Attempts to Balance Transport and Fate of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans for Baden-Württemberg

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It is a declared goal of German environmental politics to lower the human exposure to polychlorinated dibenzo-p-dioxins (PCDDs) and dibenzofurans (PCDFs) to below 1 pg I-TEQ/kg of body weight per day. The current daily exposure of adults in Germany is calculated to be about 2.5 pg I-TEQ/kg body weight. In order to achieve this goal it is necessary to concentrate in minimizing emission on the relevant sources and relevant exposure pathways. However, the question arises, whether our knowledge of the relevant sources and pathways is sufficient.
We have studied in the past years transport phenomena of PCDDs and PCDFs and have made an attempt to balance transport and fate on the basis of our current information.

The result can be summarized as follows:
With regard to its PCDD/PCDF content the surface area of the state of Baden-Württemberg can be divided in three categories: forest, agricultural land and the remaining, mainly industrialized and densly populated area. There is a fourth category, which will not be considered here, heavily contaminated, but comparatively small areas, with up 29000 ng I-TEQ/kg of soil. On the total surface area has accumulated in the last 30 to 40 years a total amount of PCDD/PCDF, which is estimated to about 18 kg I-TEQ. The basis for this calculation is shown in Table 1. In sediments of rivers and lakes we estimate at present a total amount of 0.1 kg I-TEQ.
This total PCDD/PCDF potential present in the surface area is balanced by the total deposition via dust, estimated to be about 26 kg I-TEQ, which is in the same order of magnitude. The basis for this calculation is shown in Table 1: 100 mg dust/m$^2$ and day; avarage PCDD/PCDF concentration 500 ng/kg dust; assumed time of deposition and accumulation 40 years.
In addition to the PCDD/PCDF potential in the area one has also to consider the input through PCDD/PCDF containing waste, part of which is accounted for in Table 3. Input through municipal waste and sewage sludge has to be considered as a secondary source and is difficult to account for in a transport and fate balance. The contribution of the PCDD/PCDF in solid residues from waste incineration to the deposition by redistribution is also unknown.

# TRANS

While the balance for input, calculated on the basis of surface area PCDD/PCDF content, and input, calculated from current deposition, is at least in the same order of magnitude, the known thermal emission sources to air, shown in Table 2, account only for a minor fraction of this amount of PCDD/PCDF. Municipal waste incineration amounts to only about 3 % of the total deposition.

That we obviously still have a limited knowledge of transport and fate of PCDD/PCDF is also shown by the amount of PCDD/PCDF calculated from ambient air data (Table 4). Assuming an uniform distribution of PCDD/PCDF in the air mass of 0-100 m above ground, we calculate 161 g I-TEQ for this layer and we have to assume that these PCDD/PCDF are completely washed out several times a year. So we have a problem with the sources for these PCDD/PCDF. Is it all import by long distance transport? Or does it come from redistribution? These questions cannot be answered at present.

## Table 1

### PCDD/PCDF land inventory for Baden-Württemberg

| | area (qm) | depth (d) cm (g/ccm) | ng I-TEQ/kg | Total I-TEQ in g |
|---|---|---|---|---|
| *Forest* | 397445100 | 5 (0.6) | 30 | 11923 |
| *Agricultural area* | 5260206000 | 30 (1.0) | 1.1 | 5786 |
| *Remaining area* | 49713300 | 10 (1.0) | 10 | 497 |
| *Sediments* | 600 km/10 m | 10 (1.0) | 10 | 60 |
| **Sum** | | | | **18266** |

### Deposition with dust

assumption: 100 mg dust /m^2,d
          500 ng I-TEQ/kg

| | I-TEQ g/Jahr | I-TEQ since 1950 |
|---|---|---|
| | **653** | **26100** |

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**Table 2**

**Emissions to air from known sources**

|  | I-TEQ g/Jahr | I-TEQ g since 1950 |
|---|---|---|
| 3  *Municipal waste incinerators* | 33 | 660 |
| *Woodburning* | | |
| *Houshold* | 5 | 100 |
| *Industrial* | 10 | 200 |
| *Others* | 30 | 600 |
| **Sum** | **78** | **1560** |

**Table 3**

**Input through solid residues**

|  | t/year | I-TEQ g/year | I-TEQ g since 1950 |
|---|---|---|---|
| *Municipal waste* | 3000000 | 180 | 7200 |
| *Sewage sludge* | 225000 | 18 | 720 |
| *Solid waste from MWC* | | | |
| *Filter dust* | 16500 | 215 | 4300 |
| *Slag* | 200000 | 20 | 400 |
| **Sum** | | **433** | **12620** |

