

The European Dioxin Air Emission Inventory Project—Final Results

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Abstract

Main results of the second stage of the so-called “European Dioxin Emission Inventory” are presented. They cover emission testing data gained from various facilities in the EU (among these the first emission measurements reported from Portugal and Greece) and some central European countries. Further, updated dioxin emission estimates for the most important emission sources in the 17 western European countries and an evaluation of the emission time trend from 1985 to 2005 are presented. The major conclusions are, that

- at present, iron ore sintering is likely to be the most important emission source type followed by the former “No. 1”, municipal waste incineration;
- measurement data from a considerable number of installations are still missing, in particular from the metal industries in Spain and Italy;
- there still exist an unknown number of health care waste incinerators with flue gas PCDD/F concentrations above 100 ng I-TEQ/m³ which must be considered as important local sources;
- in general, considerable emission reduction has been achieved with respect to the industrial emission sources, whereas emissions from non-industrial sources hardly decreased;
- hence, in the near future the emissions from non-industrial sources are likely to exceed those from industrial installations;
- the goal of 90% emission reduction set in the 5th EU Action Programme will be achieved for some source types only.

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Keywords: PCDD/F; Inventory; Sources; Industry; Domestic; Abatement; Environmental policy

1. Introduction

At the EU Council Conference in June 1993, the German Delegation presented a memorandum to the Council with the objective of compiling the knowledge available on dioxin emissions from industrial sources in the Member States, evaluating it and demonstrating the possibilities of limiting the emissions.

The European Commission took up the subject in 1995 and implemented the project “Identification of Relevant Industrial Sources of Dioxins and Furans; Quantification of their Emissions and Evaluation of Abatement Technologies”. Due to the fact that important data on dioxin emissions from metallurgical and other processes had been gained from the emission testing program of the German State of North Rhine—Westphalia, the State Environment Agency (Landesumweltamt NRW, LUA) was committed to carry out this project. Starting in 1995, Stage I of the project was finished in November 1997 with the publication of a

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900 page report covering the information on dioxin emissions available from 17 European Countries and an evaluation of these data to estimate the annual emissions of these countries on a comparable basis (Quaß et al., 1997).

According to that study, which reflected the emission situation of the period 1993–1995, still the emissions from municipal solid waste incinerators appeared to be the largest contributor to the overall European dioxin emission freight. Further, the emissions from iron ore sintering plants were identified as emission sources of similar importance. Considerable emissions—of the industrial facilities covered by the report—were further reported to be released from hospital waste incineration and secondary non-ferrous metal production. Besides, non-industrial processes like domestic wood and coal combustion, accidental fires and dioxin release from PCP treated wood were found to be important, but less quantifiable emission sources for PCDD/PCDF.

It became obvious quite early that considerable data gaps still existed for a number of potential and relevant dioxin emission sources; moreover, several countries had gathered no or few own related information up to that date. To reduce these uncertainties and to “catalyse” the implementation of corresponding national actions the Commission approved a second stage of the project. The objectives of this Stage II were

- to carry out case studies on emission sources;
- to update the Stage I emission inventory on the basis of the case study results and further information from the Member States;
- to evaluate whether the 5th Action Programme’s goal of a 90% reduction of dioxin emissions until 2005 compared to 1985 would be achieved;
- to provide a first overview about the dioxin emission situation in central European countries, with special regard to the accession states;
- to provide a preliminary inventory on PCDD/F emissions to land and water via non-atmospheric pathways.

This paper addresses the first three of these objectives.

