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.

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.

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
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 km².

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 (Lₐₙₛ) were calculated as follows

\[
L_{\text{afs}} = A_{\text{c}} \cdot S_{\text{m}} \cdot C_{\text{s}} \quad (1)
\]

Where, \(A_{\text{c}}\) is the national Alpine area, \(S_{\text{m}}\) the soil mass per area and \(C_{\text{s}}\) the average concentration of the pollutant in the humus and in the mineral soil.

The loads of pollutants in the Alpine forest green biomass (Lₐₙ₇) were calculated with the equation

\[
L_{\text{afb}} = A_{\text{c}} \cdot G_{\text{b}} \cdot C_{\text{n}} \quad (2)
\]

where \(G_{\text{b}}\) is the green biomass per area and \(C_{\text{n}}\) is the average concentration of the pollutant in the needles.

For the emissions we firstly calculated the per capita emissions (Eₚₜₜ) on the basis of the national emission data. The national emissions in the Alps Eₐₙ were estimated using

\[
E_{\text{ac}} = \frac{E_{\text{pc}}}{P_{\text{ac}}} \quad (3)
\]

Where \(P_{\text{ac}}\) 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.
in 2000 (Vestreng et al., 2006). Accordingly, an estimate of the emissions in the Alps was made considering 2000 as reference year. However, since sampling of soils and needles was performed in 2004 and considering that some relevant limitations in the use and production of the studied pollutants were implemented after 2000, some discrepancies between estimated and effective emissions may arise. In order to better assess the actual emissions in the year in which the samples were collected, emissions in 2004 were estimated for those SVOCs for which concurrent national emission data were available (PCDD/F, PAH, HCB and HCH), provided by MSC-E (Gusev et al., 2007).

3. Results

Bearing in mind the various uncertainties of these estimates, both the pollutant masses in the forest ecosystems and the emissions should rather be considered an estimate of magnitude. The uncertainties include, e.g., inferring from half-year-old needles to all needle ages, Norway spruce forest to other forest ecosystems, remote sites to all Alpine forest sites, but also the limited number of sites, or the use of average national per capita emissions for the subarea of the Alpine region. Given these limitations and the methods used, pollutant masses in the forests will likely be underestimated, because pollutant concentrations are those from remote sites. At the same time, emissions in the Alpine region will likely be overestimated, because local per capita emissions in the less populated and industrialised Alpine areas can be expected to be lower than the nationwide average.

Nevertheless, the total of the country-specific estimates of pollutant masses (approach “sum”) does not deviate considerably from the estimate based on average values for the whole study region (approach “mean”) although the share of the Alpine area varies significantly among countries (Table 1 and Fig. 2). This indicates a rather robust estimate of magnitude.

The PCDD/F masses in the green crown biomass and soil of forests of the Austrian, German (Bavarian), Italian, Slovenian and Swiss part of the Alpine region are either 7 kg TEQ (“sum” estimate) or 12 kg TEQ (“mean” estimate, Fig. 3). These PCDD/F masses expressed in TEQ are two orders of magnitude higher than the estimated emissions in this region in 2004 (Fig. 2). Austria and Italy are the countries with the highest share of the Alpine emissions and loads, which is in line with their higher share, in terms of both area and population, of the Alpine region (Table 1).

For PAH, estimated pollutant stocks in forests are 469 t (“sum” estimate) or 792 t (“mean” estimate). These figures are one order of magnitude higher than the corresponding emission estimates. Austria and Italy are the countries with the highest share of the emissions in the Alps but the highest loads are those in the Austrian and Swiss forest ecosystems of the Alps (Figs. 2 and 3).

For both groups of compounds the pollutant stocks accumulated in needles/leaves are negligibly small (clearly below 1/1000) compared to forest soil.

The loads of PCB in the Alpine forests fall between 7.0 and 7.5 t “mean” and “sum” estimates respectively, while emissions in 2000

<table>
<thead>
<tr>
<th>Population in the Alps</th>
<th>Area in the Alps (ha)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Austria</td>
<td>3,250,400</td>
</tr>
<tr>
<td>Germany</td>
<td>1,373,600</td>
</tr>
<tr>
<td>Italy</td>
<td>4,093,600</td>
</tr>
<tr>
<td>Slovenia</td>
<td>639,200</td>
</tr>
<tr>
<td>Switzerland</td>
<td>1,740,800</td>
</tr>
</tbody>
</table>

Table 1

Area and population in the Alpine region (Ruffini et al., 2005).

Fig. 2. Estimation of the emissions in the Alpine area of each country and in the Alps using two techniques. The “sum” approach considers the Alpine area of each country while the “mean” approach considers the whole Alpine area as a single district. AUT: Austria; DEU: Germany; ITA: Italy, SVN: Slovenia, CHE: Switzerland (ISO alpha 3 codes).
are estimated to range between 1.5 and 2.5 tons. PCB stocks in crown biomass are two orders of magnitude lower than those in the soil. The highest estimated emissions of PCB in 2000 are those in the German and Austrian part of the Alps while the highest loads were calculated for the Italian and Austrian forest ecosystems of the Alps.

