PCDD/F and PCB in spruce forests of the Alps

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Dioxin-like pollution of forests in the Alps shows strong geographic variation.

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ABSTRACT

PCDD/F and PCB concentrations in remote mountainous spruce stands of the Central European Alps show strong geographic variation. Independent of the matrix (0.5 year old needles, humus or mineral soil), the highest pollutant levels were always found at the lateral zones of the mountain range. High levels coincided with strong precipitation, particularly along the northern margin of the study region. The most volatile PCB congener propagated further into the colder, drier central Alps than the heavier species. Matrices with different accumulation history (needles and humus) repeatedly reflected different spatial immission patterns. Consistent with its much longer exposure, pollutant levels in humus exceeded those of needles by up to two orders of magnitude. Needle contamination varied with altitude but the vertical trends were highly variable between transects and changed between years, too.

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1. Introduction

Low temperatures, high precipitation and diurnal winds promote the reception of certain airborne persistent organic pollutants (POPs) by mid-latitude mountain ranges (Daly and Wania, 2005). Consequently, POP contamination of various biotic and abiotic media has been reported even from remote spots in the Alps (Carrera et al., 2001; Jaward et al., 2005). The geographic distribution of POPs across the Alps remained unclear nonetheless. Project MONARPOP1 included a cross-alpine survey of POPs in mountainous forest ecosystems to reveal geographical and altitudinal patterns on a transnational scale. Mountainous forests were chosen as a major constituent of Alpine landcover (by area, ecological and economic value) and because conifer stands both indicate and influence the atmospheric distribution of POPs (Barber et al., 2004; Horstmann et al., 1997; Weiss et al., 1998, 2000, 2003; Kirchner et al., 2006; Tremolada et al., 2007). Of the range of analysed POPs, this article focuses on polychlorinated dibenzodioxins (PCDD) and -furans (PCDF) in needles, humus and mineral soil from Norway spruce stands.

2. Materials and methods

2.1. Site selection

The monitoring network consisted of 33 irregularly spaced sites at a standard elevation of 1400 m a.s.l. plus seven height profiles of 4–5 plots (Fig. 1, description of height profiles: Kirchner et al. [2009]). For site selection, field staff was instructed to keep minimum distances from large pollutors (50 km), small scale domestic heating (500 m), minor roads (500 m), larger roads (5 km) and application of silvi-or agricultural chemicals (5 km) while also minding orographic influences (e.g., shielding of horizontal air exchange). Needles were sampled at all, humus at a subset of 55, and mineral soil at 27 plots. Note that results refer to plots at standard elevation unless (for the description of height profiles) stated otherwise.

2.2. Needles

Six month old needles of Norway spruce (Picea abies [L.] Karst.) were harvested between end of September and end of October 2004, adopting sampling instructions in Umweltbundesamt (1996). On a subset of height profile plots, sampling was repeated in October 2005 and May 2006. Two to three branches were cut from the top seventh whirls of two individuals growing in the inner of even aged (>60 y)
homogeneous stands. The current year’s (2004) twiglets were cut and densely packed to minimize the air space remaining in the glass jars. Samples were transported in insulated boxes with ice packs and/or dry ice added and kept at ca. −20 °C. After defoliation (liquid nitrogen, inert atmosphere), needle subsamples of 15–40 g (depending on analyte) were filled into small cleaned (see above), light protected and airtight glass vials for redistribution among the involved laboratories and kept/ transferred at ca. −20 °C until analysis.

2.3. Humus, mineral soil

At 2.5 points of a 5 m grid, all humus within a 30 × 30 cm square was collected down to the mineral soil border, omitting living plant parts and stones. 10 cm cores of mineral soil were taken from the center of each resulting pit. Samples were collected in glass jars and stored at ca. −20 °C. Humus and mineral soil samples were lyophilised in the original collection vessels. Lyophilised samples were then removed from the glass jars and pooled per matrix and site. Humus pools were ground under cooled conditions to a particle size <0.5 mm, and mineral soil pools were sieved to use the fraction <2 mm. Pools were then homogenised. 90–220 g humus and 30–90 g mineral soil of all samples were shipped to the laboratories, using 100 ml screw-top vials for the mineral soil and 330 ml latchtop vials for the humus. The amount delivered depended on the analytical requirements of the particular laboratory (each participating laboratory analysed its assigned compound(s) for the whole set of samples).

