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Deposition of PCBs in mountains: The forest filter effect of different forest ecosystem types

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Abstract

The effect of canopy composition and density on the forest filter effect (the deposition flux under the canopy and deposition to bare soil) for polychlorinated biphenyls (PCBs) was investigated for forests of the Italian Alps. Deposition fluxes were measured in situ using deposition samplers below canopies and in adjacent clearings at three altitudes (1100, 1400, and 1800 m above sea level). Forest sites were selected on the basis of canopy composition and density. Net forest fluxes (NFFs) were calculated by subtracting the deposition flux under canopies by deposition fluxes to clearings and represent the net contribution of forests to PCB deposition. NFF trends are discussed in relation to canopy development. Mean deposition velocities were also calculated and a direct correlation with the/octanol air partition coefficient (K_{OA}) was found. A leaf area index (LAI) was used to calculate a specific, canopy density-independent, deposition velocity for each forest type. This parameter can be used to calculate the deposition of PCBs in forests, given their LAI. Results show that forests significantly enhance the deposition of PCBs to soil and that this effect is controlled by temperature, leaf dynamics, and K_{OA}.

Keywords: PCBs; POPs; Deposition; Mountains; Alps; Forest filter effect; Organic; Carbon; LAI; K_{OA}

1. Introduction

Atmospheric transport and deposition to soils and vegetation are key factors for the study of environmental exposure to persistent organic pollutants (POPs). Methods to evaluate atmospheric deposition fluxes by both direct and indirect measurements have been developed and applied in different scenarios (Strachan and Eisenreich, 1988; Galassi et al., 1993; Gatz et al., 1994; Gregor et al., 1996; Jurado et al., 2004). The uptake of POPs by vegetation is a relevant step by which pollutants enter the terrestrial food web (McLachlan, 1996; Gouin et al., 2002). Forests, in particular, may strongly influence deposition fluxes to the ground (Horstmann and McLachlan, 1996; Brorström-Lundén and Löfgren, 1998). The capacity of leaves to accumulate organic pollutants (Bacci et al., 1990; Reischl et al., 1987; Umlauf et al., 1994; Di Guardo et al., 2003), the extent of organic surface area, together with high soil organic matter content, and infrequent biomass harvesting create the conditions for forests to be an important storage or "retardation" compartment for POPs in the terrestrial environment.

A model known as the "forest filter effect (FFE)" proposed by McLachlan and Horstmann (1998), predicted that the average annual deposition of some POPs on forested soils is approximately three times greater than that on bare soil. In this model, forest canopies are seen as cumulative compartments (with lipid-soluble POPs accumulating on leaf surfaces and in leaf cuticular waxes) able to redirect POPs to the soil following different processes: rain washout, wax erosion, and transport due to litter fall. The model is expressed as a function of K_{OA} and K_{AW} (*n*-octanol/air and air/water partition coefficients, respectively) and predicts a low

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filter effect for compounds with $\log K_{OA} < 7$ and $\log K_{AW} < -6$ and pronounced effects for compounds with $7 < \log K_{OA} < 11$ and $\log K_{AW} > 6$. Few experimental data are currently available to better characterize the filter effect (Horstmann and McLachlan, 1996, 1997a, b, 1998). Considering the large percentage of the planet surface involved, the global role of forests in the fate of semivolatile organic compounds is receiving greater attention (Wania and McLachlan, 2001; Wegmann et al., 2004). Considerable uncertainties still exist concerning the variability of the FFE for different ecological scenarios, given the few experimental data available. This is particularly true for mountains, where different forest types are generally present at different altitudes.

In this study, the FFE was estimated for certain organochlorine compounds using deposimeters in forests and clearing sites along an altitudinal gradient (from 1000 to 1800 m above sea level (a.s.l.)) in the Italian Western Alps. These forests are characterized by different compositions, densities, and leaf phenology. In the alpine scenario, environmental conditions could determine the presence of contamination hotspots due to enhanced deposition (Daly and Wania, 2004). The main goal of this study is to evaluate the deposition of polychlorinated biphenyls (PCBs) in an alpine scenario and, more specifically, to characterize the role played by different forest types in deposition fluxes to soils.