The estimated mass of PBDE present in the Alpine forests is between 1.5 and 3.2 t ("mean" and "sum", respectively, which is one order of magnitude higher than the estimated emissions in 2000. As for PCB, PBDE stocks in the needles are 100 times lower than those observed in the soil. Italy and Austria are the most important emitters of PBDE in the Alpine region and Italy is the country with the highest loads in its Alpine territory.

The estimated HCB loads in needles and soils of Alpine forests are 1.5–3.2 t ("sum" vs. "mean"), which is two orders of magnitude higher than the estimated emissions in 2004. Italy is the country with the highest emissions while the highest loads are those calculated for Austria and Italy.

The only country still officially emitting HCH in 2000 is Italy. Therefore, the HCH emission data in the Alpine region are supposed to represent the estimated Italian emission in this region only. The interpretation of data regarding this pollutant is, however, difficult since the estimations of emissions in literature are quite variable. Depending on the authors (Pacyna et al., 1999; Denier van der Gon et al., 2006) the estimated emissions in the Alps vary from 0.16 to 10 t a⁻¹ while the load in the Alpine forests ranges between 4.5 and 4.7 t. Moreover, HCH stocks in Alpine forest ecosystems in 2004 still reach almost the same order of magnitude as the annual emissions during the lastest years of use. Austria reported 2.3 t and Germany 14.5 t national HCH emissions in 1997, their last year of use (EEA, 2007). According to the estimation method adopted in the present study this corresponds to 0.9 t HCH use in the Austrian part of the Alps and 0.2 t HCH use in the German part of the Alps in 1997, which is similar to the estimated HCH masses in the Alpine forests in 2004: 1.7 t for Austria and 0.2 t for Germany.
Similar has been found for DDT plus metabolites, however with longer periods since their last uses in the studied countries. The two approaches result in 14 and 17 t DDT plus DDD and DDE accumulated in the forest ecosystems of the Alps respectively. This is approximately the same amount that – according to our estimates on the basis of the total national emissions estimated by Pacyna et al. (1999) – has been used in 1980 in the Alpine region of the studied countries respectively. Since 1985 DDT was no longer used in Austria, western Germany and Switzerland. In Italy no uses are reported since 1996.

4. Discussion

The uncertainties of the, albeit coarse, estimates of emissions and loads in the Alps made in this work are comparable to those of the more sophisticated and state-of-the-art mass balances. The uncertainty of the TEQ emissions should be considered an order of magnitude while actual PAH emissions were assessed with a factor of 2–5 (Pacyna et al., 1999).

Also the uncertainties of HCB emissions are large, in particular those from combustion processes, and the estimates of recent PCB emissions rather assess the order of magnitude (Shatalov et al., 2005).

Our emission estimates for the Alps were checked by comparing them with other studies at European Level.

The estimates of SVOC emissions in the Alps obtained in the present study for the years 2000 and 2004 are comparable with those provided by the regional multicompartment model MSCE-POP for 2005 for the same region (http://www.msceast.org/pops/pop_index.html) (Table 2).

For HCH, the estimates made on the basis of data reported by Pacyna et al. (1999) are comparable with those provided by the MSCE-POP model. However emission estimates based on Vestreng et al. (2006) are considerably higher.

The most relevant discrepancy was observed for HCB where the modelled emissions in 2005 are one order of magnitude higher than those estimated in this study. On the other hand, emission estimates for BaP are comparable, in particular between 2004 and 2005. For dioxins and furans the estimates of the model for 2005 are comparable with those obtained in this study for 2000.

Among the pollutants considered in this research, PAH have the highest bulk emissions and loads in the Alps. It is remarkable that still – despite of its long ban – considerable DDT masses are bound in the Alpine forests that are only one order of magnitude lower than those of PAH. Austria and Italy are with few exceptions the countries with the highest estimated emissions and loads in their respective Alpine areas – a result that reflects the higher extent of the Alpine area and population in these countries.

For all the studied pollutants the loads in the Alps are significantly higher than the actual emissions of these compounds in the corresponding regions. This imbalance is even more striking considering that only the loads bound in forests (i.e. half the landcover) of the Alpine area were taken into account and that the input concentrations represent remote sites. This suggests:

- a) the persistence of these compounds and their potential for accumulation even for compounds of shorter half-life like HCH;
- b) a significant additional input of pollutants from outside the Alpine area;
- c) an underestimation of emissions within the Alpine area;
- d) higher emissions in earlier years.