2. Approach

For the planned emission measurements in various countries of the European Union a broad co-operation with experienced national measuring institutes and local environmental authorities was needed and several sub-projects were to be installed (see Table 1). LUA took over the tasks of negotiating with potential partner institutions, standardisation of data transfer, quality check by cross-analysing emission sample extracts in its

own dioxin laboratory and reporting to DG Environment. Moreover, some special research programs including emission measurements at potential dioxin emission sources were carried out by LUA’s emission testing group as well (Table 1). During an international “kick-off” workshop held in 1997 a preliminary ranking list for facilities to be tested was agreed on. It was further decided not to address municipal solid waste incineration due to the large data set already available. However, it revealed impossible—for various reasons—to obtain the permission for carrying out all the scheduled emission testing. In particular this was the case regarding sinter and non-ferrous metal plants in Spain and the UK. Contrarily, measurements could finally be done also in countries which did not participate in the workshop like Portugal and Greece.

In order to update the emission inventory, results from case studies and additional information from inventory programs of the Member States were considered. In general, emission estimates were generated by multiplication of emission factors with corresponding activity rates (so-called “top-down” approach). In certain cases more detailed information was available allowing to use the “bottom-up” method by adding emission values of the particular facilities. To provide an impression on the uncertainty the emission estimates are presented in terms of minimum/maximum ranges, which mostly are based on the range of emission factors reported.

For the evaluation of the emission time trend between 1985 and 2005 a retrospective inventory for the year 1985 and a prospective inventory for the year 2005 had to be compiled.

However, information on dioxin emissions for the year 1985 is scarce. At that time, emission testing had been carried out at waste incinerators and a few further sources only; in many cases, results were not reported using a toxicological equivalence factor and are therefore difficult to compare with today’s values. In view of these difficulties and having in mind, that the goal was to evaluate if a 90% emission reduction would be achievable or not, it was considered to be sufficient to provide maximum emission estimates for 1985. Any source type with a reduction rate less than 90% compared to this maximum estimates certainly would have even less reduction rates compared to any lower (and probably more realistic) 1985 estimate.

Data from central European countries were gathered by co-operation with various organisations in Poland, the Czech Republic, Estonia and Latvia. In the case of Poland and the Czech Republic already emission measurement data were available which could be referred to.

The task of compiling emission data for non-atmospheric pathways was committed to a sub-contractor (AEA Technologies plc, UK). As done for the air emission inventory, source types were arranged according to the Corinair nomenclature; emission estimates were

Table 1
Overview on case studies and other sub-projects carried out within Stage II of the European Dioxin Inventory Project

Country	Institute(s)	Topic	No. of measurements	Results
A, PL, N, NL	FTU, Cracow Uni-vers., NILU, TNO	Development of a concept for a research program on domestic wood combustion in Europe	None	Research proposal submitted to DG Environment
D/EE	LUA/MoE	Oil shale power plant	Two (raw oil shale, filter dust)	Very low PCDD/F concentrations, hence no indication for dioxin emission
D	LUA	Iron foundries (cold air cupola)	>30 filter dust, 6 × 3 emission samplings	Large variation of PCDD/F in filter dust (<0.1 up to 12 µg TEQ/kg dm); stack gas conc. up to 0.2 ng TEQ/m ³
D	LUA	Domestic coal combustion (Geueke et al., 2000)	36 emission samplings (6 fuels, 2 stoves)	Between 0.1 and 10 ng I-TEQ/m ³ at 0% O ₂ , depending on fuel and stove type
D	LUA	TiO ₂ -prod.	Three emission samplings	No PCDD/F emission found
D	LUA	Combustion of sulfur containing solid and liquid wastes for SO ₂ -production.	Three process material analyses; three emission samples	No significant indication for emission of PCDD/F or their sulfur analogues
D	LUA	Diesel engine emissions (emergency power supply, modern truck diesel) (Geueke et al., 1999)	Six emission samplings	Emissions around detection limit (4.5 pg I-TEQ/m ³)
B	ISSeP	Iron ore sinter plants	Six emission samplings	Plant 1: 0.7 ng I-TEQ/m ³ ; Plant 2: 6.8 ng I-TEQ/m ³ ; total annual emission ≈28 g I-TEQ/year
DK	NERI/Dk-Teknik	Hospital waste co-combustion (Vikelsee et al., 2000)	Six emission samplings	No difference in PCDD/F emission with/without co-combustion at level 0.4 ng TEQ/m ³
GR	GSF (D)	Hospital waste incineration; electric arc furnace (EAF); olive residue drying	10 emission samplings	Incinerator >300 ng I-TEQ/m ³ ; EAF: 0.3–2.1 ng I-TEQ/m ³ ; Olive oil plant: 0.3 ng I-TEQ/m ³
P	ERGO (D)/IDAD (P)	Hospital waste incineration (two old, one new facility) (Coutinho et al., 2000); secondary aluminium smelter; secondary copper smelter; electric arc furnace	13 emission samplings	Very high emission (>80 ng I-TEQ/m ³) at one hospital waste incinerator; sec. aluminium plant ~7 ng I-TEQ/m ³ ; other plants around/below 1 ng I-TEQ/m ³
UK	AEA Technology	Study on dioxins to land and water (Wenborn et al., 1999)	None	Large range of emission estimates; probably most important source: pesticide production

