (POPs), has led to a decline of their atmospheric concentrations in many cases (UNEP, 2001; Nizzetto et al., 2010; Dvorská et al., 2009). However, the long range atmospheric transport (LRAT) potential and

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slow degradability of some of these compounds sustain their occurrence, from urban and industrial areas to background and remote environments (Galbán-Malagón et al., 2013a, 2013b; Cabrerizo et al., 2013b, 2014, 2016).

Soils are a major reservoir and sink for hydrophobic SVOCs due to their high affinity to organic matter (Dalla Valle et al., 2005). Aside from direct inputs, the occurrence of SVOCs in soils and the atmosphere depends also on the mechanisms of atmospheric deposition and volatilization. The net direction of the soil-air and snow-air diffusive exchange (i.e. transport of gas phase SVOCs at the soil/snow-air interface) is driven by environmental factors, mainly temperature and the soil organic matter (SOM) (Cabrerizo et al., 2011a, 2011b, 2013a, 2013b, 2014). Soil coverage (i.e. soil with grass versus snow) can also affect SVOCs air-surface partitioning. Soils can have higher amounts of organic matter, while snow has a large specific surface area. The study of the direction of soil-air diffusive exchange for a specific chemical can be approached by measuring the chemical's fugacity in air and soil. Fugacity describes the escaping tendency of a chemical from a specific medium, such as soil, snow or air (Mackay and Paterson, 1981). The previously described soil fugacity sampler (Cabrerizo et al., 2009) allows its direct measurement under field conditions, and thus the soil/snow fugacity does not need to be estimated from POP concentrations in soils and/or models. Previous fugacity based field studies of soil-air exchange of POPs focused on temperate European background sites (Cabrerizo et al., 2011a, 2011b, 2011c, 2013a; Degrendele et al., 2016), and polar regions in the southern hemisphere (Antarctica) (Cabrerizo et al., 2013b, 2014). In a recent study, diurnal variations in air and soil fugacity were characterized at a background region of central Europe (Degrendele et al., 2016), and tied to temperature changes among other factors. However, the seasonal effects of temperature and other climatic/biogeochemical variables on the soil-air diffusive exchange have only been partially explored in regions from the northern Iberian Peninsula (Cabrerizo et al., 2011a, 2011b, 2011c), but not in other climatic regions, and few or no data exist in polar regions from the northern hemisphere. The soils from high latitude regions can have a high reserve capacity for POPs (Dalla Valle et al., 2005), and are important for the global distribution of POPs and to study the potential impact of these toxic chemicals in these fragile ecosystems.

The main objective of this study was to investigate, for the first time, the seasonality of the air-soil/snow exchange of PCBs, OCPs and PAHs in an arctic region (Tromsø 69°N) where soils are frozen, seasonally covered with snow, and with a high temperature range (25 °C) between summer and winter. In order to achieve this objective, a 14-month sampling campaign using a fugacity sampler was performed.

## 2. Materials and methods

### 2.1. Sampling

All samples were collected in a carefully selected urban site in the main island of Tromsø (Tromsøya), Norway (N 69° 38' 35.8" E 18° 56' 54.5"). A fugacity sampler (Cabrerizo et al., 2009) was deployed for 5 or 6 days in 14 sampling events between May 2013 and July 2014. This sampler allows for the simultaneous collection of an ambient air sample and a sample of air equilibrated in-situ with the soil/snow surface for each sampling event. The latter is a measure of the fugacity of the chemicals in the top soil/snow (Cabrerizo et al., 2009, 2013a, 2013b). Air-soil/snow equilibrium is ensured by operating at a low sampling flow rate of 8–10 L min<sup>-1</sup>, thus the air flowing below a stainless-steel plate of 1 m<sup>2</sup> located 3 cm above the soil/snow surface equilibrates with the underlying surface as demonstrated for soil surfaces (Cabrerizo et al., 2009). The application of this sampler to snow surface has also provided consistent results (Cabrerizo et al., 2013a, 2013b). In addition, a reference air sample collected at 1.5 m height allows the monitoring of contaminant air concentrations in the lower atmosphere. In both samples, the air passes through pre-combusted glass fiber filters (GF/F,

47 mm diameter) to remove particulate matter and 10 × 2 cm pre-cleaned polyurethane foam plug (PUFs), in which the gas phase compounds are retained. The average air volume sampled was 67 ± 15 m<sup>3</sup>. Sampling events, dates and air temperatures are given in the Supplementary Information (Table S1, SI).

The ground at the sampling site was frozen and/or covered by ice or snow between October 2013 and April 2014. Eight soil samples were taken after each soil fugacity samplings between May and September 2013 and between May and July 2014, by gently collecting the soil surface layer (top 0.5–1 cm). After sampling, all samples were stored in freezers at -20 °C until analysis. Soil was not sampled during the snow covered period, from November 2013 to April 2014.