Probably all of the above mechanisms apply to varying extents, depending on the pollutant. Already previous estimates for Austria for the year 1993 indicated considerably lower emissions than loads of SVOCs in the environment (Weiss, 1997, 1998). The results at hand do not differ from those earlier findings although the quality of emission estimates improved. Meanwhile, the emissions of several pollutants have decreased substantially, some were banned. The present estimates indicate that environmental loads respond slowly to decreasing emissions and/or that there is significant input from sources outside the studied regions. Particularly high masses of compounds like DDT and HCH in the Alpine forests, despite their ban in the studied countries many years ago, underline this assumption. Nevertheless, previous estimates of Weiss (1998) showed similar masses of γ-HCH in forest canopy and -soil in Austria, while the present study reveals soil stocks of γ-HCH which are two orders of magnitude higher than those in the canopy. This could be the result of the γ-HCH ban in almost all of the studied countries and the much longer accumulation period in the soil.

According to the results of the MONARPOP monitoring network, levels of POPs in the border areas of the Alps are higher than those in the central Alps which correlates with the higher precipitation in the peripheral parts of the Alps and with the barrier effect of mountain ranges for the long range transport of pollutants (Offenthaler et al., 2009a). Higher inputs at peripheral regions of the Alps have been also documented for inorganic compounds (Mutsch, 1998; Schneider, 1998). A quantitative estimation of the long-range transport towards the Alps can be obtained considering the country-to-country depositions estimated with the MSC-E POP model for PCDD/F in 2004 and BaP in 2005 (Gusev et al., 2006). According to this model, 9–78% of the deposition in the countries studied here origins from national sources. Switzerland and Austria, the countries with the highest share of Alpine territory (over 60% of the national area), are those with the lowest own contribution to country-wide depositions of BaP and PCDD/F, with values ≤30%.

This evidence supports the hypothesis that high precipitation and low temperatures favour the deposition of SVOCs transported by air masses into the Alpine region (Kaiser and Scheifinger, 2009; Offenthaler et al., 2009b).

The third hypothesis requires further investigation, especially for PCDD/F and PAH for which emissions from biomass burning are suspected to be relevant at a local scale (Hays et al., 2005; Prévôt et al., 2006). In this context, recent studies (e.g. Zhang et al., 2008) have demonstrated that the biomass combustion may be a relevant PCDD/F source which was not duly taken into account in the past.

To assess the accumulation over long time periods, total national emissions in the 15 years prior to the sample collection were recalculated for the Alpine area of the studied countries only and compared with the load of pollutants in the Alpine region (Table 3). The load of PCDD/F in the forest soils of the Alps is one order of magnitude higher than the cumulated total national emissions over the preceding 15 years. Despite the long half-life of these compounds

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Table 2: Comparison of the POP emission estimates for the Alpine region of the present study with those of the regional multicompartment model MSCE-POP.

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Units</th>
<th>MSC-E 2005a</th>
<th>MONARPOP 2004b</th>
<th>MONARPOP 2000c</th>
</tr>
</thead>
<tbody>
<tr>
<td>TCDD/F</td>
<td>g TEQ y⁻¹</td>
<td>82</td>
<td>30 46</td>
<td>52 57</td>
</tr>
<tr>
<td>BaP</td>
<td>kg a⁻¹</td>
<td>6683</td>
<td>5220 5933</td>
<td>9150 15375</td>
</tr>
<tr>
<td>HCH</td>
<td>kg a⁻¹</td>
<td>244</td>
<td>155 155</td>
<td>10137d</td>
</tr>
<tr>
<td>HCB</td>
<td>kg a⁻¹</td>
<td>660</td>
<td>63 87</td>
<td>273 407</td>
</tr>
</tbody>
</table>

a Based on MSC East: http://www.msceast.org/pops/pop_index.html.

b Based on Gusev et al., 2007.

c Based on Vestreng et al., 2006.

d Based on the HCH use in Italy in 2000.
in subsurface soils (25–100 years; Paustenbach et al., 1992) and their high affinity for soils with high organic matter content, these figures demonstrate the relevance of the advection of these pollutants from areas outside the Alps.

The mass of the four studied PAH in the Alpine forest soils corresponds approximately to the emissions in the Alps during the preceding 15 years. In the atmosphere, HCB predominates in the gaseous phase (92–100%, Lane et al., 1992). It is resistant to wet deposition, and at high latitudes (or altitudes) it tends to precipitate with the solid phase (Wania and Mackay, 1993). Due to its low water solubility HCB has a low mobility in the soil and is unlikely to be removed by leaching. However, it has a higher vapour pressure and lower boiling point than PAH or PCDD/F and therefore HCB losses from soil by volatilisation may explain the lower concentrations of HCB in the solid matrices with respect to PCDD/F and heavy PAH. Nevertheless, a half-life of HCB in soils of six years or less (Mackay et al., 1992; Rippen, 1997) conflicts with the similarity of forest soil stocks and 15 years’ cumulated emissions in the Alpine area. Again, additional HCB inputs from outside the Alps are consistent with the observed loads.