derived by the top-down approach using emission factors and activity rates. A comprehensive report on this topic was accomplished by 1998 and is available in the World Wide Web for download (Wenborn et al., 1999).

3. Results and discussion

3.1. Case studies

The results of all case studies performed within Stage II are summarised in the last column of Table 1. The case studies addressed mainly the sectors of iron and steel production and health care waste incineration. Further, various industrial facilities belonging to other sectors were investigated which had a certain probability

of dioxin emissions, e.g. TiO₂ production via chlorine process, secondary aluminium and copper smelters, olive residue drying). Non-industrial sources were investigated by a comprehensive test-rig investigation of hard coal and lignite burning in single-room heating stoves; additionally a proposal was prepared covering the research needs to lower the degree of uncertainty regarding the emission estimates of domestic solid fuel combustion. A further study (Geueke et al., 1999), which also belonged to this project was carried out on PCDD/F emissions from diesel engines.

The following major conclusion can be drawn from the case study results:

- The Belgian iron ore sintering plants tested confirmed the importance of these sources for the annual

dioxin emissions in Europe. One plant, which had been equipped with a new sinter cooling facility prior to the measurements, exhibited surprisingly low emissions; at the other plant, however, emissions higher than usually found (~ 2 ng I-TEQ/m³) were detected.

- Hospital waste incinerators tested showed a large variability of emissions. Two small facilities, operated at the site of the hospitals, revealed extraordinarily high emissions. Due to their small capacity and short operation times these plants contribute only moderately to the total annual emissions, by mostly less than 1 g I-TEQ/year per site. Co-combustion of hospital waste in municipal waste incinerators—a practice being applied in several EU countries—appears to be a feasible way to go without increasing the dioxin emissions to air; however, it should be noted that this result was obtained on an emission level slightly above the emission limit; further, if this result is representative has not yet been evaluated.
- Domestic burning of hard coal and brown coal in small single-room heating stoves may cause significant emissions depending on the type of oven and fuel. Confirming results obtained by the Austrian EPA (Moche and Thanner, 1998, 2000) hard coal from Poland generates particularly high emissions. Emissions from low power single stoves (~ 5 kW) reveal to be increased compared to central heating facilities with 35 kW (Bröker et al., 1992, 1994).
- Electric arc furnaces (EAFs) and secondary metal smelters were shown to have moderate annual emissions slightly below or above 1 g I-TEQ/year per site.
- A number of further facilities suspected to have PCDD/F emissions fortunately did not prove so. This emphasises the general experience that prediction of dioxin formation and emission from reasoning about technologies and processes is difficult and maybe misleading in both directions.

