

Temporal Trends and Spatial Distribution of PCDD, PCDF, and PCB in Pine and Spruce Shoots

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Abstract: In Germany, there is a lack of consistent and comparable data for the time dependent behaviour and spatial distribution of dioxin-like and indicator PCB in ambient air, deposition and plants. The aim of this study was to improve the data on PCDD/PCDF, dioxin-like PCB and non dioxin-like PCB in spruce and pine shoots from different locations and years by retrospective monitoring. The survey was conducted with archived samples of one-year old spruce shoots (*Picea abies*) and pine shoots (*Pinus sylvestris*) from the German environmental specimen bank. Two sets of samples from locations in urbanized areas in western and eastern Germany (Warndt and Duebener Heide Mitte, respectively) were investigated as time series. Additionally, spruce shoots from seven different rural locations sampled in the years 2000–2004 were analyzed in order to get an overview about the spatial distribution of PCB and PCDD/PCDF. The analytical results of the samples from the two urbanized areas clearly show that the atmospheric contamination with PCDD and PCDF has declined by about 75% between 1985 and 1997 at Warndt and about 40% between 1991 and 1997 at Duebener Heide. However, concentrations stayed virtually constant at both locations from 1997 to 2004 at a level of about 1 ng WHO-TEQ/kg dry matter (d.m.). Similarly, the investigation of spruce shoots from rural locations from 2000 to 2004 did not reveal a temporal trend at any site. PCDD/PCDF levels were between 0.1 and 1.0 ng WHO-TEQ/kg d.m. At the urbanized location Warndt the six indicator PCB as well as the 12 dioxin-like PCB according to WHO revealed a significant decline by more than 75% between 1985 and 1999. Thereafter, PCB levels stayed virtually constant. At the location Duebener Heide an overall decrease of PCB concentrations in pine shoots of about 60% was detected between 1991 and 2004. Spruce shoots from all locations showed a relevant contribution of dioxin-like PCB to the total WHO toxicity equivalent (PCDD/PCDF + PCB). In most samples, the contribution of dioxin-like PCB was between 21% and 41%. The TEQ contribution of PCB in the samples from three rural sites was higher and similar to the TEQ value of PCDD/PCDF. The investigated pine shoots from the urbanized site Duebener Heide showed a 15–28% contribution of dioxin-like PCB to total TEQ and thus lower than in spruce shoots from different locations. In all samples except one PCB 126 contributed to more than 80% to the PCB-TEQ.