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Short communication

Persistent organic pollutants in the atmosphere of the Antarctic Plateau

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HIGHLIGHTS

- The occurrence of POPs in the Antarctic Plateau is demonstrated for the first time.
- The mass per sample of PCBs, HCHs and HCB show a minimum at the South Pole.
- POPs reach the Antarctic Plateau by atmospheric transport from the free troposphere.

GRAPHICAL ABSTRACT

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ABSTRACT

Persistent organic pollutants (POPs) bioaccumulate in biota, have long residence times in the environment, and potential for long range atmospheric transport. Here, we show the first measurements of legacy POPs in the atmosphere of the Antarctic Plateau from 73° South to the South Pole. Samples were taken using passive samplers. The amount of polychlorinated biphenyls (as $\sum_{26}$PCBs) per sample ranged from 0.8 ng to 26 ng. The mass per sample of hexachlorobenzene (HCB) and γ-hexachlorocyclohexane (γ-HCH) in the gas-phase ranged from 0.67 ng to 2.7 ng and from non-detected to 2.6 ng, respectively. The lowest amounts of POPs were observed at the South Pole. This work shows that POPs have also reached the remotest region of Earth from primary sources. The assessment of the air mass back trajectories and current knowledge of atmospheric circulation over the Antarctic continent suggests that POPs reach the Antarctic Plateau by subduction of air masses from the free troposphere.

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1. Introduction

Persistent organic pollutants (POPs) have the potential to bioaccumulate in organisms, to undergo long range transport reaching remote regions, and affect ecosystems (Lohmann et al., 2007; Nizzetto et al., 2010). The occurrence of POPs has previously been reported in the atmosphere, water and land for all Earth's regions except for most of the vast Antarctic Continent (Lohmann et al., 2007; Bengtson Nash, 2011; Galbán-Malagón et al., 2013a). Long range transport of semivolatile POPs occurs, generally, through successive volatilization and atmospheric deposition mediated by atmospheric transport, a process known as

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"grasshopping", with presumably oceanic transport being of minor importance (Wania and Mackay, 1996; Jurado and Dachs, 2008). The Antarctic continent is not only the remotest region from primary sources of POPs (Von Waldow et al., 2010), but it is also isolated from inputs of POPs from lower latitudes due to the dominant atmospheric and oceanic circulation patterns. Indeed, the Antarctic circumpolar current acts as a barrier limiting the oceanic transport of POPs to the Antarctic continent (Bengtson Nash et al., 2010). However, semivolatile POPs undergo long range atmospheric transport and can be deposited to remote regions, including locations south of the Antarctic circumpolar current (Risebrough et al., 1976; Galbán-Malagón et al., 2013a; 2013b; Bigot et al., 2016). Legacy and emerging POPs, such as polychlorinated biphenyls (PCBs), have been described in soils and vegetation (Borghini et al., 2005; Cabrerizo et al., 2012), the atmosphere (Baek et al., 2011; Kallenborn et al., 2013; Galbán-Malagón et al., 2013a; Gambaro et al., 2005), seawater (Galbán-Malagón et al., 2013b; Bigot et al., 2016), and biota (Risebrough et al., 1976; Miranda-Filho et al., 2007) from the maritime Antarctica. In the Southern Ocean, which is characterized by high primary productivity and phytoplankton biomass (Strutton et al., 2012), the biological pump effectively sequesters hydrophobic POPs from the atmosphere and surface waters by settling of organic matter bound-POPs to the deep ocean (Galbán-Malagón et al. 2012, 2013a). Therefore, even if POPs reach the Southern Ocean and its atmospheric boundary layer, their atmospheric and seawater concentrations will be reduced by oceanic sequestration (Galbán-Malagón et al., 2013a, 2013c).