In contrast with other pollutants the HCH mass in the Alpine forest soils represents only a low share of the emissions of these compounds in the previous 15 years (Table 3). These substances have a higher solubility in water than HCB and, in the atmosphere, occur either in the vapour phase or adsorbed to water droplets or snowflakes. Wet gaseous deposition is the dominant deposition process for this family of pollutants (Lei and Wania, 2004). In the soil HCH are adsorbed to soil particles and have low mobility. However, volatilization of HCH from the soils seems to be relevant and is controlled by temperature and moisture (Samuel and Pillai, 1990). Hence, the low levels observed in the Alpine soils could be associated with losses in the warm season (in particular in spring when moisture is relatively high) and with the shorter half-life of most HCH-isomers in soil.

Nevertheless, the disagreement in the HCH emission estimations between different authors suggests that the discrepancy between emissions and loads is mostly due to the uncertainty in the former.

In addition to the properties of the substances, the residence time of pollutants is related to the physical, chemical and biological properties of the soil. In particular the content of soil organic matter (SOM) is widely accepted as the most important component for the retention of hydrophobic organic compounds (HOC) in most soils and sediments (Chiou et al., 1983; Kile et al., 1995; Means et al., 1980; Schwarzenbach and Westall, 1981). The enrichment factor of compounds like PCDD/F and PAH in soil has been associated with its content of organic carbon (OC) and of fine particles (Schulz et al., 2005). Moreover, a significant correlation between the concentrations of some HOCs and the fraction of organic carbon per dry mass of soil has been reported by Schwarzenbach and Westall (1981). The partitioning of HOCs between particulate and dissolved organic matter increases with hydrophobicity of the compound (Frankki, 2006). The average concentration of OC in the studied soils of the Alpine forests is relatively high, 37% in the humus and varies between 11 and 19% in the mineral layer, and contributes to the retention of SVOCs in this compartment.

### Table 3

Comparison between the cumulated emissions in the Alpine range of the five studied countries in the time window 1990–2004 (estimated on the basis of the national emissions by MSC-E) and the load of pollutants in the Alpine forest soils (this study).

<table>
<thead>
<tr>
<th></th>
<th>PCDD/F</th>
<th>SUM 4 PAH</th>
<th>HCB</th>
<th>HCH</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Cumulated emissions in the Alpine range 1990–2004</strong></td>
<td>kg TEQ</td>
<td>t</td>
<td>t</td>
<td>t</td>
</tr>
<tr>
<td>Load in Alpine forest soils (<strong>&quot;mean&quot;-approach</strong>)</td>
<td>1.25</td>
<td>254</td>
<td>1.4</td>
<td>80</td>
</tr>
<tr>
<td>Load in Alpine forest soils (<strong>&quot;sum&quot;-approach</strong>)</td>
<td>7</td>
<td>469</td>
<td>2.5</td>
<td>4.5</td>
</tr>
<tr>
<td><strong>Load in Alpine forest soils</strong></td>
<td>12</td>
<td>791</td>
<td>3.2</td>
<td>4.7</td>
</tr>
</tbody>
</table>

5. Conclusions

POPs in the Alpine forests are stored mainly in the soil PAH being those with the highest emissions and loads in the Alps.

For all the studied pollutants the loads in the Alpine region are significantly higher than the corresponding emissions in the same area, even for compounds like DDT and HCH whose release to the environment is supposed to have stopped several years ago. In this regard, it is worthy of mention that the approach adopted in the present study (based on average national per capita emissions) tendentially estimates low load/emission ratios since it includes emissions from industrial and metropolitan areas outside the Alps that may result in an overestimation of the Alpine emissions for certain compounds.

The imbalance between emissions and loads underline the persistence and accumulation of POPs in the environment. Characteristically low temperatures and high organic carbon content of Alpine forest soil contribute to increase residence times (and hence persistence) of dioxins, PCB, and the heaviest PAH, up to one or more decades.

On the other hand, the emissions-loads gap also indicates a significant input of pollutants from outside the Alpine area and/or that the emissions within the Alps are underestimated.

The hypothesis of the Alps being a net sink for SVOCs is supported by previous work on PCDD/F and BaP and probably applies to all POPs. Nevertheless, recent studies indicate that emissions of dioxins and PAH from biomass burning from local diffuse sources in the valleys may be relevant for the local mass balance of these compounds and probably have not been sufficiently considered in the available emission estimates.

Very likely all the factors listed above influence the loads of POPs in the Alps, however more research is needed to quantify the relative contribution of each of them for the different substances in order to reduce the considerable uncertainties in the estimation of the emissions.

### Acknowledgements

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