### 2.2. Analytical procedures

All concentrations were determined from single samples. All samples were Soxhlet extracted with acetone:hexane (3:1, v:v) for 24 h and fractionated as described elsewhere (Cabrerizo et al., 2009, 2013a). Hexachlorobenzene (HCB), hexachlorocyclohexane isomers ( $\alpha$ -HCH,  $\beta$ -HCH,  $\gamma$ -HCH,  $\delta$ -HCH) and PCB congeners were analysed by a gas chromatograph equipped with a  $\mu$ -electron capture detector (GC- $\mu$ -ECD, Agilent Technologies, model 7890 N) with a 60 m (0.25 mm i.d. × 0.25  $\mu$ m film thickness) DB-5 capillary column. The instrument was operated in splitless mode (closed for 1.5 min) and the oven temperature program started at 90 °C to 190 °C at 15 °C/min, then to 203 °C at 3 °C/min (held for 5 min), to 290 °C at 3 °C/min, and finally to 310 °C at 5 °C/min (held for 10 min). Injector and detector temperatures were 280 and 320 °C, respectively. Helium and nitrogen were used as carrier (1.5 mL/min) and makeup (60 mL/min) gases, respectively, and 2  $\mu$ L of sample were injected. The following PCB congeners were analysed: tri-PCB 18, 17, 31, 28; tetra-PCB 52, 49, 44, 74, 70; penta-PCB 95, 99/101, 87, 118; hexa-PCB 110, 151, 149, 153, 132, 105, 138, 158, 128, 169; hepta-PCB 187, 183, 177, 171, 156, 180, 191, 170; octa-PCB 201/199, 195, 194, 205; nona-PCB 206, 208; deca-PCB 209.

PAHs were analysed by a gas chromatograph coupled to a mass spectrometer (GC/MS). Samples were injected in an Agilent 6890 GC System with a 30 m (0.25 mm i.d. × 0.25  $\mu$ m film thickness) capillary column (HP-5MS) coupled with a mass spectrometer Agilent 5973 (GS-MS) operating in selected ion monitoring (SIM) and electron impact mode (71 eV). The oven temperature was increased to 90 °C (held for 1 min), then increased to 175 °C at 6 °C/min (held for 4 min), increased to 235 °C at 3 °C/min, increased to 300 °C at 8 °C/min (held for 8 min), and finally to 315 °C at 3 °C/min (held for 8 min). Injector and transfer line temperatures were 280 °C and 300 °C, respectively, and 2  $\mu$ L of sample were injected.

The following parent PAHs and alkyl homologues were analysed (53 individual PAHs): acenaphthylene (Act), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), dibenzothiophene (DBT), fluoranthene (Flt), pyrene (Pyr), benzo[a]anthracene (B[a]ant), chrysene (Cry), benzo[b]fluoranthene (B[b]f), benzo[k]fluoranthene (B[k]f), benzo[e]pyrene (B[e]pyr), benzo[a]pyrene (B[a]pyr), perylene (Pery), dibenzo[a,h]anthracene (Dib[a,h]ant), benzo[g,h,i]perylene (B[g,h,i]pery), indeno[1,2,3-cd]pyrene (In[1,2,3-cd]pyr), benzo[g,h,i]fluoranthene (B[g,h,i]f), methylphenanthrenes ( $\Sigma$ MP, sum of 4 isomers), dimethylphenanthrenes ( $\Sigma$ DMPD, sum of 7 isomers), methyl dibenzothiophenes ( $\Sigma$ MDBT, sum of 3 isomers), dimethyldibenzothiophenes ( $\Sigma$ DMDBT, sum of 5 isomers), methylpyrenes ( $\Sigma$ MPyr, sum of 5 isomers), dimethylpyrenes ( $\Sigma$ DMPyr, sum of 8 isomers), methylchrysenes ( $\Sigma$ MCry, sum of 2 isomers).

### 2.3. Quality assurance/quality control

To minimize contamination during analysis, PUF plugs were initially pre-cleaned by Soxhlet extraction (with acetone/hexane (3:1, v:v), 24 h) before sampling. Filters were individually wrapped in aluminium foils and pre-combusted at 450 °C for 4 h.

Procedural and/or field blanks (4 and 4, respectively) were analysed with each batch of 4–5 PUF samples to monitor potential contamination during sampling and extraction. Soil potential contamination was monitored by 3 procedural blanks. Field blanks consisted of PUFs that had travelled and been handled together with the field samples. Procedural blanks were the result of analytical procedures without PUF. The limits of quantification (LOQs) were defined as the mean concentration of field or procedural blanks (using always the more restrictive of the two) plus three times the standard deviation of the blank response. For the analytes which were not found in procedural blanks, LOQs were derived from the lowest standard in the calibration curve.

To monitor recoveries, samples were spiked prior to extraction with PCB congeners 65 and 200 and deuterated PAH standards (acenaphthene-d10, phenanthrene-d10, crysene-d12 and perylene-d12) which were used as surrogate standards. The quantification followed the internal standard procedure, using anthracene-d10, pyrene-d10, p-terphenyl-d14, and benzo[b]fluoranthene-d12 for PAHs and PCB congeners 30 and 142. Average recoveries ranged from  $58 \pm 20\%$  for acenaphthene-d10 to  $110 \pm 22\%$  for crysene-d12 for PUF samples, and from  $58 \pm 16\%$  for acenaphthene-d10 to  $110 \pm 11\%$  for perylene-d12 for soil samples (Table S2, SI). Recoveries of PCB surrogates ranged from  $70 \pm 28\%$  to  $80 \pm 29\%$  for PUF samples, and from  $74 \pm 22\%$  to  $81 \pm 10\%$  for soil samples (Table S2, SI). One sample was discarded for PAH results due to low PAH surrogate recoveries ( $<10\%$ ).