The Antarctic continent is dominated by the katabatic winds (Bintanja et al., 2014) driven by the low temperatures found in the high elevation continental Antarctica. The average annual temperatures in the Antarctic Plateau are around − 50 °C (Jones and Lister, 2014). These air masses originate in the free troposphere and descend from the high elevation plateau to the Southern Ocean (Bintanja et al., 2014; Parish and Bromwich, 2007). The direction of the winds over the Antarctic continent is fairly constant, even during summer when the katabatic winds are weaker, due to the influence of the Antarctic orography (Parrish and Casano, 2003). Atmospheric circulation models show that transport of air masses from the coastal Antarctica boundary layer to the Antarctic plateau is unlikely as coastal air masses are too cold to rise to the Antarctic Plateau (Stohl and Sodemann, 2010). Therefore, subduction of air masses from the free trophosphere may transport POPs to the inner regions of the continent. However, the free troposphere has been largely unexplored in terms of POPs, except for measurements of POPs at mid-latitude mountains (Van Drooge et al., 2002; Lohmann et al., 2007; Wang et al., 2016), and few measurements from aircrafts (Knap and Binkley, 1991; Harner et al., 2005). Modelling exercises have shown that meridional transport and subduction of air masses from the free troposphere can contribute to the transport of POPs to the Arctic (Zhang et al., 2010), but the relevance of this process has not been previously assessed for Antarctica. The objective of this work is to report the first measurements of atmospheric POPs over the Antarctic Plateau.

2. Materials and methods

2.1. Sampling approach

Within the framework of the ACCIONA Wind Power expedition, a wind-driven sledge attached to a kite crossed the Antarctic Plateau (around 3000 m asl) in December 2011, covering 3100 km from Novolazarevskaya station (75 km from the coast) to Glaciar Union (Table S1 in supplementary material). During this transect, five atmospheric samples were taken for the analysis of POPs starting at 73°01' South, 05°24' East, until the South pole, and then finishing at 80°29' South, 79°55' West (Fig. 1, Table S1).

The sampling strategy was designed according to the limitations of performing sampling of atmospheric POPs from a wind powered sledge (Fig. S1), and by the likely low POP concentrations to be found in Antarctic air. High volume air samplers are the common approach used for sampling large volumes of air (Galbán-Malagón et al., 2013a), but this would have required electrical power which was not available during the expedition. The use of passive samplers is an alternative to overcome these constraints. Directional flow-through passive samplers (Xiao et al., 2007) allows for sampling large volumes of air within a time period of days with the high wind speeds found in Antarctica. We simplified the Xiao and coworkers' sampler design (Xiao et al., 2007) in order to gain in robustness. The sampler consisted in a horizontally aluminium tube of 60 cm length, 10 cm internal diameter, fixed onto one side of the wind powered sledge at approximately 1.5 m height (Fig. S1). The sampler contained a polyurethane foam (PUF), an anemometer and a temperature sensor connected to a data logger (Fig. S1). The PUFs diameter and length were of 10 cm. The PUF density was of 30 kg m^{-3}. This sampler allowed taking gas-phase samples of semivolatile organic compounds without the requirement of electrical power and with minimal work load for the expedition participants. Prior to the sampling campaign, all PUFs were precleaned with acetone-hexane 3:1 for 48 h, with the solvent exchanged every 24 h. Afterwards, PUFs were dried in a desiccator and stored in air-tight teflon bags. Before the shipping of the sampling equipment to the campaign, PUFs were fortified with PCB 65 and PCB 200 for their use as depuration compounds. Depuration compounds allow estimating the sampling rate (Pozo et al., 2004), and thus determining the effective air volumes provided these are depleted enough during the exposure time. In total, five samples (samples S1 to S5), covering the transect of 3500 km from Novolazarevskaya station (75 km from the coast) to Union Glacier, were taken and analysed (Fig. 1 and Table S1 show the location of