Humus dry mass was determined for 2 g aliquots of each site’s humus homogenate by drying at 105 °C until no further weight loss was recorded. Pollutant concentrations in the lyophilised sample were then converted to dry mass concentrations by multiplying with the ratio lyophilised:dry mass. Likewise, 100 needles of each sample were randomly drawn and desiccated to a constant weight at 105 °C. Again, the obtained ratio was used to convert fresh to dry mass concentrations.

2.4. Chemical analysis

All samples were spiked with 13C-labeled PCDD/F and PCB standards (Cambridge Isotope Laboratories, Andover, MA, USA). Humus and needles were extracted in a Soxhlet extractor, mineral soil in an ASE (Dionex Corporation, Sunnyvale, CA, USA), all with toluene as a solvent. The clean-up consisted of the following three liquid column chromatographies: i) column filled with H2SO4/Celite; elution with n-hexane, ii) mixed layer column from FMS; elution with n-hexane, iii) alumina column from FMS, consecutive elution with n-hexane, n-hexane/dichloromethane = 9:1 and n-hexane/ dichloromethane = 1:1, where fractions 2 and 3 separated PCB from PCDD/F. Prior to injection into high-resolution mass spectrometers (HRMS), 13C-labeled standards were added to each fraction to determine recovery. PCDD/F were measured with an HRMS type Finnigan MAT90 (Thermo Electron GmbH, Bremen, Germany), coupled to an Agilent GC 5890 (Agilent Technologies, Palo Alto, CA, USA). PCB with an HRMS type Finnigan MAT 955 coupled with an Agilent GC 6890. The chromatographic separation was achieved by splitless injection on a DB5MS column for PCBs and on a DB5 and a DBDIOXIN column for PCDD/F (J&W, GmbH, Sulzbach, Germany). The mass spectrometers were operated in SIM mode at a resolution of 6000–10 000, respectively.

All glassware was rinsed twice with deionised water.2 Large glass jars (3 l for needle and 5 l for humus collection) were then heated with rubber gaskets removed for 10 h at 120 °C, small vials for the distribution of needles were heated for 24 h at 200 °C to destroy organic contaminants. Before use and after sample collection, the mounds of the jars were covered with household aluminium foil, untreated, to prevent contact with the gaskets. Glassware for the distribution of humus and mineral soil to the laboratories underwent similar cleaning, heating and sealing (except the rubberless screw-top vials which did not receive the additional aluminium layer).

2.5. Precipitation data

Average annual precipitation 1994–2003 has been calculated from data in Efthymiadis et al. (2006).

2.6. Statistical tests

To test for spatial concentration differences, sites were grouped by longi- or latitude (resulting in three zones of 10–14 observations each). If Kruskal-Wallis’ rank sum test showed significant (α ≤ 0.05) differences among the zones, Wilcoxon’s test was used for pairwise comparison of zones. Alternatively, ANOVA and Student’s t-test were used to examine normally distributed and homoscedastic samples. Shapiro-Wilk’s test was applied to see whether untransformed or naturally loga- rithmised data deviated from normal distribution with an error probability of α ≤ 0.05. The R 2.8.0 environment (R Development Core Team, 2008) was used for computation and display of data.

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2 Elix 10 linked up to a MilliQ, both: MilliPore, Massachusetts, USA.
Table 1
PCDD/F and PCB content of the humus layer of Norway spruce stands.

