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A comparison of Alpine emissions to forest soil and spruce needle loads for persistent organic pollutants (POPs)

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The loads of POPs in the Alps are higher than their emissions in the Alpine region.

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ABSTRACT

The project MONARPOP analysed the concentrations of semivolatile organic compounds (SVOCs) in two important sink compartments, needles of Norway spruce (*Picea abies* [L.] Karst.) and forest soil from 40 remote Alpine forest sites in Austria, Germany, Italy, Slovenia and Switzerland.

In the present study the load of PCDD/F, PCB, PBDE, PAH, HCB, HCH and DDT in the Alps calculated on the basis of measured data are compared with their estimated emissions in the Alpine region. It comes out that the masses of the studied pollutants stored in the forests are higher than the corresponding emissions in the Alpine area indicating that the Alps are a sink for POPs advected from surrounding areas. It is assumed that local emissions of PCDD/F and PAH deriving from biomass burning are probably underestimated and that the pool of these pollutants in the forests represents the accumulation over some decades.

1. Introduction

Forests play an important role in regulating the atmospheric concentrations and the transport of persistent organic pollutants (POPs) in the environment and are important sinks for semivolatile organic compounds (SVOCs). These ecosystems contain up to 80% of the aboveground carbon in the world so that their interactions with air-borne lipophilic substances like POPs influence the environmental fate of these pollutants at a global scale (Nizzetto et al., 2007).

Deposition of POPs from air to the terrestrial environment is higher in forested areas than in other kinds of natural or anthropogenic environments (Wania and Mclachlan, 2001). The last is in part the result of the greater roughness elements of the woodland landscape which enhance downward fluxes of both gaseous and particle-bound POPs (Howsam et al., 2001).

Mountainous areas in general, and the Alps in particular, are geographical and meteorological traps for atmospheric pollutants as a consequence of barrier effects, high precipitation, and low ambient temperature (Weiss et al., 2007). Considering that forests are the prevailing ecosystem type in the Alps, approximately 50% of the area (7.5 Mio. ha) is covered by them, it follows that they play a major role in the cycle of the pollutants in this region.

The present study, carried out within the framework of the project MONARPOP, has analysed two important sink compartments for atmospheric pollutants, needles of Norway spruce (*Picea abies* [L.] Karst.) and forest soil of remote Alpine forest sites in Austria, Germany, Italy, Slovenia and Switzerland, for the concentrations of SVOCs.

The present contribution compares coarse estimates of pollutant masses in the Alpine forests with their emission in the Alpine region of the investigated countries.

The objectives of this research are to make an indirect estimation of the SVOCs masses emitted in the Alpine area, to compare the

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emitted pollutant masses with their load in the Alps, and to assess whether the masses present in the Alpine ecosystems are consistent with the emissions in the area or are the result of other processes like long-range transport, accumulation, etc.

2. Methodological approach

The study area includes the Alpine regions (as defined by the Alps Convention) of Austria, Germany, Italy, Slovenia and Switzerland representing an area of approx. 150.000 km^2.

Samples of humus, mineral soil and half year old Norway spruce needles were collected in a monitoring network consisting of 40 sites located in remote areas at an altitudinal range between 1200 and 1400 m a.s.l (Fig. 1).

The concentration of PCDD/F, PAH, PCB, HCB, HCH, DDT, and PBDE among others were determined on the three mentioned matrices. More details on sample collection and chemical analyses are available in (Knoth et al., 2006; Belis et al., 2007; Offenthaler et al., 2008).

For concentrations below the detection limit zero was used. The pollutant masses per hectare in humus and the uppermost 10 cm of mineral soil of each site were upscaled from sample volume (humus: measured depth and defined cross section, mineral soil: cores of defined depth and cross-section), sample dry mass and the dry mass concentrations of pollutants. Forest areas in the Alpine regions of the individual countries were taken from the investigated countries' national forest inventories (NFIs: BFW, 2007; LWF, 2007; Ministry of Agriculture, Food and Forestry, 2007; SFI, 2007; WSL, 2007a). The green crown biomass of the forests was obtained for Switzerland from the Swiss NFI (WSL, 2007b) or, for the other countries, estimated from results of various forest ecosystem studies from representative ecological regions. According to a compilation of such studies (Ellenberg et al., 1986; Reichle, 1981) the following figures were used: 17.5 tons (t) needle mass per ha for Norway spruce forest and 3.5 t ha^{-1} needle and leave mass for all other tree species.