3.2. Update of emission inventory—time trend

Compared to the situation described in the Stage I report considerable improvement of the data base regarding dioxin air emissions can be stated today. Fig. 1(a) and (b) indicate the state of inventory activities in the western European countries for the years 1995 and 2000, respectively. At present, there is only one country (Greece) without any known official activity to assess the national emission sources. Quite recently, inventory work started in Spain and Italy which also comprise emission measurements and first results were reported already (De Lauretis, 1999; Fabrellas et al., 1999); however, so far no data for the metal industries have been published. Also in Central Europe (not shown in Fig. 1), particularly in Poland and in the Czech Republic, quite comprehensive inventories have been compiled

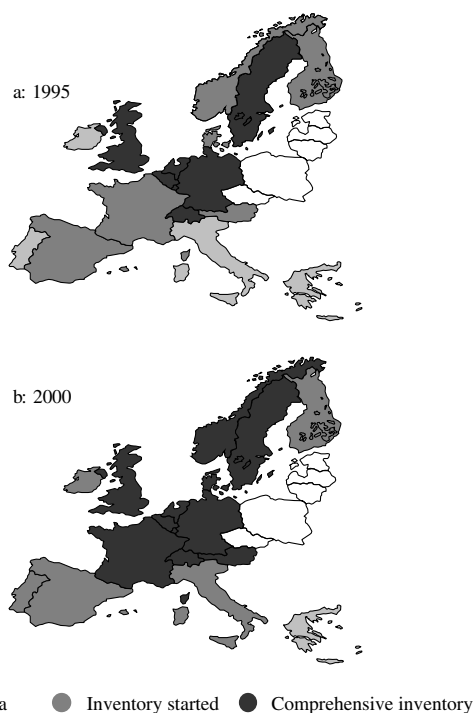


Fig. 1. State of available information about PCDD/F air emissions in Western Europe (EU 15 + Norway, Switzerland).

(Grochowalski, 1998; Holoubek et al., 2000). This work will be extended to the Baltic countries and other EU accession states in the near future (De Santi, 2001).

4. Industrial sources

Fig. 2(a)–(f) show the estimates for PCDD/F air emissions from various source types for the reference years 1985, 1995, 2000 and 2005. As outlined before, for 1985 only maximum estimates are given, whereas for the other years min–max ranges are presented.

- Iron ore sintering: The emission factor for 1985 was assumed to be somewhat (50%) higher than in 1995 taking into account that in the 1980s the use of chlorinated compounds like PCBs, which might have entered the input materials for sinter plants, was more frequent. Data for the years 2000 and 2005 were extrapolated from the time trend of sinter production rates using the 1995 emission factor of 7.5 μ g I-TEQ/t (which corresponds to 3 ng I-TEQ/m³ in the flue gases). Lower emissions due to abatement systems already installed at Austrian, Dutch and German plants have been accounted for;
- EAFs: The annual emission from this source type is comparably low; however, time trends of production

