Steps Towards an European Dioxin Emission Inventory

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Introduction

In February 1993, within the "5th European Program of Policy and Action", the European Council of Ministers laid down its political goal to reduce the entry of dioxins and furans (PCDDs/PCDFs) into the environment and with it into the human food chain as a precautionary measure. This program strives to reduce the emissions of dioxins until the year 2005 by 90 % as compared to the reference year 1985.

To identify PCDD/PCDF emitting sources and to quantify the emitted amounts the European Commission, DG XI commissioned a research program in 1994 to the North Rhine-Westphalia State Environment Agency. This program consists of two stages:

Stage I — already finished — aimed at the collection of information available in 17 Western European Countries (15 EU members, including Norway and Switzerland) concerning PCDD/F emissions to air, via waste water and residual matter for the reference period 1993 to 1995. Based on this collected information the most important emission sources as well as the main data gaps were to be identified. The results of Stage I are available as a comprehensive report [1]. Stage II, which started in 1997, comprises of international dioxin emission measurement programs to fill in identified data gaps. In this paper the results of Stage I are summarised and the current and near future activities of Stage II will be presented.

Methodology

The inventory of dioxin emissions for the European countries [1] was set up by a 4 step process:

1. information from emission inventories and results of measurement programs performed so far by the national authorities were collected and reported after rearrangement to a common structure. For comparison purposes and to allow for use of the data in ongoing international emission inventories the CORINAIR '94 emission source categorisation (SNAP 94 [2]) was adopted with some modifications.
2. the collected data were analysed in order to set up a list of the most important ***air*** emission sources. The emission freight, flue gas concentrations and uncertainty considerations were taken into account to select those emission sources which are likely to generate about 90% of the European dioxin emissions.
3. For the selected sources a re-estimation of the annual PCDD/F emissions to air was made based on default average emission factors and statistical activity rates

considering all 17 countries including those states which provided no or incomplete emission data.

4. The results of the re-estimation were evaluated and commented with respect to the major emission sources and the main uncertainties and data gaps.

Results and discussion

Emissions to air

The re-estimation presented in our study reveals that the European dioxin air emissions are nearly twice the annual emission freights reported by the available national inventories (table 1). This is not unexpected, since almost no or few information was obtained from a number of countries. Moreover, scope and quality of existing emission inventories differ widely ranging from comprehensive reports based on numerous emission measurements to simple one-page tables with estimates purely based on foreign results. Clearly, there were considerable improvements during the last decade in a number of European countries on the way to a survey on dioxin air emissions. However, even the re-estimate presented here must be regarded as considerably uncertain due to the lack of many basic data or their low quality, respectively.

	national reports	re-estimation
A	29	121
B	727	484
CH	182	183
D	600	840
DK	43	50
E	134	327
F	621	1119
GR	no data	122
I	no data	1050
IRL	no data	33
L	29	50
N	45	41
NL	89	117
P	no data	127
S	36	89
SF	25	69
UK	715	928
Total	3273	5750

Table 1 Reported and re-estimated annual PCDD/F air emissions

According to the re-estimation results the European annual dioxin emissions to air appear to be dominated by a few emission source categories, which however represent a large number of individual sources (table 2). In the reference period 1993-1995 the largest emission could be assigned to municipal waste (msw) incineration followed closely by iron ore sinter plants. Meanwhile these two source types are likely to have exchanged there place in the ranking due to abatement measures on msw incinerators installed in several countries. The reduction of flue gas concentrations of msw incinerators to values below 0.1 ng I-TEQ/m³ is state of technology and would decrease the European air emissions to about 20 g I-TEQ/year if applied throughout Europe. By contrast, to decrease the emissions from sinter plants appears to be much more difficult; a reduction by a factor 5 compared to our re-estimate seems feasible if most modern abatement technologies are applied to all operating plants.