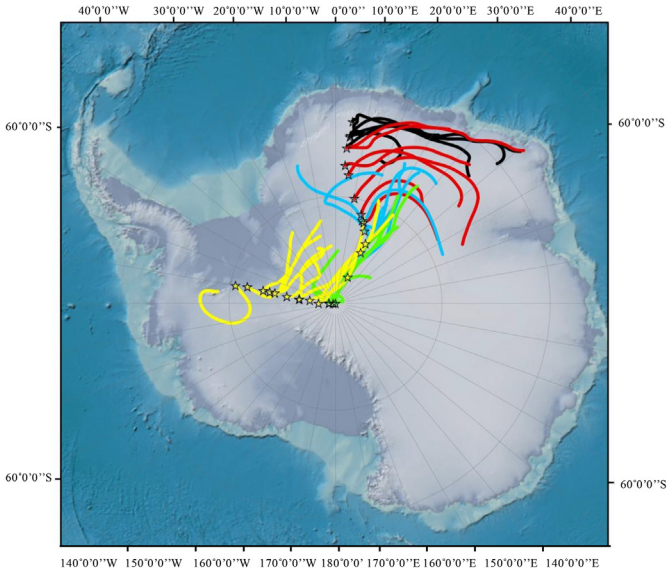

Fig. 1. Sampling transect in the Antarctic Plateau followed by the wind driven sledge from December 2011 until January 2012. One air mass back-trajectories (BT) is shown for every sampling day, the colour of the BT indicates to which one of the samples it contributed. Five samples were taken during transects as indicated in black (S1), red (S2), blue (S3), green (S4) and yellow (S5). The 48 h BT were estimated using the NOAAs Hysplit model at 25 m above ground level. Supplementary Material Fig. S2 shows the BT at 50 and 100 m above ground level. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

samples S1–S5 and their ancillary data). Samples were deployed for periods between 5 and 9 days integrating a transect of air above the Antarctic Plateau as shown in Fig. 1. After sampling, samples were kept into two teflon bags. In addition, blank samples were also included at a rate of one field blank for each field sample. Field blanks were deployed for less than one hour into the sampler, just before the sampling period of the corresponding field sample. Field blanks were kept in the same manner than field samples.

2.2. Sample extraction and quantification

All samples were Soxhlet extracted during 24 h with acetone:-hexane 3:1. Prior to the extraction, PCB 54 and PCB 155 were added as surrogate standards in order to control potential losses during the analytical procedure. The extracts were reduced in a rotary evaporator to 0.5 ml and cleaned and fractionated using 3 g of 3% deactivated alumina, eluted using 6 ml of hexane (Cabrerizo et al., 2012). The targeted POPs were PCBs, hexachlorobenzene (HCB) and hexachlorocyclohexanes (α- and γ- HCH). PCBs, HCB and HCHs were analysed by a gas chromatograph equipped with a μ-electron detector capture (GC-μ-ECD, Agilent Technologies, model 7890N) as reported elsewhere (Cabrerizo et al., 2012). The targeted PCB congeners were: tri-Chlorine PCB congeners 18, 17, 31, 28, 33; tetra-chlorine PCBs 52, 49, 44, 74, 70; penta-chlorine PCBs 95, 99/101, 87, 118; hexa-chlorine PCBs 110, 151, 149, 153, 132/105, 138, 158, 128, 169; hepta-chlorine PCB 187, 183, 177, 171/156, 180, 191, 170; octa-chlorine PCBs 201/199, 195, 194, 205; nona-PCB 206, 208, 209). PCB 30 and PCB142 were added before sample injection as internal standards for quantification.

2.3. QA/QC

All analytical procedures were monitored using strict quality assurance and quality control measures. Recoveries were routinely monitored using PCB 54 and PCB 155 and these were in the range between 60% and 114% for PCB 54, and between 70% and 95% for PCB 155. We corrected the levels by recoveries. One field blank was analysed for each one of the five field samples. Only 8 out of the 41 targeted congeners were detected in field blanks. PCB mass in samples are only reported when they were above the mean amount in blanks plus three times the standard deviation. Following this criteria, PCB 101 is not reported. Blank levels were at concentrations significantly below (less than 5%) the field levels for all other congeners. PCB 118 is not reported due to a co-elution. α-HCH was not detected in the field samples.