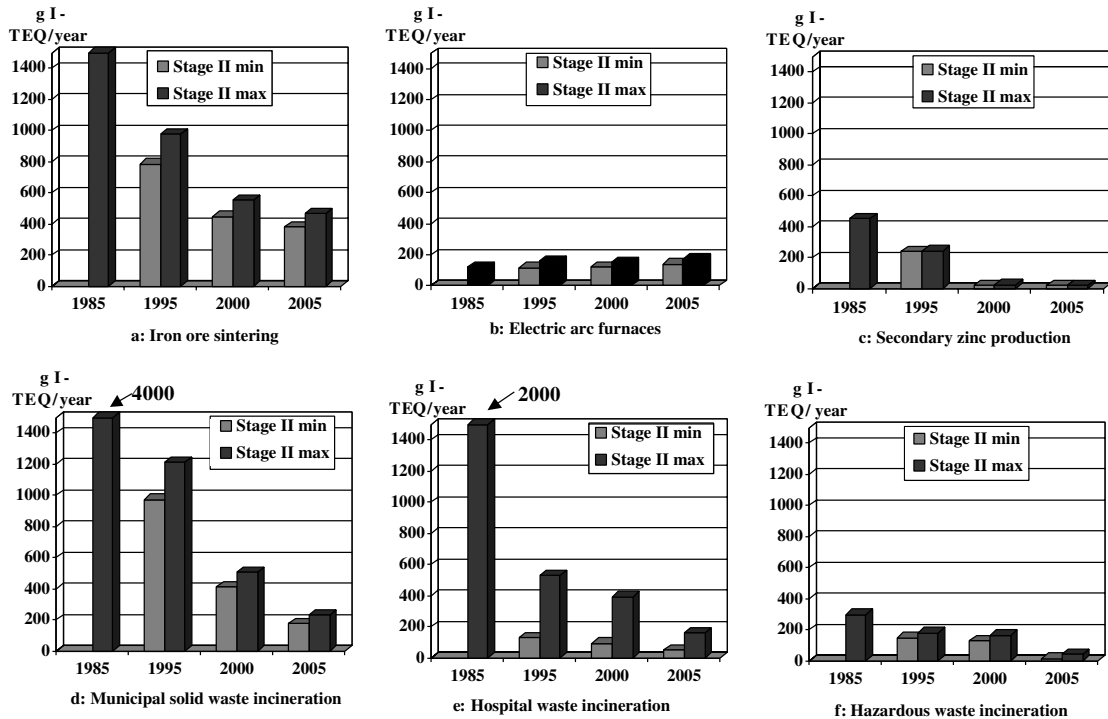


Fig. 2. (a)–(f) Time trend of estimated PCDD/F emission from various industrial sources in Europe (EU 15 + Norway, Switzerland).

and plant numbers suggest strong increasing activity. As in general no considerable dioxin reducing abatement measures are installed, the emission factor was held constant for all years, thus yielding increasing emission projections for the future. It should be noted that this is the only industrial source with positive emission trend;

- **Municipal solid waste incineration:** A dramatic decrease of the emissions by about 90% between the years 1985 and 2000 can be seen from the diagram (Fig. 2). Further decrease is likely; however, as important countries (France, Spain, Italy) started their measurement and inventory programs only recently it cannot be assumed that all installations in Europe will achieve to comply with the 0.1 ng I-TEQ/m³ emission limit until 2005. Hence an additional 50% reduction was assumed for this year compared to the present situation.
- **Hospital waste incineration:** Considerable uncertainty still exists about the PCDD/F emissions from this emission source type. By end of the 1980s the first facilities were shut down due to their high dioxin emissions which could reach several hundred ng I-TEQ/m³ (Hagenmaier et al., 1986; Bremmer et al., 1994). It may be estimated that some 1000–2000 facilities were operated in Europe by 1985 (Kempf et al., 1998). As the case studies carried out in this project showed, an annual emission value of 1 g I-TEQ/year and plant can be assumed reasonably; thus the upper estimate for 1985 was set to 2000 g I-TEQ/year. But, how many facilities comparable to the investigated installations are in operation today is entirely unknown. The emission estimates for 1995–2005 thus result from country-by-country considerations, partly based on plausibility assumptions.

• **Secondary zinc recovery:** This source is directly linked to the EAFs, since filter dust from steel production processed in special plants for zinc (and other non-ferrous metal) recovery. Four plants are said to be operated in Europe within the Berzelius company (Berzelius, 2002); two of them—located in Germany (Hiester and Radermacher, 1996) and France (France, 2000a,b), respectively—had been identified previously as very important dioxin emission sources. These facilities meanwhile have installed abatement systems (Rütten, 1999) minimizing their PCDD/F emissions. The second German plant is known to have comparably low emissions, no data are available yet for the plant located Italy. The same is valid for a Spanish installation which formerly also belonged to Berzelius but has been sold recently (Berzelius, 2000). For the 2000 and 2005 emission projections an optimistic but yet not proven approach was chosen assuming that the emissions from these installations are not comparable to their sister plants.