### 3. Results and discussion

#### 3.1. Atmospheric concentrations

Results from the air samples collected at 1.5 m height revealed ambient air concentrations for  $\sum_{53}\text{PAHs}$ ,  $\sum_{40}\text{PCBs}$  and  $\sum_{5}\text{OCPs}$  in the ranges  $2.6\text{--}34 \text{ ng m}^{-3}$ ,  $64\text{--}460 \text{ pg m}^{-3}$  and  $26\text{--}270 \text{ pg m}^{-3}$ , respectively (Fig. 1, Tables S3–4, SI). These atmospheric levels are higher than in other rural and remote areas of Europe (Shahpoury et al., 2015; Nizzetto et al., 2010, 2017; Anttila et al., 2016; Halse et al., 2011; Hung et al., 2010, 2016), yet below the average concentrations of northern European urban areas (Li et al., 2009). Although the sampling site was based in an urban coastal area, these low SVOC atmospheric concentrations are consistent with Tromsø's low population density (around 3000 persons  $\text{km}^{-2}$ ).

The PCB and PAH concentration profiles in the gas phase were dominated by the lower molecular weight (MW) compounds with the higher MW compounds frequently below LOQ (Tables S4–5, Figs. S1–2, SI), consistent with previous studies (Shahpoury et al., 2015; Nizzetto et al., 2010, 2017; Anttila et al., 2016; Halse et al., 2011). PAHs with 3 aromatic rings contributed on average to  $71 \pm 15\%$  of  $\sum_{53}\text{PAHs}$ , with Phe as the main contributor ( $29 \pm 7.9\%$ ) to  $\sum_{53}\text{PAHs}$  air concentrations.  $\sum_{\text{Hexa-PCBs}}$  and  $\sum_{\text{Tetra-PCBs}}$  were the main contributors to  $\sum_{40}\text{PCBs}$  ( $32 \pm 22\%$  and  $32 \pm 17\%$ , respectively).  $\sum_{5}\text{OCPs}$  composition was dominated by HCB ( $84 \pm 8.9\%$ ), with HCB air concentrations ranging from  $<\text{LOD}$  to  $270 \text{ pg m}^{-3}$ , and  $\sum_{4}\text{HCHs}$  air concentrations from 2.6 to  $42 \text{ pg m}^{-3}$ .

Generally, atmospheric concentrations showed a marked seasonal variability with highest concentrations for most of the targeted SVOCs during the warmest months (July and August 2013), and minimum concentrations during the winter (November 2013 to February 2014) (Fig. 1). The temperature dependent seasonality in atmospheric concentrations was supported by significant correlations of the gas phase concentrations with the inverse of air temperature for  $\sum_{40}\text{PCBs}$  ( $r^2_s = 0.30$ ,  $p < 0.05$ ) and  $\sum_{4}\text{HCHs}$  ( $r^2_s = 0.48$ ,  $p < 0.01$ ), as well individual HCHs, PCB congeners and some PAHs (Fig. 2, Table S5, SI). This is consistent with previous studies reporting temperature dependent atmospheric concentrations of semivolatile POPs (Wania et al., 1998; Brunciak et al., 2001; Cortes et al., 1998; Castro-Jiménez et al., 2009), and constitutes evidence of a dominant role of surface-air exchange as a driver of the atmospheric occurrence of POPs (Wania et al., 1998;