<table>
<thead>
<tr>
<th></th>
<th>F</th>
<th>Mean</th>
<th>sd</th>
<th>Min</th>
<th>P25</th>
<th>Median</th>
<th>P75</th>
<th>Max</th>
</tr>
</thead>
<tbody>
<tr>
<td>PCDD</td>
<td>2</td>
<td>158.83</td>
<td>116.15</td>
<td>43.3</td>
<td>65.2</td>
<td>119</td>
<td>206.75</td>
<td>477</td>
</tr>
<tr>
<td>PCDF</td>
<td>2</td>
<td>155.01</td>
<td>80.44</td>
<td>63</td>
<td>91.25</td>
<td>132.6</td>
<td>211.5</td>
<td>311</td>
</tr>
<tr>
<td>PCDD/F</td>
<td>2</td>
<td>313.84</td>
<td>188.47</td>
<td>115.7</td>
<td>155.85</td>
<td>262.3</td>
<td>404.55</td>
<td>758</td>
</tr>
<tr>
<td>PCDD/F in TEQWHO</td>
<td>2</td>
<td>4.44</td>
<td>2.79</td>
<td>1.37</td>
<td>2.55</td>
<td>3.70</td>
<td>5.21</td>
<td>10.81</td>
</tr>
<tr>
<td>dl-PCB</td>
<td>2</td>
<td>1591</td>
<td>748</td>
<td>439</td>
<td>1067</td>
<td>1482</td>
<td>1583</td>
<td>3269</td>
</tr>
<tr>
<td>dl-PCB in TEQWHO</td>
<td>1</td>
<td>2.32</td>
<td>1.06</td>
<td>0.70</td>
<td>1.51</td>
<td>2.06</td>
<td>3.01</td>
<td>4.92</td>
</tr>
<tr>
<td>6 PCB</td>
<td>1</td>
<td>8301</td>
<td>3605</td>
<td>3160</td>
<td>5005</td>
<td>7670</td>
<td>11 040</td>
<td>17 220</td>
</tr>
</tbody>
</table>

Unit: ng kg⁻¹ dry mass; 31 samples; F, distribution type: 1 – normal, 2 – lognormal; Pnth percentile; sd, standard deviation.

3. Results

3.1. Concentration ranges

Table 1 lists the pollutant concentrations in needles, humus and the topmost 10 cm of mineral soil of Norway spruce stands in the study area (note that these statistics include all plots, not only those at 1400 m standard altitude). Humus showed the highest pollutant levels (Table 4).

3.2. Concentration differences between matrices

Concentrations decreased from humus over mineral soil to needles, and differences could be up to two orders of magnitude. PCDD/F varied stronger between media than PCB, which showed similar relative contributions of PCDD/F and PCB to total dioxin-like activity (in WHO toxic equivalents for humans: TEQWHO) varied between media: PCB amounted to 91% of total TEQWHO in needles, and differences could be up to two orders of magnitude.

Although pollution loads never culminated in the central region, some pollutants were lowest in a lateral zone, resulting in a monotonous longi- or latitudinal trend across the study region. Such a progression was observed only for humus PCDD concentrations, however. There was an eastwards decrease of PCB 77 and a decline from north to south for the congeners no. 118, 138, 153, 156, 167, 169, 180, 189.

Table 2 shows compounds with significant differences among longi- or latitudinal site groups and indicates the site group with the highest (median) concentrations. Significant differences from north to south (latitudinal) were more frequent than from east to west (longitudinal). Longitudinal variation was more pronounced in needles than in humus. Despite small differences between medians, northern and southern concentration ranges were comparable for needles, at markedly lower loads in the central zone (Fig. 2). The inner zone of the study region never showed the highest concentrations (Figs. 3a, b and 4).

While highest PCB concentrations in humus were always observed at the periphery, the more volatile congener 28 moved farther towards the inner of the Alps than the heavier congener 180 (Fig. 4). No similar contrast was found between tetra- and octachlorinated dioxin or furan homologues, despite their physicochemical differences.

Table 3 shows compounds with significant longi- or latitudinal concentration differences. Although pollution loads never culminated in the central region, some pollutants were lowest in a lateral zone, resulting in a monotonous longi- or latitudinal trend across the study region. Such a progression was observed only for humus PCDD concentrations, however. There was an eastwards decrease of PCB 77 and a decline from north to south for the congeners no. 118, 138, 153, 156, 167, 169, 180, 189.