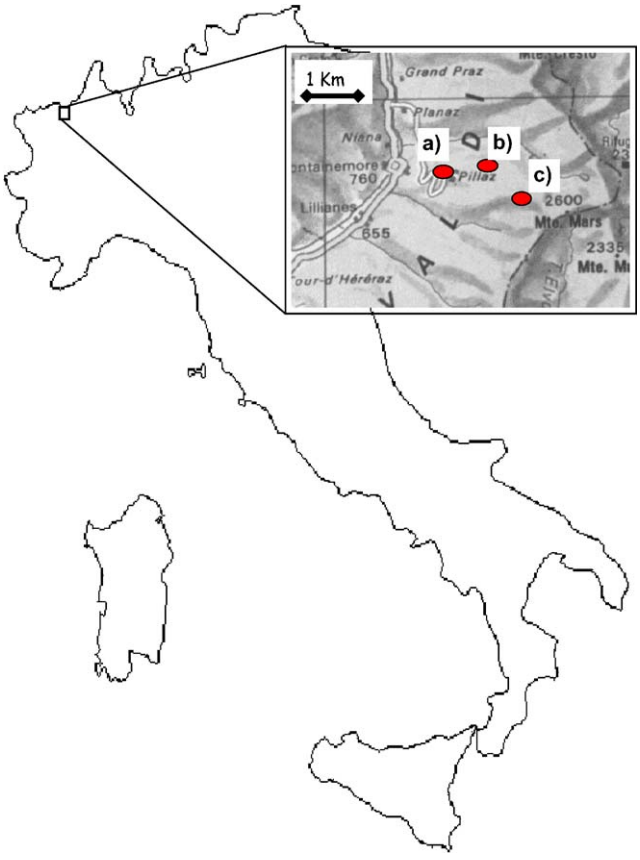

Fig. 1. Geographical location of sampling sites: (a) 1100, (b) 1400, and (c) 1800 m a.s.l.

2. Materials and methods

2.1. Reagents

All solvents used were pesticide residue grade. Acetone and *n*-hexane were purchased from Fluka, Buchs, Switzerland. Methanol, dichloromethane, toluene, and chlorotrimethylsilane were purchased from Sigma–Aldrich, Seelze, Germany. Anhydrous sodium sulfate and florisil for residue analysis (60–100 mesh) were purchased from Merck, Darmstad, Germany. Empore extraction disks (C-18, 47 mm) came from 3M, St. Paul, MN, USA, and gel permeation SX-3 beads (200–400 mesh) were purchased from Bio-Rad, Hercules, CA, USA. PCB single-congener analytical standards were purchased from AccuStandard, New Haven, CT, USA. Purities were above 98%.

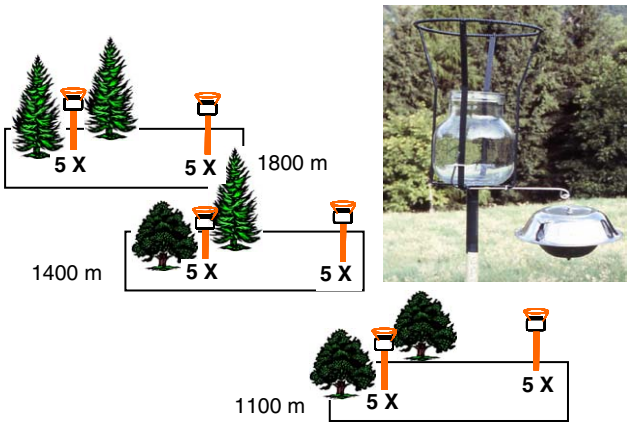

Fig. 2. Schematic representation of the experimental design.

2.2. Sampling sites

Sampling was performed in 2003 at six sites (three forest sites plus three adjacent clearings) in the Lys Valley, Aosta, Italy, situated at 1100, 1400, and 1800 m a.s.l. in an alpine region (Figs. 1 and 2). These sites are not directly affected by local sources of contamination (Jaward et al., 2005). The Lys Valley is in the Western Alps and presents a North–South orientation, with the main peak (Mount Rosa, 4634 m a.s.l.) at the northern extremity. All sampling sites were located on the east side of the valley, to normalize ecological conditions such as sun exposure and wind path. Deposition samplers within forest sites were located at least 100 m from the margins of the forest. Clearing sites were selected in the vicinity of the corresponding forest site, and samplers were located not less than 100 m from the forest margin to avoid possible interference due to the