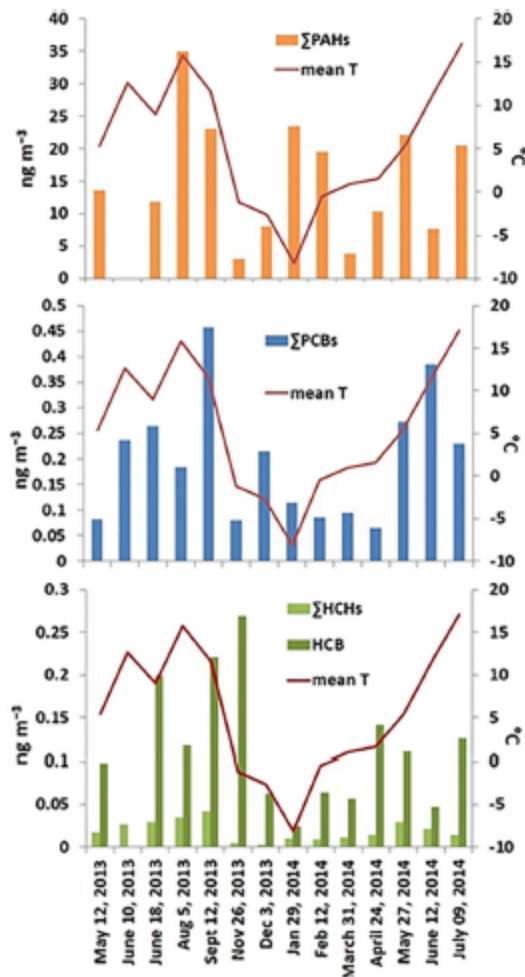


Fig. 1. Gas phase concentrations of  $\sum_{5}\text{OCPs}$ ,  $\sum_{40}\text{PCBs}$ , and  $\sum_{53}\text{PAHs}$ , and mean air temperature ( $^{\circ}\text{C}$ ). Soil was snow covered from November 2013 to April 2014.

Cabrero et al., 2013a, 2013b, 2014), probably from proximate land regions. However, gas-particle SVOCs partitioning is also driven by temperature (Finizio et al., 1997), which could contribute to higher gas phase concentrations at higher temperatures. In contrast, isolated arctic areas at similar or even lower latitudes have been shown to present a lack of temperature dependent atmospheric concentrations which may reflect long range transport (Oehme et al., 1996; Stern et al., 1997; Wania et al., 1998), especially over oceanic regions where air and water are often at dis-equilibrium. The concentrations of  $\sum_{\text{MePAHs}}$  and some of the analysed PAHs (DBT,  $\sum_{\text{MDBT}}$ ,  $\sum_{\text{MP}}$  and  $\sum_{\text{DMPD}}$ ) were also significantly correlated with the inverse of air temperature (Fig. 2, Table S5, SI), however, this correlation was not significant for  $\sum_{53}\text{PAHs}$  as most of the individual PAHs did not show a clear seasonality pattern. This lack of temperature dependence of PAHs in the gas phase is consistent with reports at temperate urban regions where local anthropogenic sources, such as vehicular traffic or domestic heating, have a strong influence on PAHs atmospheric concentrations during the winter (Gigliotti et al., 2000). The observed high  $\sum_{53}\text{PAHs}$  atmospheric concentrations from December to February are likely related to an increase of wood-based household heating during winter in this region (Launhardt et al., 1998; Solli et al., 2009).

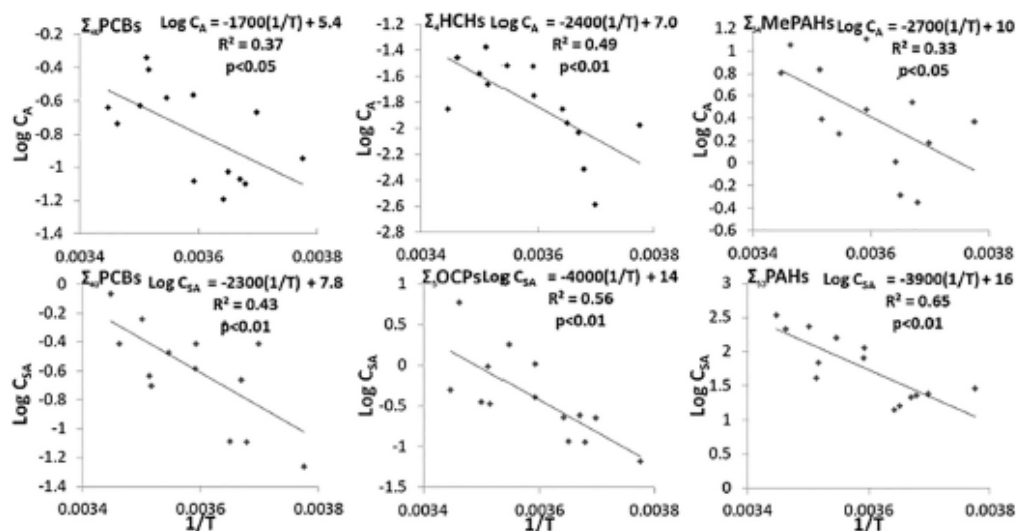


Fig. 2. Linear least squares regressions of ambient gas phase concentrations ( $C_A$ ,  $\text{ng m}^{-3}$ ) (upper panels), and the gas phase concentrations equilibrated with the soil/snow surface ( $C_{SA}$ ,  $\text{ng m}^{-3}$ ) (lower panels), for  $\Sigma_5OCPs$ ,  $\Sigma_{40}PCBs$ ,  $\Sigma_{Me}PAHs$  and  $\Sigma_{53}PAHs$  versus the inverse of the mean air temperature (K).

72-Hour air mass back-trajectories were also evaluated every 12 h for each sampling event using the US National Atmospheric and Oceanic Administration (NOAA) Hysplit model (Draxler, 2010) (Fig. S3, SI). Due to the long duration of each sampling event, samples integrate the concentrations of air masses with diverse back-trajectories, in some cases from opposite directions. Higher concentrations air samples did not coincide with a dominance of southern (mainland) back-trajectories, nor lower concentrations with northern air masses. This is consistent with a larger relevance of local sources and remobilization processes driving the atmospheric concentrations in Tromsø.