Fig. 2. Total PCDD/F content [ng kg⁻¹ dry mass] of 0.5 year old Norway spruce needles from (top down) northern, central, and southern parts of the Alps (horizontal shapes indicate boxplot statistics; outlier annotated with site code).

Table 4
Median pollutant concentrations in humus, mineral soil and needle samples.

<table>
<thead>
<tr>
<th></th>
<th>PCDD/F</th>
<th>PCDD/F in TEQWHO</th>
<th>dl-PCB</th>
<th>dl-PCB in TEQWHO</th>
<th>6 PCB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Humus</td>
<td>261.6</td>
<td>4.04</td>
<td>1459.5</td>
<td>2.053</td>
<td>8235</td>
</tr>
<tr>
<td>Mineral soil</td>
<td>77.5</td>
<td>2.00</td>
<td>122.0</td>
<td>0.228</td>
<td>747</td>
</tr>
<tr>
<td>Needles</td>
<td>2.0</td>
<td>0.01</td>
<td>115.4</td>
<td>0.104</td>
<td>853</td>
</tr>
</tbody>
</table>

Unit: ng kg⁻¹ dry mass; parallel observations from 20 sites at standard altitude.

Table 5
PCDD/F and PCB with significant longi- or latitudinal concentration differences.

<table>
<thead>
<tr>
<th></th>
<th>Highest median</th>
<th>Longitudinal median</th>
</tr>
</thead>
<tbody>
<tr>
<td>Humus</td>
<td>ΣPCDD/F, ΣPCDF/F in TEQWHO</td>
<td>West PCB 77, 105, 157</td>
</tr>
<tr>
<td>Center</td>
<td>–</td>
<td>Middle –</td>
</tr>
<tr>
<td>South</td>
<td>–</td>
<td>East –</td>
</tr>
<tr>
<td>Needles</td>
<td>North PCB 77, 105, 118, 126, 156, 167, 180, 189, 26 PCB</td>
<td>West PCB 105, 114, 118, 126, 156, 167, ΣPCB in TEQWHO</td>
</tr>
<tr>
<td>Center</td>
<td>–</td>
<td>Middle –</td>
</tr>
<tr>
<td>South</td>
<td>ΣPCDF/F, ΣPCDD/F in TEQWHO</td>
<td>East –</td>
</tr>
<tr>
<td>ΣPCDD/F</td>
<td>–</td>
<td>CH−07</td>
</tr>
</tbody>
</table>

Fig. 2. Total PCDD/F content [ng kg⁻¹ dry mass] of 0.5 year old Norway spruce needles from (top down) northern, central, and southern parts of the Alps (horizontal shapes indicate boxplot statistics; outlier annotated with site code).
Fig. 3. a: Geographical distribution of PCDD/F in six month old Norway spruce needles. (Dot area proportional to concentration.) b: Geographical distribution of PCDD/F in humus of Norway spruce stands. (Dot area proportional to concentration.)
At the northern fringe of the study region, high PCDD/F and PCB contamination of humus coincided with strong precipitation. This association of precipitation and pollution levels was not so pronounced in the south (Fig. 5).

### 3.4. Altitudinal changes

There was no common altitudinal pattern to all seven height profiles. Different profiles could even display opposed concentration trends (e.g. downhill vs. uphill increases at profiles Klosters and Wechsel in Fig. 6). Other profiles showed nondescript vertical variation (Figs. 6 and 7). As observed with the spatial patterns (Fig. 3a and b), humus and needle samples would reveal contrasting aspects of the immission regime. Along the sample profile, needles and humus showed contrasting or even opposed vertical concentration gradients. For example, PCDD/F toxic equivalents in needles, converse to those in humus, increased with height at Klosters and declined at Wechsel. Different compounds, in turn, could exhibit strikingly similar altitudinal distributions at the same profile (Fig. 8).

