Contributions of Diesel-Powered Vehicles and Wood Burning to overall PCDD/PCDF Emissions

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Introduction

There have been attempts to balance the PCDD/PCDF emission from known sources with either the PCDD/PCDF deposition /1,2/ or the human intake of PCDD/PCDF /3/. In all three studies its was found, that there is a considerable difference between the PCDD/PCDF emission (in TEQ) estimated from known sources and either the PCDD/PCDF deposition or human exposure actually measured. The most likely explanation for this discrepancy is either an incorrect quantitative estimation of PCDD/PCDF emission from known sources or the existence of still unknown sources of dioxin emission.
Recently Jones /4/ suggested that dioxin emissions from heavy duty diesel trucks might be one of those underestimated sources of dioxin emission. Another source of dioxin emission which is difficult to asses quantitatively is wood burning in residential and industrial facilities. The attempt is made to analyse the contribution of these two emission sources on the basis of currently available data.

Balancing studies for dioxin emission in Germany

In Germany two balancing studies have been carried out, one for the state of Baden-Württemberg and one for the western part of Germany, comparing the deposition of PCDD/PCDF with the PCDD/PCDF emission from known (major) sources (Table 1). Wintermeier and Rotard calculated for 88% of the area of Germany a deposition rate of 5 to 20 pg TEQ/(m^2·d), while we determined for the state of Baden-Württemberg an average deposition of 30 pg TEQ/(m^2·d) for the years of 1991 to 1993. The total amount deposited, as calculated in the two studies, is shown in Table 1. The emission of PCDD/PCDF calculated for known sources both for the state of Baden-Württemberg, based mainly on our own data, and for the western part of Germany show a considerable discrepancy compared to the deposition. In Tables 2 and 3 average PCDD/PCDF emission from various sources are listed.

Table 1: Comparison of dioxin emission and dioxin deposition

	Baden-Württemberg /1/	Germany /2/
Deposition	0.400 kg TEQ/year	1 to 4.5 kg TEQ/year
Emission from known sources	0,068 kg TEQ/year	0.8 to 1.4 kg TEQ/year

Table 2: Estimated dioxin emissions from known sources for Baden-Württemberg

	g TEQ/year
Municipal waste incineration	37
Wood burning	
residential	10
industrial	10
Hospital waste incineration, after 1990	<0,01
Secondary aluminium production	3
Steel production	6
Cement production	1
Coal and oil fired power plants	0.4
Crematories	0.7
Automobiles	0.05
Total emission	*68*

Table 3: Estimated dioxin emissions from major known sources for Germany/2/

	g TEQ/year
Waste incineration	360 - 630
Metal recycling	400 - ?
Residential heating	20 - 45 (260)
Automobiles	11 - 50
Total emission	*800 - 1200 (1400)*

PCDD/PCDF emissions from diesel-powered vehicles

When the published data on PCDD/PCDF emission from gasoline-powered vehicles are compared (Table 4), there is rather good agreement between the various studies with the exception of the data reported by CARB for cars run on unleaded gasoline equipped with a catalyst. The data for the leaded fuel in Table 4 were obtained, however, with fuel that contained still halogenated scavengers. In Germany, and probably other countries as well, leaded fuel does no longer contain these scavengers. One measurement carried out in 1993 by our laboratory on scavenger free leaded gasoline showed PCDD/PCDF emission values comparable to the unleaded gasoline. There are no data available on the PCDD/PCDF emissions from cars run on current fuels which, due to the ban of halogenated scavengers, may altogether have lower PCDD/PCDF emission than previously reported, because the chlorine content in unleaded gasoline may in part have been due to the use of scavengers.

PCDD/PCDF emissions from diese-powered vehicles

The literature on PCDD/PCDF emission from diesel-powered vehicles is quite limited, especially as far as heavy-duty trucks are concerned, and somewhat contradictory. The available data are summarised in Table 5. While we found rather low PCDD/PCDF emission (0.075 ng TEQ/l) for light-duty vehicles, comparable to those found for cars run on unleaded gasoline with a catalyst, the CARB study and the tunnel-study by Oehme et al. show rather high emissions of 27 and 18 ng TEQ/l, respectively.

When the PCDD/PCDF emission from diesel-powered vehicles is calculated for Germany on the basis of these data the PCDD/PCDF input is either 1.4 g TEQ/year (with 0.075 ng TEQ/l) or 450 g TEQ/year (with 27.4 ng TEQ/l). With the latter value the emission from diesel vehicles would be in the same range as the emission from 43 municipal waste incinerators in

1991. But even if this high value would be the correct one for diesel-powered vehicles, one could not completely close the gap between deposition and emission.

Table 4: PCDD/PCDF Emission from Gasoline-Powered Vehicles

Vehicle class	Data source	ng TEQ/km	ng TEQ/l
Light duty leaded fuel	Oehme et al./5/	0.27	2.7[a]
	Marklund et al./6/	0.06	0.55
	Hagenmaier et al./7/	0,12	1.08
	CARB/8/	0.20	1.79
Light duty unleaded fuel no catalyst	Marklund et al./6/	0.01	0.09
	Hagenmaier et al./7/	0.01	0.09
Light duty unleaded fuel with catalyst	Marklund et al./6/	0.01	0.09
	Hagenmaier et al./7/	0.002	0.02
	CARB/8/	0.24	4.33

[a]Calculated from ng/km with 10 l of gasoline per 100 km

Table 5: PCDD/PCDF Emission from Diesel-Powered Vehicles

Vehicle class	Data source	ng TEQ/km	ng TEQ/l
Heavy-duty truck	Oehme et al./5/	5.4	18[a]
	CARB/8/	4.9	27.44
Bus	CARB/8/	0.38	5.9
Light duty car	Hagenmaier et al./7/	0.005	0.075

[a]Calculated from ng/km with 30 l of diesel per 100 km

Recently we have analysed the PCDD/PCDF emission of a bus, run either on steady state or on the "Berlin-cycle". In both cases we were unable to detect PCDD/PCDF with a detection level of 1 pg/l for individual congeners. Brominated and mixed chlorinated-brominated dioxins and furans were also below detection limit. This is a strong indication that the main source of halogens for dioxin formation observed previously, were the scavengers used in leaded gasoline which entered the diesel fuel when the same transport tanks were used for both gasoline and diesel transport.
Recent analyses of street dust and wall wipes from tunnels also do not support the assumption that emission from vehicles and diesel trucks in particular contribute more to the overall PCDD/PCDF emission than previously calculated. The contribution might be even lower where the use of halogenated scavengers has been banned.
There clearly are more up to date data necessary to realistically estimate the input of dioxin from vehicle emission.

PCDD/PCDF emission from wood burning

A large quantity of wood is burned at industrial operations, but the practice has not been well characterised. A number of studies have found dioxins in the emissions and ash/soot from wood fires in non-industrial situations. The major problem in estimating an average PCDD/PCDF emission for a particular area is the wide variation of PCDD/PCDF emission found for various facilities and operating conditions.

In a study by Schatowitz et al./9/ PCDD/PCDF emission ranged from 0.02 ng TEQ/m^3 to 14.4 ng TEQ/m^3 for facilities with 6 to 1800 kW. In a study carried out recently we found for facilities with 39 to 800 kW PCDD/PCDF emission in a range of 0.02 ng TEQ/m^3 to 16.0 ng TEQ/m^3/10/. Since there are thousands of these industrial wood burning facilities in Germany, it is difficult to asses their PCDD/PCDF input to the environment on currently available data. Wood burning, both non-industrial and industrial might be one of the underestimated sources of PCDD/PCDF emission.

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