### 3.2. Occurrence in surface soils

Concentrations of  $\Sigma_{53}PAHs$ ,  $\Sigma_{40}PCBs$  and  $\Sigma_5OCPs$  in soils ranged from 67 to 290  $\text{ng g}_{\text{dw}}^{-1}$ , 0.3 to 0.95  $\text{ng g}_{\text{dw}}^{-1}$  and 0.67 to 1.5  $\text{ng g}_{\text{dw}}^{-1}$ , respectively (Tables S6–7, SI). PCB composition was dominated by  $\Sigma$ Hexa-PCBs (40  $\pm$  2.3%) to  $\Sigma_{40}PCBs$ , followed by  $\Sigma$ Penta- and  $\Sigma$ Hepta-PCBs, while the lighter  $\Sigma$ Tri-PCBs had a small contribution to  $\Sigma_{40}PCBs$  (3.5  $\pm$  3.9%) (Fig. S2, SI). PAH profile was also dominated by the 4 aromatic rings parent- and methylated-hydrocarbons (Flu, Pyr, Cry, B[a]ant,  $\Sigma$ MPyr,  $\Sigma$ DMPyr and  $\Sigma$ MCry), which accounted for 46  $\pm$  3.4% of  $\Sigma_{53}PAHs$  (Fig. S1, SI). These concentrations and profiles are comparable to results from previous studies at high latitude or temperate European regions (Zhu et al., 2015; Meijer et al., 2002; Ma et al., 2008; Nam et al., 2008; Cabrerizo et al., 2011a, 2011b, 2011c; Bosch et al., 2015; Holubek et al., 2009; Schuster et al., 2011). HCB was the main contributor (96  $\pm$  3.9%) to  $\Sigma_5OCPs$ , with soil concentrations ranging from 0.61 to 1.5  $\text{ng g}_{\text{dw}}^{-1}$ .

SOM is generally regarded as a key factor governing the soil reservoir capacity (Dalla Valle et al., 2005) and soil-air partitioning and exchange for hydrophobic pollutants (Hamer et al., 2001; Cabrerizo et al., 2011a, 2011b, 2011c, 2013a). Correlations between POPs concentrations in soil and SOM (or the fraction of organic carbon ( $f_{oc}$ )) have been reported for several biomes including high latitude polar regions (Meijer et al., 2003; Ribes et al., 2003; Cabrerizo et al., 2012; Schuster et al., 2011). In this study, however, no significant correlations were found between  $f_{oc}$  and the concentrations in soil of the target compounds. This lack of correlation does not undermine the role of SOM in air-soil partitioning but rather reflects the limited  $f_{oc}$  variability among

samples, ranging from 6.5 to 10%, since all samples were collected at the same sampling site, and thus were not covering a spatial range as in other studies. Furthermore, SOM quality and structural composition also play an important role in air-soil partitioning (Mader et al., 1997; Valsaraj et al., 1999), even though the chemical composition of OC was not evaluated in these samples. Average air temperatures for the soil sampling periods ranged from  $-5.4$  to  $17$   $^{\circ}\text{C}$ . Concentrations of PAHs, PCBs and OCPs in soils showed no clear seasonality and did not present significant correlations with air temperature for any of the target compounds, reflecting the relevance of soils as a reservoir of SVOCs, due to its fugacity capacity hundreds of times higher than in the atmosphere (Dalla Valle et al., 2005).

### 3.3. Soil/snow-air partitioning of SVOCs

The profiles of gas phase concentrations equilibrated with the soil or snow surface ( $C_{SA}$ ), directly proportional to the soil or snow fugacity and obtained with the fugacity sampler, were similar to those found in ambient air ( $C_A$ ), with an even lower contribution of high MW compounds (Figs. S1–2, SI). PAHs with 3 aromatic rings contributed on average to 90  $\pm$  6.9% of  $\Sigma_{53}PAHs$  and  $\Sigma$ Tetra-PCBs were the main contributors to  $\Sigma_{40}PCBs$  (47  $\pm$  17%).  $C_{SA}$  concentrations were generally higher than  $C_A$  (Fig. S5, Tables S8–9, SI) and showed stronger seasonal patterns with higher SVOC concentrations during warmer months. Furthermore,  $C_{SA}$  presented significant correlations with air temperature for  $\Sigma_{53}PAHs$ ,  $\Sigma_{40}PCBs$  and  $\Sigma_5OCPs$  (Fig. 2), consistent with the role of temperature as a primary control on the soil-air and snow-air partitioning and exchange of SVOCs. Information on the statistically significant  $C_{SA}$  correlations with air temperature for 17 of the target compounds are compiled in Table S10 (SI). Unlike  $C_A$ ,  $C_{SA}$  seasonality patterns were likely unaffected by local PAHs sources due to its higher concentrations.