The emissions from the three source types next in the ranking list (residential wood combustion, clinical waste incineration, accidental fires) could only be assessed with high uncertainties and the estimates should therefore be considered cautiously. On-site incinerators for clinical waste already have been shut down in some countries but still are operated in other regions. A large number of facilities exist in the non-ferrous metal sector, in particular using secondary materials (scrap, filter dusts etc.). Many of them are likely to

CORINAIR SNAP	Source type	PCDD/F I-TEQ/a	remark	uncertainty of EF/AR
090201	msw incineration	1467 +174	decreasing trend *)	low/low
030301	Sinter plants	1010 +115	**)	medium/low
0202	Residential wood combustion	945	use of contaminated wood uncertain	medium/high
090207	Clinical waste incineration	816	few plant data and statistics	high/high
06 04 06	wood preservation	381	from PCP-treated goods[4]	v. high/v. high
12	Fires	380	based on ref. [5]	v. high/v. high
030308-10	non ferrous metals	136	Cu, Al, Zn	medium/low
07	road transport	111	mainly leaded fuel; decreasing trend	low/low
	TOTAL	**5535**		

Table 2 Most important air emission sources as revealed from the European Emission Inventory (EF: emission factor; AR: activity rate)
*): illegal domestic burning of msw;**) Sinter plant for recycled materials

have flue gas concentrations above 0.1 ng I-TEQ/m³ (for instance, up to 200 ng I-TEQ/m³ were found recently in the flue gas of a German zinc producing plant [3]. As the emissions from industrial sources will decrease non-industrial sources gain importance. Unfortunately the emissions from wood combustion, accidental fires and from PCP treated wood are difficult to assess because emission factors and corresponding activity rates are not available or highly uncertain.

Emissions via residual materials and waste water
Much less data are available for non-atmospheric emissions of PCDD/F. These data, in most cases reported from few countries, concern a small number of source types only. Emission factors are not reported generally; hence a calculation of an emission estimate for Europe is almost impossible. Reported annual emissions amount up to ca. 3,500 g I-TEQ/a for residues (table 3) and 17 g I-TEQ/a for waste waters. The real European emissions are expected to be much higher with residual emissions exceeding atmospheric emissions by far. However, PCDD/F bound to solids are unlikely to enter the food chain on a short-time scale except for materials like sewage sludge which to some extent is spread on agricultural land.

Main data gaps/future activities
As PCDD/F enter the food chain mainly via the atmospheric pathway there is an urgent demand for identification of the main emission sources. With respect to the sources shown in table 1 data gaps still exist for msw incinerators in Italy, Spain and in particular from the large number of small facilities in France. These gaps are to be closed partly by the ongoing French national dioxin program and by a measurement campaign carried out in Spain within Stage II of the project presented here. No emission measure-ments have been

(officially) reported so far for iron ore sinter plants being operated in Belgium, Spain, France, Italy and Portugal. Since slight variations of the flue gas concentra-tions may have conside-rable im-pact on the European emission freight each facility should be subjected to measurements. With-in Stage II this will be done at Belgian and Spanish plants. Regarding cli-ni-cal waste incine-ra-tion an im-prove-ment of basic data (number and type of facilities, waste statistics) is required. In Stage II emission measurements are scheduled at facilities in Spain, Denmark and Poland to assess the range of emission factors. Further sub-projects comprise measurements at non ferrous metal production sites in the UK as well as a specific study to improve the knowledge about non-atmospheric emissions in Europe. Finally, attempts will be made to improve the knowledge about dioxin emissions from wood combustion and regarding heavy duty diesel engines.

source type	material	emissions g I-TEQ/a	countries considered
09 10 04 Land filling	msw	1623	D
09 02 01 msw incineration	ESP ash, fly ash, slags	1511	B, NL, S
12 Fires	deposited soot	220	D
04 05 03 VC production	Catalyst sludge	68	B, D
09 10 03 sludge spreading	sewage sludge	37	D
TOTAL		**3459**	

Table 3 Main PCDD/F emissions via residual materials as reported by the surveyed documents

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