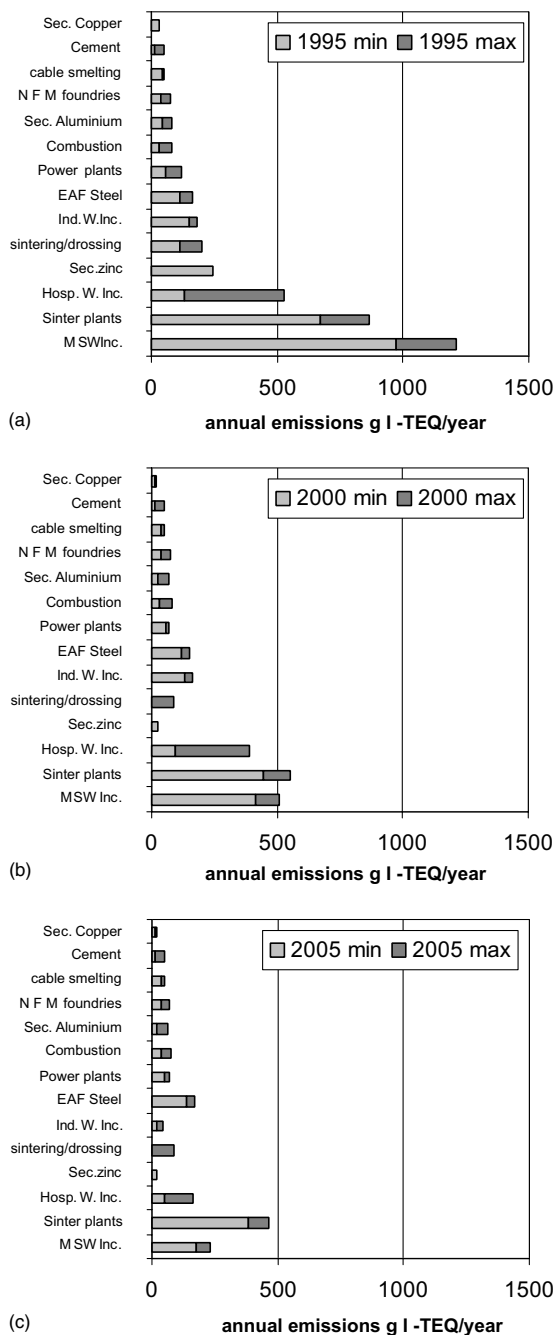


Fig. 3. (a)–(c) Ranking chart of dioxin emissions from industrial sources.

- Hazardous waste incineration: This source type has been the first which was addressed by an European directive to limit dioxin emissions (EC Directive 94/67/EC, 1994). This might be somehow surprising in view of the generally low overall emissions caused

by these plants and could be explained by the occurrence of heavy environmental pollution at some particular locations. For the estimates presented here a growing percentage of facilities complying with the emission limit is assumed.

Also for the majority of the other industrial sources considered decreasing or at least constant emissions can be stated. Taken together, the large variety and number of these other sources, (c.f. Fig. 3(a)–(c)), contribute by ≈ 200 – 400 g I-TEQ/year and are thus comparable in their effect to MSW incinerators and sinter plants. Fig. 3(a)–(c) shows the ranking of emission sources for the years 1995, 2000, and 2005, respectively. As mentioned before, lowering total emissions (decreasing from 3500 ± 650 in 1995 to $\approx 1400 \pm 350$ g I-TEQ/year in 2005) are accompanied by changes of the relative importance of the source types with the most striking feature that sinter plants will become the most important single source type.

5. Non-industrial sources

Except for the contribution of automotive traffic emissions generally much higher uncertainties exist regarding the emissions from non-industrial sources (see Table 2). Most important, emissions from domestic solid fuel combustion (wood and coal) make up more than 60% of all non-industrial PCDD/F emissions. Further large contributors are accidental fires, release of PCDD/F from pentachlorophenol treated wood products and illegal incineration of household wastes. Particularly regarding these last mentioned source types the estimates can be based only on emission factors which are derived from few studies in combination with highly uncertain activity data, large differences between min and max emission values are obtained.