Soil-air partition coefficients ( $K_{SA}$ ) were calculated as:

$$K_{SA} = \frac{C_S}{C_A} = \frac{f_{oc}}{\rho_{OC}} \left( \frac{\gamma_{OC-MW_{OC}}}{\gamma_{OC-MW_{OC}}} \right) K_{OA}$$

where  $C_S$  ( $\text{ng kg}_{\text{dw}}^{-1}$ ) is the individual SVOC concentration in surface soil, and  $C_A$  ( $\text{ng dm}^{-3}$ ) is the gas phase concentration that has been

equilibrated with the soil or snow surface (Tables S8–9, S1) as measured using the soil fugacity sampler.  $K_{SA}$  ( $L\ kg^{-1}$ ) has been previously modelled as a function of the octanol air partition coefficient ( $K_{OA}$ ), where  $\gamma_{OCT}$  and  $\gamma_{OC}$  are the activity coefficients of SVOCs in octanol and organic carbon, respectively,  $MW_{OCT}$  ( $g\ mol^{-1}$ ) and  $MW_{OC}$  ( $g\ mol^{-1}$ ) are the molecular weight of octanol and organic carbon, respectively,  $\rho_{OCT}$  is the octanol density, and  $f_{oc}$  is the fraction organic carbon of the soil (Finizio et al., 1997).  $K_{OA}$  was temperature corrected as reported elsewhere (Pamis et al., 2015; Shoeib and Harner, 2002; Zhang et al., 1999), and the value of  $\frac{(\gamma_{OCT}MW_{OCT})}{(\gamma_{OC}MW_{OC})}$  was assumed to equal unity (Cabrero et al., 2009).

Least squares linear regressions of  $\log K_{SA}$  versus  $\log K_{OA}$  showed statistically significant correlations for PAHs and PCBs (Fig. 3) explaining between 20 and 46% of the variability. It is noteworthy that these  $K_{SA}$  have been estimated from measurements of the fugacity in soil and not from models. These results are consistent with previous studies and support the role of SOM in soil-air partitioning of hydrophobic compounds (Finizio et al., 1997; Cabrero et al., 2011a, 2011b, 2011c). The slopes of  $\log K_{SA}$  versus  $\log K_{OA}$  differed considerably from 1 (Fig. 3). Additionally, there were large differences between the value of  $\log K_{SA}$  and  $\log K_{OW}$  for some compounds, which undermines the use of  $K_{OA}$  as a singular air-soil partitioning predictor. Furthermore, OCPs linear regression presented a negative slope since HCB presented higher  $K_{SA}$  values than HCHs (Fig. 3). However, the regression was not statistically significant for HCB or  $\sum_4$ HCHs considered separately (Fig. S4, S1).

### 3.4. Fugacity in soils, snow and air

The fugacities of the SVOC in air ( $f_a$ , Pa) and in the snow or soil ( $f_s$ , Pa) were calculated by:

$$f_a = 10^{-9} C_a RT / MW$$

$$f_s = 10^{-9} C_s RT / MW$$

where  $R$  is the gas constant ( $8.314\ Pa\ m^3\ mol^{-1}\ K^{-1}$ ),  $T$  is the temperature (K), and  $MW$  is the chemical molecular weight ( $g\ mol^{-1}$ ).

Least squares linear regressions of  $\log f_s$  versus  $\log f_a$  (Fig. 4) also showed statistically significant correlations for PAHs, OCPs and PCBs with  $R^2$  values of 0.63, 0.56 and 0.18, respectively. In general,  $f_s$  values were lower than  $f_a$ , which is reflected in linear regressions that presented slopes below 1. This is consistent with previous fugacity based studies on background temperate areas (Cabrero et al., 2013a) and Antarctica (Cabrero et al., 2013b), and further supports the relevance of secondary emissions as drivers of the atmospheric concentrations and points to the close coupling between air and soils/snow surface in terms of SVOC occurrence.

The comparison of  $f_s$  and  $f_a$  values allows determining the direction of soil/snow – air exchanges.  $f_s/f_a$  ratios higher than 1 reflect a net volatilization of a chemical. In return,  $f_s/f_a$  ratios lower than 1 indicate the net deposition of the chemical. The effect of the uncertainty in the measurements has been previously quantified (Cabrero et al., 2011a, 2011b), and air-soil/snow equilibrium is considered for  $\ln f_s/f_a$  values between  $-1.2$  and  $0.53$ .

$f_s/f_a$  ratios showed a large variability for individual PAHs, PCBs and OCPs depending on the compound hydrophobicity (Tables S11–12, S1, Fig. 5) and ambient temperature (Fig. 6). Statistically significant least squares linear regressions of  $\ln f_s/f_a$  versus  $\log K_{OA}$  showed that in general the most hydrophobic compounds were closer to equilibrium or showed a net deposition, regardless of the season (Fig. 5). However, all target compounds with  $\log K_{OA}$  lower than 10 showed a net volatilization during part of the year, especially the warmer months. This is consistent with previous studies and supports the relevance of soil volatilization as a source of lighter SVOCs (Cabrero et al., 2011a, 2011b,

