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Polychlorinated biphenyls (PCBs) in the atmosphere of sub-alpine northern Italy

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Advection of air masses and re-volatilization from local sources play a dominant role as drivers of the PCB atmospheric concentration in sub-alpine northern Italy.

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ABSTRACT

The main objective of this work was to assess the atmospheric concentrations and seasonal variations of selected POPs in a sub-alpine location where few data are available. A monitoring and research station was set up at the JRC Ispra EMEP site (Italy). We present and discuss a one-year data set (2005–2006) on PCB air concentrations. ∑7PCBs monthly averaged concentration varied from 31 to 76 pg m^{-3}. Concentrations in the gas phase (21–72 pg m^{-3}) were higher than those in the particulate phase (3–10 pg m^{-3}). Advection of air masses and re-volatilization from local sources seem to play a dominant role as drivers of PCB atmospheric concentrations in the area. Indications of seasonal variation affecting PCB congener patterns and the gas/particulate partitioning were found. Modeling calculations suggest a predominant importance of the wet deposition in this region (1 μg m^{-2} yr^{-1} ∑7PCBs yearly total wet deposition flux; 650–2400 pg L^{-1} rainwater concentrations).

1. Introduction

Atmospheric transport and deposition are important mechanisms determining the fate of many persistent organic pollutants (POPs) in the environment. Due to atmospheric transport these pollutants can be moved from sources to distant locations and therefore contaminate remote terrestrial and aquatic environments. Research has been carried out in the last decades on ambient concentrations and controls of numerous POPs families. Experimental measurements alone (active and passive sampling) or combined with model estimations have been used to study the POPs environmental processing, transport and behavior in specific regions (Stern et al., 1997; Lohmann et al., 2001; Hung et al., 2005a,b; Shen et al., 2005; Pozo et al., 2006; Cleverly et al., 2007; Klánová et al., 2007; Dvorska et al., 2008). However, the global distribution of these pollutants, the difficulties in their sampling and analyses and the associated high economic cost of those procedures make it complicated to obtain experimental data from all areas of interest. In addition, the current scarcity of consistent experimental data sets on POPs concentrations poses a problem for proper validation of models. Data from "independent sampling stations" or sampling campaigns at a local or regional scale and from national or international monitoring networks, at more global scales (e.g. AMAP, EMEP, TOMPS in the Arctic region and Europe and NJADN and CBADS in North America, among others) are commonly used for POPs model validation (Fig. S1, supporting material). However, there are many "non-monitored areas" and for numerous zones no information on POPs ambient levels is available. Such is the case for instance with Southern–Western Europe that remains uncovered by the EMEP POPs monitoring network.

The present work focuses on the atmospheric measurements of polychlorinated biphenyls (PCBs) conducted on the east shore of Lake Maggiore, Lombardy, northern Italy. PCBs are a POPs family of anthropogenic origin constituted by 209 congeners that enter the environment as a result of primary and secondary sources. PCBs bioaccumulate and have been identified as toxic compounds able of producing a wide spectrum of adverse health effects in biota and humans such as disruption of the endocrine system and carcinogenicity, among others (Safe, 1984, 1990). Since the mid-1970s these chemicals have been removed from active use in most countries but they are still present in almost all environmental compartments. Few data are available on PCB atmospheric concentrations in sub-alpine northern Italy and adjacent areas. Passive and active sampling air measurements of PCB have been conducted along the Lys Valley, Aosta Valley (Jaward et al., 2005), and PCB gas phase concentrations are available for Venice Lagoon,

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Veneto (Manodori et al., 2006). Those places are located ~60 km to the west and ~300 km to the east, respectively, from our sampling site (see Section 2.1.). Dioxin-like and indicator PCB atmospheric concentrations (air and particulate phases) have been recently reported for one week of sampling in the Lake Maggiore area (Vives et al., 2007; Castro-Jiménez et al., 2008). However, no long-term study has ever been conduced in this area.

Our main objective was to initiate systematic (weekly) POPs atmospheric measurements (gas and particulate phases) in order to assess the atmospheric concentrations and seasonal variations of selected POPs in a sub-alpine location in one of the "non-monitored areas". To achieve this goal, a monitoring and research station was set up at the Joint Research Center (JRC) Ispra EMEP site, Italy. The generated data sets will be incorporated in the current monitoring network databases and used for model development and validation. Regular monitoring activities started on April 2005. A data set on PCBs' (CB-28, -52, -101, -118, -153, -138 and -180) air concentrations obtained for the period April 2005–2006 is presented in this work. Modeling estimations of the wet and dry deposition fluxes of PCBs, based on the obtained experimental data, for the same period are also presented and discussed in terms of their seasonality.

2. Experimental

2.1. Sampling location description

Air samples were collected at the EMEP monitoring and research station at the JRC Ispra Site. The sampling site is located in a semi-rural area by the eastern shore of the sub-alpine Lake Maggiore, in the Varese province (Lombardy), northern Italy (45°49′N, 8°38′E, 209 m a.s.l.) (Fig. 1). Although the Varese province is one of the most industrialized districts in Italy with a relatively elevated population for a semi-rural area, the station is several tens of km away from large emission sources like intense road traffic or big factories. Milan (1 300 000 population), 60 km to the south-east; Novara (100 000 population), 40 km south; Varese (82 000 population), 20 km east; and Gallarate – Busto Arsizio (130 000 population), about 20 km south-east, are the main urban nucleus around. Due to the topography and its location, this region is characterized by low wind speed occasionally interrupted by North-Föehn events, where relatively warm and dry downslope wind from the Alps flows over the area (Rodríguez et al., 2005).