The anemometer deployed inside the directional air-sampler did not work properly during the campaign presumably due to the low ambient temperatures. Therefore, the use of depuration compounds was the only method left to determine the effective air volumes for the five samples. However, the recoveries of the depuration compounds were between 84 and 100% after correcting by surrogate recoveries. Therefore, the loss of depuration compounds was small or negligible during the sampling periods, and it was always lower than the recommended loss for their use for estimating the effective air volume. This is probably due to the very high PUF-Air partition constants at the average temperatures registered during the campaign, which ranged from −29 °C to −23 °C. Therefore, we do not provide the gas phase concentrations as the effective sampled volumes could not be estimated. We report the mass of the targeted POPs per sample.

3. Results and discussion

The mass of PCBs per sample ($\sum PCB_{26}$) ranged from 27 ng/sample in the outer part of the Plateau (Station S1, Fig. 1), to a minimum in the South Pole of 0.8 ng (Fig. 2). For HCB, these ranged from 0.67 ng/sample to 2.7 ng/sample, with maximum concentrations in the outer region of the plateau. There are not known effective degradation mechanisms of HCB in the atmosphere and seawater, and thus HCB is a paradigm of persistent organic pollutant (Galbán-Malagón et al., 2013c). HCB concentrations are fairly constant in the atmosphere of the Southern Ocean and maritime Antarctica, ranging from 10 to 30 pg m^{-3} (Baek et al., 2011; Kallenborn et al., 2013; Galbán-Malagón et al., 2013; Bigot et al., 2016). The effective sampling air volume of the Plateau's samples would be of about 90 m^3 if we assume a concentration of 15 pg m^{-3} for HCB. Presumably, the effective sampling volumes for the more hydrophobic PCBs would be much larger since they have higher affinity for the PUF used as passive sampler, and thus larger breakthrough volumes. The mass of γ-HCH ranged from below

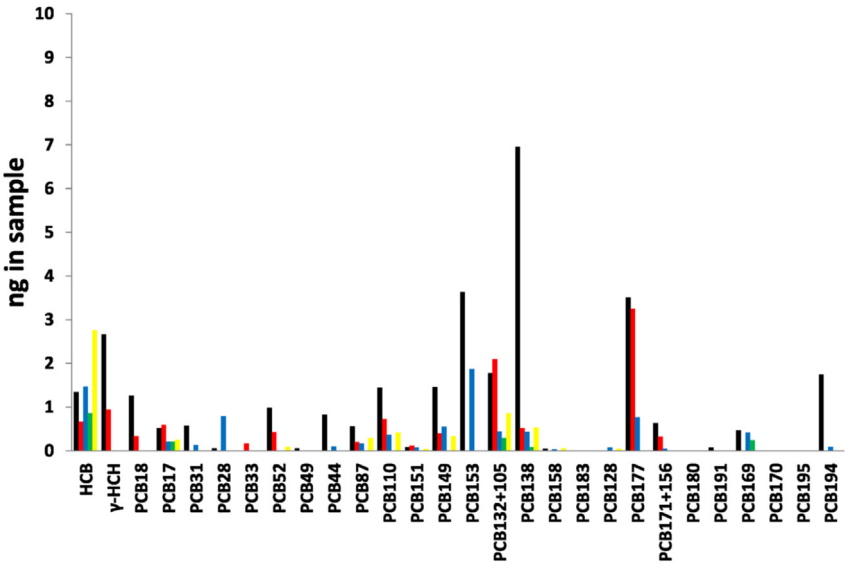

Fig. 2. Mass per sample (ng $sample^{-1}$) of individual PCBs congeners, HCB and γ-HCH in the five atmospheric samples from the Antarctic plateau. Samples S1 to S5 have the same colour code than in Fig. 1. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

detection limit in the central plateau and South Pole, to 2.7 ng/sample in the outer plateau (Fig. 2).