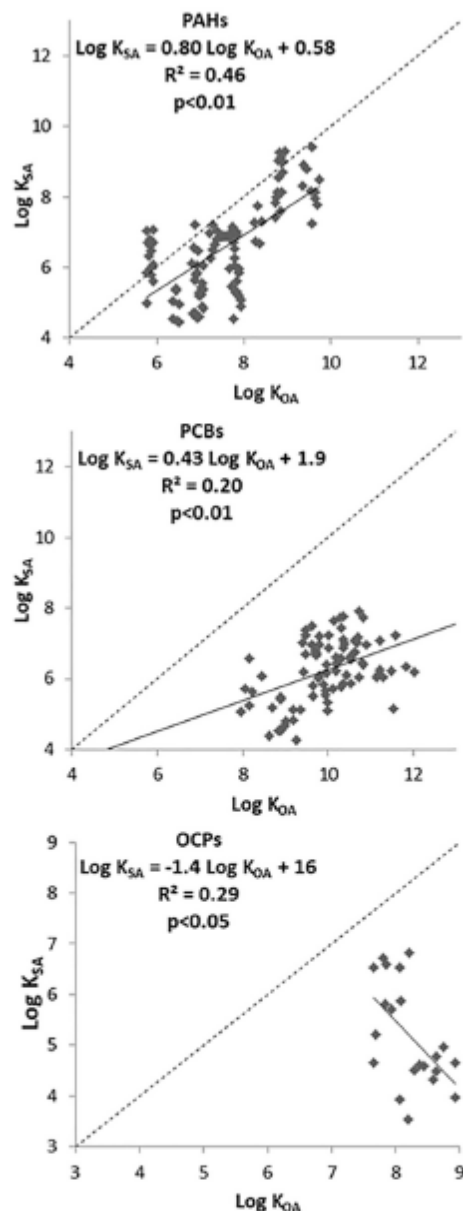


Fig. 3. Correlations of the soil-air partition constants ( $K_{SA}$ ) versus the octanol-air partition constant ( $K_{OA}$ ) for PAHs, PCBs and OCPs. Dashed lines mark  $K_{SA} = K_{OA}$ .

2011c, 2013a). However,  $\ln f_s/f_a$  showed poor correlations with  $\log K_{OA}$ , explaining only 5 and 11% of variability for PAHs and PCBs, respectively. OCPs  $\ln f_s/f_a$  ratios presented no significant correlation with  $\log K_{OA}$  (Fig. S6, S1), although for its main contributor, HCB, a linear regression was statistically significant and explained 42% of variation due to the influence of temperature on  $K_{OA}$  (Fig. 5).

PAHs average  $\ln f_s/f_a$  ratios were between  $-1.2$  and  $0.53$  (near equilibrium) from January to March, and higher than  $0.53$  (volatilization) during the rest of the year (Table S11, S1). In general, the maximum individual PAH  $\ln f_s/f_a$  ratios were detected during June and July of 2013 and 2014 (Table S11, S1). This is consistent with a higher relevance of soils as a source of PAHs, HCB and other SVOCs during warm conditions,

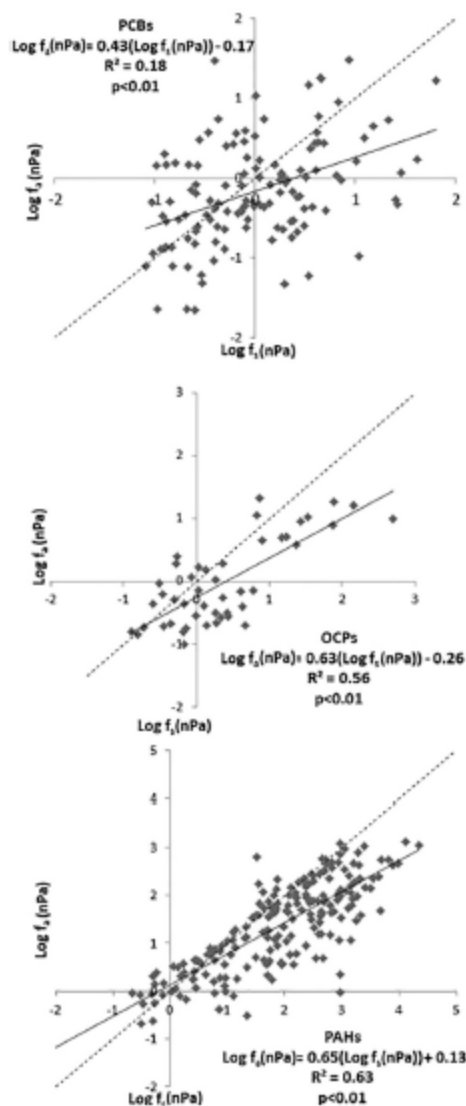


Fig. 4. Fugacity in air ( $f_a$ ) versus fugacity in soil ( $f_s$ ) for PCBs, OCPs and PAHs.

contributing to the temperature dependent gas phase concentrations (Fig. 2), through a close air-soil coupling of concentrations (Fig. 4). The low ratios observed in winter reflect the high sorbing capacity of snow for SVOCs, thus lowering the escaping tendency of PAHs from the surface. The  $\text{Ln } f_a/f_s$  ratios for HCB, Phe, Ant, Flt and Cry, were significantly correlated with temperature (Fig. 6, Table S13, SI). Conversely, the lack of correlation of  $\text{Ln } f_a/f_s$  ratios for PCBs and HCHs with temperature can be explained by their simultaneous  $C_{SA}$  and  $C_A$  correlations with temperature and remarkable seasonal patterns, which induces a lack of seasonality in  $f_a/f_s$  ratios for these compounds.