### 3.5. Changes between years

Despite identical sites, needle samples from 2005 showed considerably different contamination from those taken in 2004. 2004 PCB concentrations ($\Sigma$dl-PCB, $\Sigma$6 PCB) were markedly higher at all height profiles, whereas the altitudinal variation at each profile was similar (Fig. 9). In contrast, higher dioxin or furan levels occurred either in 2004 or 2005, even at the same height profile. As a consequence, vertical trends of dioxin and furans in needles changed between years, too (Fig. 10).

![Fig. 4. Geographic distribution of two PCB congeners with different physicochemical properties. Top: congener no. 28, bottom: congener no. 180.](image-url)
4. Discussion

Pollutant levels in humus and needles gave different impressions of the geographic distribution of PCDD/F and PCB loads. Humus showed much higher concentrations than mineral soil or needles, which is common for coniferous forests: humus contains more organic matter and, hence, has a higher affinity for lipophilic pollutants than mineral soil. In the absence of soil tillage and rapid biomass removal, humus of mountainous conifer forests accumulates litter and throughfall over at least two decades until full mineralisation, giving an account of the long-time pollution. The six month old needles, on the other hand, reflect the current year’s situation. These different accumulation spans of humus and needles are thought to be the main reason that either matrix yields a – sometimes substantially – different picture of the geographical pollution load. Concentrations in humus will respond more slowly to reduced immissions than in needles. In fact, humus PCDD/F levels in 2004 were two thirds of those in 1994 (compared for seven Austrian sites already investigated in an earlier study: Weiss et al., 2000) while needle levels had decreased to one 1/40 during this period of steadily declining emissions in Austria and neighbouring EU countries (Anderl et al., 2007; EEA, 2007). Besides, exposition time will not only influence the amount of pollutants received but might also shift the chemical cocktail under the influence of degradation, elution and revolatilisation processes (e.g., photolysis on the needle surface: Niu et al., 2003).

Concentrations of different dioxin-like substances can be compared by their TEQ (toxic equivalents to the “Seveso” dioxin), which gives an estimate of their environmental concern. Interestingly, the contribution of dioxin-like PCBs to overall (PCB + PCDD/F) TEQ concentration decreased from needles over humus to mineral soil and, hence, with the age of the matrix. TEQ in needles were mainly (9/10) caused by PCB, while PCDD/F contributed twice as many TEQ as PCB in humus and were responsible for most TEQ found in mineral soil (Table 4). In soil, dioxin-like PCB persist with a half-life of ca. 10–19 years (congeners 126 and 169: Sinkkonen and Paasivirta, 2000), which is shorter than the 29–270 years of relevant (2,3,7,8-substituted) PCDD/F. Their faster degradation might thus contribute to the lower dioxin-like activities of PCB in humus or mineral soil compared to needles (under the simplifying assumption that neither compound class is preferentially captured or retained by needles, humus or mineral soil).

Although longi- and latitudinal concentration gradients varied between pollutants and media, peak concentrations were never found in the middle part of the study region. Highest concentrations thus always occurred at the fringe of the alpine region. Two main conclusions can be drawn from this observation. First, that the main POP load to the Alps originates outside the Alps. Second, that the alpine range is a barrier for long-range atmospheric POP transport. The barrier effect is thought to arise from precipitation and temperature gradients across the alpine range. The alpine range is characterised by northern and southern zones of high precipitation which flank the drier inner alpine region (Fig. 5). Wet deposition of advected airborne contaminants should therefore be higher at the latitudinal extremes of the investigated area. Indeed, needles from the north and the south showed significantly higher PCDD/F-concentrations than those from the central part of the study region (Fig. 2), while differences between north and south were not significant. Humus PCDD/F-concentrations, however were still significantly higher in the north, but in the south one homologue (notably the least volatile furane: OCDF) reached significantly higher humus levels than the central Alps. Graphically speaking, the needle samples indicate pollution curtains along both sides of the mountain range while humus samples suggest that, on a several years’ average, PCDD/F loads mainly wash over from the north. This could merely reflect the difference between a 2004 snapshot (needle samples) and the long term record (humus) of the atmospheric situation. A deviation of 2004 from the perennial average is not unlikely, but to date sufficiently long time series of appropriate air monitoring data (cf. Offenthaler et al., 2009) are not yet available. Apart from differences in atmospheric supply, however, ambient temperature might also help to explain different humus concentrations at comparable precipitation rates. Lower average temperatures can be expected to increase condensation (and inhibit summerly revolatilisation; cf. Tremolada et al., 2007) of gaseous POPs. As a consequence, more volatile POPs should propagate farther towards the colder, drier sites in the centre of the