**Table 4**

**Transport by air**

assumption:   45 fg I-TEQ/m^3

uniform PCDD/PCDF concentration in air layer from 0-100 m

Air mass contains:        **161 g I-TEQ**

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Another unexplained phenomenon is the discrepancy of the homologue and congener patterns from "thermal" sources, ambient air and surface coverage in forests on the one hand, deposition and agricultural land on the other hand. While in emissions from thermal sources the lower chlorinated dibenzodioxins and especially the lower chlorinated dibenzofurans contribute considerably to the total PCDD/PCDF, we find this distribution more or less conserved in ambient air and in the organic surface layer of forest areas, but no longer in deposition and agricultural land (Figure 1). Obviously, considerable amounts of lower chlorinated PCDD/PCDF has been "lost", which means that even a larger amount of emission is unaccounted for.
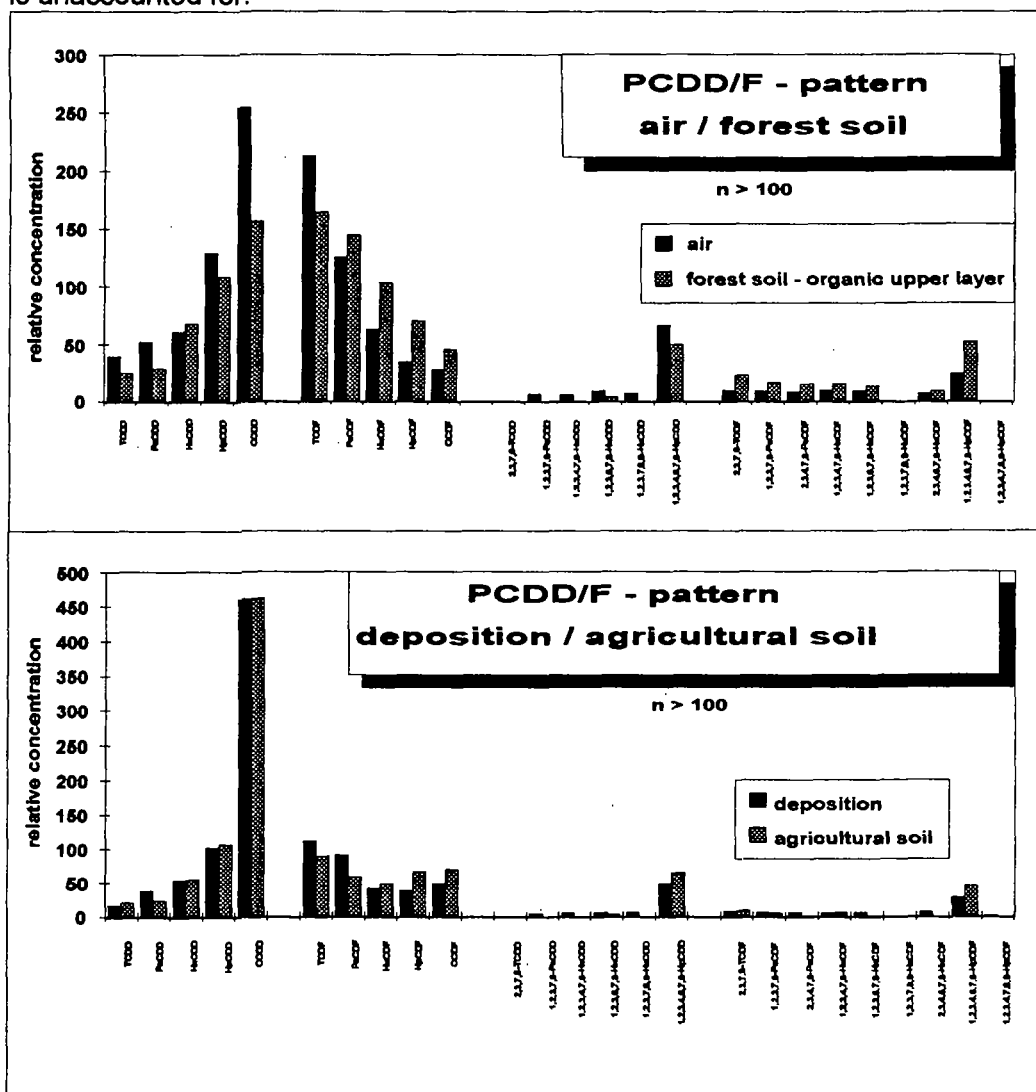


Figure 1 Comparison of the homologue and 2,3,7,8-substituted congener profiles for air and forest soil (upper graph) and of deposition (dust) and agricultural soil (lower graph).

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