#### 4. Conclusions

In the present study, the role of temperature in the ambient concentrations and the soil/snow-air partitioning of SVOCs was evaluated for a large number of organic chemicals representing a wide range of physical-chemical properties. On the one hand, the atmospheric

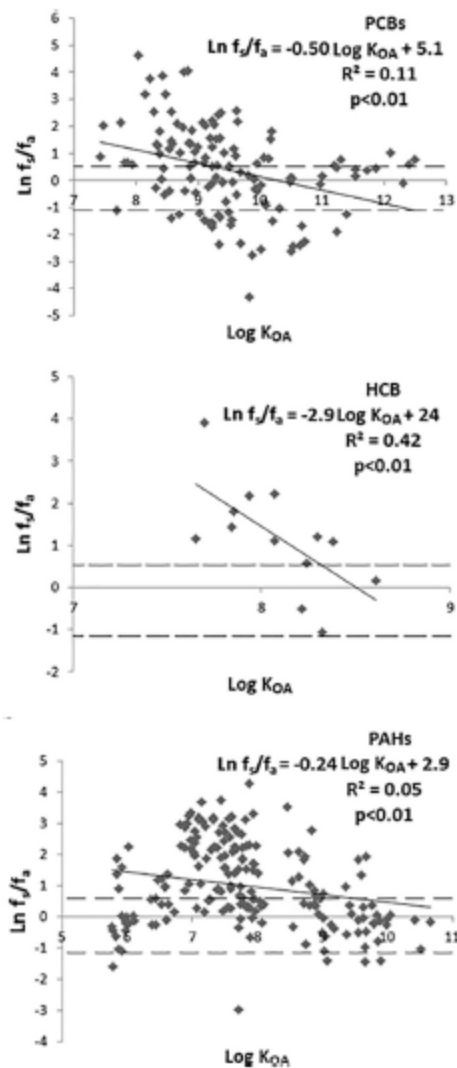


Fig. 5. Variation of the soil-air fugacity ratio ( $\text{Ln } f_a/f_s$ ) versus the octanol-air partition coefficient ( $K_{OA}$ ). The horizontal dashed lines mark ratios  $-1.2$  and  $0.53$ , interval for air-surface equilibrium.

concentrations ( $C_A$ ) and the concentrations in the gas phase equilibrated with the surface soil/snow ( $C_{SA}$ ) for PCBs and HCHs showed a clear seasonality correlated with temperature. In addition, soil/snow fugacity was correlated with air fugacity which constitutes evidences of the relevance of soils as dominant local sources of these SVOCs in the atmosphere, except for the higher MW PCBs. The more hydrophobic POPs were close to equilibrium or showed a net diffusive deposition. A close coupling between the atmospheric and soil/snow compartments for PCBs and HCHs in this arctic site has been confirmed during the study period as reflected by their  $f_a/f_s$  ratios (close to equilibrium conditions with a lack of a clear seasonality). On the other hand, PAHs and HCB showed a clear seasonality in  $f_a/f_s$  ratios with equilibrium conditions during winter months and net volatilization during the rest of the year, which constitutes evidence of the relevance of soils as secondary sources and highlights the role of temperature on the soil/snow-air exchange of SVOCs.

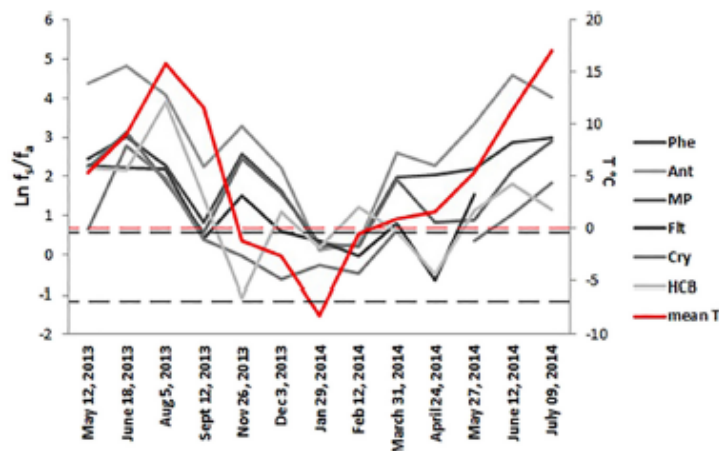


Fig. 6. Seasonality of the fugacity ratio ( $\ln f_s/f_a$ ) for HCB and some PAHs. The horizontal grey dashed lines mark ratios  $-1.2$  and  $0.53$ , the interval for air-surface equilibrium. The horizontal red dashed line marks  $0\text{ }^\circ\text{C}$ . Soil was snow covered from to November 2013 to April 2014. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.04.330>.

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