Fig. 5. Precipitation and PCDD/F-concentrations in humus of Norway spruce stands (size of grey dots increases with precipitation).
Alpine region. In fact, the most volatile dioxin (TCDD) was the only homologue of which humus from the coldest (central) part of the Alps had significantly higher levels than the warmer southern zone. Such an assumed temperature-driven fractionation might also be reflected by the concentration of some PCB in humus, where congener no. 28 (vapour pressure of the liquid $p_L = 24$ mPa: Mackay et al., 1991) was found deeper inside the alpine area than the less volatile congener no. 180 ($p_L = 0.16$ mPa): Fig. 4. Following recent simulations by Wania and Westgate (2008), the accumulation of airborne POPs in mountains – as opposed to high latitudes – is governed by rapid scavenging processes (because air masses pass relatively fast compared with the transport to polar latitudes). The washout of particulates would thus assume a dominating role in the transfer of atmospheric PCB and PCDD/F to the surface. As air temperatures drop with altitude and towards the inner of the Alps, the gaseous fraction of the contaminants will successively transit into the particulate phase, from which it is washed out much more efficiently than from the gas phase. This could explain why the most volatile gaseous PCB or PCDD/F reach the inner Alpine region where, finally, temperatures force them into the efficiently trapped particulate phase, while the heavier congeners – already prevailing as particles – hardly cross the precipitation curtain at the northern and southern fringe of the Alps.

There was no uniform trend for altitudinal concentration changes. Quite contrarily, continuous vertical trends – if any – could turn out opposite ways: pollution levels could increase or decrease...
with height, depending on the particular height profile. Dioxin or furan concentrations in needles could even show different gradients between consecutive years. Altitude alone was obviously no simple proxy for PCDD/F or PCB levels in our study, despite its climatological conjunction with precipitation and temperature (cf. Kirchner et al. 2009). Local climate, especially precipitation and air currents, can vary within relatively short distance in the rugged terrain of the Alps. This may be the main reason for the weak correlation of pollution with altitude, particularly at such height profiles where accessibility prevented the selection of sites up the same slope. However, even transsects of identical exposition (e.g. “Rauris” profile⁴) could lack a clear altitudinal dependency of pollutant levels (Figs. 6–8). “Klosters” is an exception among the

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⁴ 3D – views of height profiles can be explored with Google Earth © and the corresponding overlay at www.monarpop.at/downloads/MONARPOP_height_profiles.kml.
height profiles because it ascends from the Swiss town Klosterson (3900 habitants): the increase of some PCDD/F levels towards the valleyground (Fig. 6) is plausibly attributable to local sources.

5. Conclusions

The occurrence and amount of PCDD/F and PCB in forest sites across the greater part of the Alps indicates that the alpine range is a barrier for the long-range atmospheric transport of these pollutants. The highest pollutant concentrations were never found in the inner of the study region, and the coincidence of high pollution and precipitation at the fringe of the study region suggests that washout plays a dominant role in the transfer of airborne PCDD/F to the ground. There was evidence that some more volatile PCB congeners propagate further into the inner alpine area with lower temperatures and presumably increased cold condensation. The geographic and altitudinal pollution gradients conveyed by needle or humus data were incongruent, as were the results of needle data of consecutive years. Interpretation of environmental data obviously has to consider a given receptor’s window of exposure, for instance the snapshot character of needle bio-indication in contrast to the pollution archives kept in forest humus. The results from seven altitudinal transsects were too heterogeneous for any general conclusions on vertical pollution gradients. This is a likely consequence of the high climatic variability throughout the study region and illustrates the challenges for high-resolution pollution models in areas of complex orography.

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