2.2. Air sampling

Integrated weekly samples were collected from April 2005 to April 2006 using high volume samplers (Echo PUF high volume sampler, TCR Tecora, Milan, Italy). Two air samplers were set up and used interchangeably to gather samples throughout the one-year period. Air particle phase was retained by using a 102 mm diameter quartz fiber filter (QFF) (QM-A type, particle retention: 2.2 μm, Whatman International Ltd., Brentford, Middlesex, UK), whereas the gas phase was trapped with a polyurethane foam (PUF) plug of 65 mm diameter, 75 mm length and 0.22 g cm^{-3} in density from Tisch Environmental, Inc. (Cleves, Ohio, U.S.). An average flow of 87 ± 2 L/min was achieved during sampling intervals gathering an averaged volume of 860 ± 27 m^3 for each sample. All details of sampling intervals are presented in Table S1 (supporting material). Atmospheric temperature and pressure were recorded at the site. In addition, temperature, precipitation, relative humidity and wind direction and speed for the sampling period were obtained from the Ranco meteorological station, located 4 km southwest from the EMEP station.

2.3. Analytical determinations

QFFs and PUFs were soxhlet extracted separately with *n*-hexane/acetone (220:30 v/v) for 48 h after being spiked with ^{13}C-labelled internal standards (PCBs 28, 52, 101, 118, 153, 138 and 180). All organic solvents were dioxin analysis grade from Sigma–Aldrich (Buchs SG, Switzerland). PUFs were directly loaded into the soxhlet, whereas filters were first placed in extraction thimbles and then loaded into the soxhlet. Extract purification was executed with an automated clean-up system (Power-Prep P6; Fluid Management Systems, Inc., Watertown, MA, USA). The purification method was previously described by Abad et al. (2000). Purified extracts were evaporated to ~0.5 ml under low nitrogen flow by using an automatic

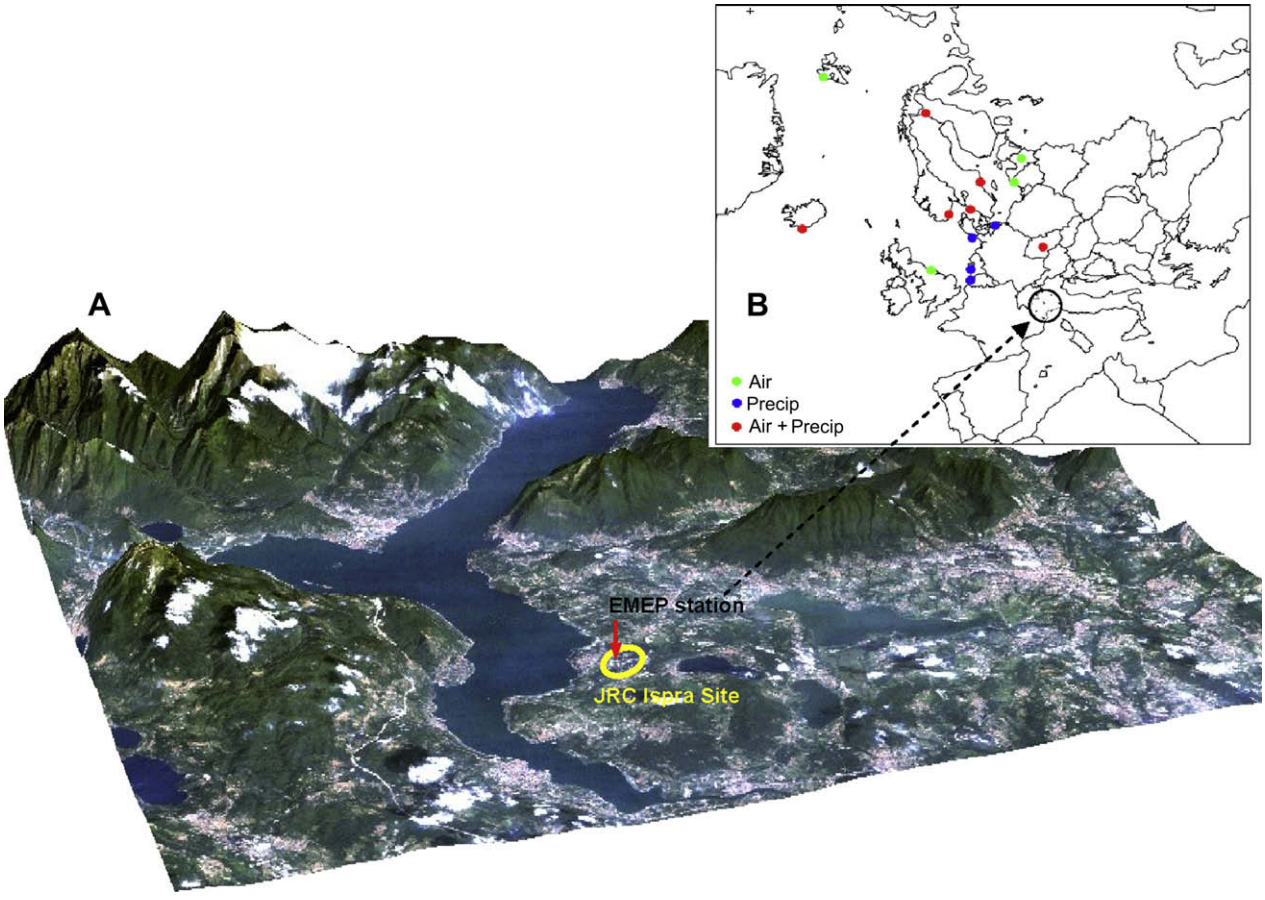

Fig. 1. Sampling site location (A). EMEP POPs monitoring sites in 2005 (from EMEP Report, 2007) and location of the Ispra sampling site in the current uncovered area (B).