The air mass back trajectories during the sampling events followed the general pattern of the katabatic winds (Parish and Bromwich, 2007) during most of the sampling periods (Fig. 1). The air masses originate at elevations between several hundred to thousands of meters above ground level from high elevation regions of Antarctica (Figs. S3 and S4), and descended to the Antarctic Plateau. This indicates that the air masses sampled during the campaign originated in the free troposphere as the boundary layer over the Antarctic plateau is thinner than in other world regions, consistent with current knowledge of atmospheric circulation over the Antarctic continent (Parish and Cassano, 2003; Parish and Bromwich, 2007; Stohl and Sodemann, 2010). POPs may have reached the free troposphere in other world regions as part of the general meridional transport of air masses from low/mid latitudes to the poles (Parish and Bromwich, 2007; Paulis et al., 2008).

It has been previously suggested that "grashopping" is the main mechanisms for POP transport to the Polar Regions (Wania and Mackay, 1996; Jurado and Dachs, 2008). The results presented here are consistent with a "high altitude – grasshopping" of POPs, mediated by the free troposphere and occurring over long distances. The influence of atmospheric transport of POPs from the free troposphere has been previously being documented for other high elevation regions (Van Drooge et al., 2002; Lohmann et al., 2007; Wang et al., 2016).

The occurrence and pattern of POPs in the atmosphere of the Antarctic Plateau may reflect the POPs in the southern hemisphere free troposphere, which is largely unexplored in terms of POPs. Samples 2, 3 and 4 were taken at different locations of the same air mass (Fig. 1), and thus reflected the variation of POPs mass during atmospheric transport over the Antarctic Plateau (at 1.5 m height). A decrease of mass is observed when going from the outer section of the plateau to the South Pole (Fig. 2). In fact, PCB levels for many congeners were below detection limit in the South Pole. The processes driving the decrease of concentrations are probably a combination of i) degradation of PCB congeners and γ-HCH due to reaction with OH radical which have been reported to be abundant in the Antarctic's Plateau's atmosphere (Mauldin et al., 2004, 2010), and ii) exchange of PCBs with the surface ice/snow during the atmospheric transport. The pattern of PCBs is especially depleted in the less chlorinated congeners, such as congeners 18, 28, 52, 110, when compared with other gas phase patterns measured at lower latitudes such as around the Antarctic Peninsula (Galbán-Malagón et al., 2013). This depletion would be consistent with PCBs reacting with OH radicals, which affects mainly the congeners with fewer chlorines (Anderson and Hites, 1996). However, the decrease in concentrations is also observed for other congeners. Measurements of snow-air exchange performed at Livingston Island (Southern Shetlands, Antarctica) indicated close air-snow equilibrium to a net volatilization of PCBs from snow (Cabrerizo et al., 2013). However, the temperatures at the Antarctic Plateau are 25–30 °C lower than at the Southern Shetland Islands during the austral summer. It is feasible that there was a decrease of the atmospheric concentrations due to a net deposition of PCBs, HCB, and γ-HCH to ice during transport. On the long term, this net deposition would lead to an accumulation of POPs in the Antarctic Plateau.

Even though the occurrence of POPs in the maritime Antarctica was identified in landmark pioneering studies four decades ago (Risebrough et al., 1976), the present study is the first to report the occurrence of POPs in the atmosphere from the South Pole and Antarctic Plateau. This also confirms that POPs pollution is truly global, since POPs are present not only in the Antarctic Plateau, but also in the global free troposphere. This work shows that within the scientific challenge to monitor and understand the global occurrence and transport routes of POPs, Antarctica serves as a sentinel of global pollution.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2016.11.015.

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