The emission data (PCDD/F, PAH) were taken from the national emission reports under the UN "Convention on Long Range Transboundary Air Pollution" (Vestreng et al., 2006). Since emissions of dioxins and furans available in literature are expressed only in toxic equivalents (TEQ) the same unit had to be used in the present study. Emission and masses are reported for only four PAH: benzo(a)pyrene (BaP), benzo(k)fluoranthene (BkF), benzo(b)-fluoranthene (BbF) and indeno(c,d)pyrene (IcdP). PCB concentrations in this study are presented as the sum of 6 congeners (#28, #52, #101, #138, #153, #180), while HCH includes α, β, γ, δ and ϵ isomers. PBDE is the sum of 8 significant congeners in the three commercial formulations pentaBDE (#28, #47, #99, #100, #153, #154), octaBDE (#183) and decaBDE (#209) and DDT is the sum of p,p'- and o,p'-DDT, DDD and DDE (also referred to as DDX).

The loads of pollutants in the soil of the Alpine forests (L_{afs}) were calculated as follows

$$L_{afs} = A_c.Sm.C_s \quad (1)$$

Where, A_c is the national Alpine area, Sm the soil mass per area and C_s the average concentration of the pollutant in the humus and in the mineral soil.

The loads of pollutants in the Alpine forest green biomass (L_{afb}) were calculated with the equation

$$L_{afb} = A_c.Gb.C_n \quad (2)$$

where Gb is the green biomass per area and C_n is the average concentration of the pollutant in the needles.

For the emissions we firstly calculated the per capita emissions (E_{pc}) on the basis of the national emission data. The national emissions in the Alps E_{ac} were estimated using

$$E_{ac} = E_{pc}.P_{ac} \quad (3)$$

Where P_{ac} is the population in the Alps for each country taken from Ruffini et al. (2005).

These estimates according to the equations (1)–(3) were done individually for each country and their sum ("sum approach") represents the result for the entire Alpine region (except France which was not covered by MONARPOP).

To appraise the robustness and uncertainty of these figures, a second way of estimating was carried out. In the "mean approach" the same equations were used considering the five countries and all their Alpine area as one single district (the estimates were carried out on the basis of overall means of the MONARPOP concentrations and the Alpine emissions).

A reliable estimate of the emissions requires complete and updated data. EMEP provides the most recent official dataset on national emissions for almost all of the studied pollutants in Europe

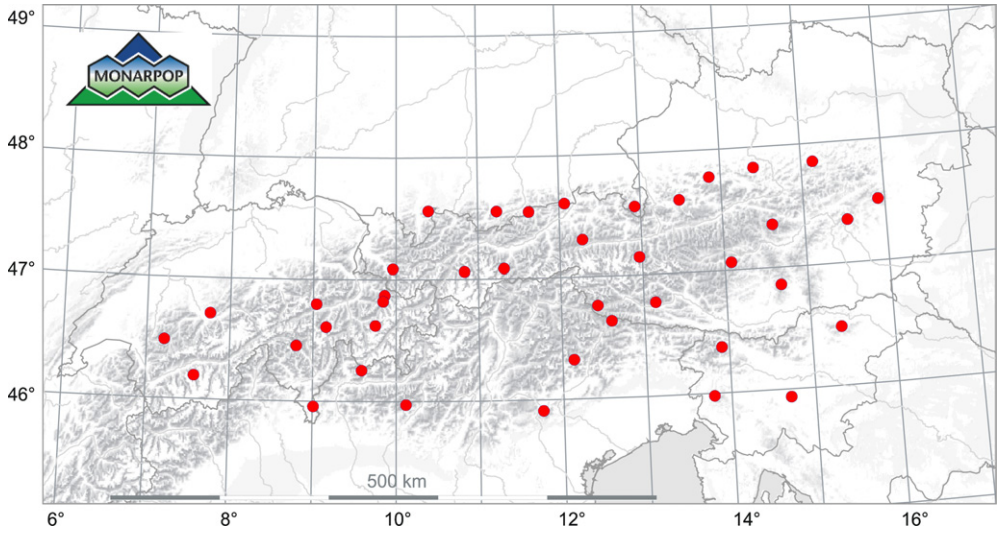

Fig. 1. Location of the 40 sites of the MONARPOP monitoring network.