In the case of domestic combustion the situation is different. Numerous studies have been performed on wood and coal burning which reveal a broad range of emission factors depending on the fuel and oven types (Bröker et al., 1994; Erken et al., 1996; Launhardt et al., 1996, 1998; Vikesoe et al., 1994; Thuß et al., 1995; Moche and Thanner, 1998, 2000); however, still research needs exist with respect to particular fuels (e.g. sea salt laden wood used in Scandinavia). Uncertainty is introduced also by unknown amounts of the different fuel types burned in single-room heating stoves or open chimneys, which generally proved to be more important than central heating appliances (Geueke et al., 2000; Moche and Thanner, 1998, 2000).

Decrease of emissions—mainly caused by lowering emissions from traffic due to decreasing use of leaded gasoline and also by increased use of natural gas and oil

Table 2
Comparison of year 1985 maximum emission estimates with year 2005 estimates for all considered source types

SNAP			1985 upper esti- mate	2005		Reduction/ increases (%)		Trend	90% re- duction likely?
				Min	Max	Max	Min		
01	Power plants	Fossil fuels	666	50	67	-92	-90	↓↓↓↓	Yes
0202	Res. combustion: boilers, stoves, fireplaces	Wood	989	523	969	-47	-2	↓	No
0202	Res. combustion: boilers, stoves, fireplaces	Coal/lignite	900	82	337	-91	-63	↓↓↓	No
0301	Combustion in industry/boilers, gas turbines, stationary engines		238	39	78	-84	-67	↓↓↓	No
030301	Sinter plants		1650	387	470	-77	-71	↓↓↓	No
030308	Secondary zinc production		450	20	20	-96	-96	↓↓↓↓	Yes
030309	Secondary copper production		29	15	17	-49	-40	↓↓	No
030310	Secondary aluminium production		65	21	60	-68	-7	↓↓	No
30311	Cement		21	14	50	-32	+137	↔	No
030326	Other: metal reclamation from cables		750	40	50	-95	-93	↓↓↓↓	Yes
040207	Electric furnace steel plant		120	141	172	+17	+43	↑	No
040309	Other: non-ferrous metal foundries		50	38	72	-25	+44	↔	No
040309	Other: sintering of special materials and dressing facilities		200	1	1	-100	-100	↓↓↓↓	Yes
060406	Preservation of wood		390	118	310	-70	-20	↓↓	No
0701	Road transport		262	41	60	-84	-77	↓↓↓	No
090201	Inc. of domestic or municipal wastes	Legal combustion	4000	178	232	-96	-94	↓↓↓↓	Yes
090201	Inc. of domestic or municipal wastes	Illegal (domestic) combustion	200	116	187	-42	-6	↓	No
090202	Inc. of Industrial wastes	Hazardous waste	300	16	45	-95	-85	↓↓↓	No
090207	Inc. of hospital wastes		2000	51	161	-97	-92	↓↓↓↓	Yes
090901	Cremation: Inc. of corpses		28	13	22	-55	-23	↓↓	No
1201	Fires		382	60	371	-84	-3	↓↓	No
Total of sources considered (g I-TEQ/year)			13 690	1963	3752	-86	-73	↓↓↓	No
Industrial sources (g I-TEQ/year)			10 539	1011	1495	-90	-86	↓↓↓	No
Non-industrial sources (g I-TEQ/year)			3151	952	2257	-70	-28	↓↓	No

for heating purpose—is much less pronounced for the non-industrial source if compared to the industrial sector. Thus, the non-industrial sector is going to become relatively more important in the near future.

5.1. Emission reduction compared to the goal set in the 5th European Action Program

From the data shown before it can be concluded, that neither the industrial sector nor the non-industrial sector taken as a whole is likely to achieve an emission reduction by 90% in 2005 compared to the 1985 situation. However, for a couple of industrial sources exceedance of 90% reduction is evident or at least probable (Table 2).

This is the case for MSW incineration as well as for hospital waste incineration, with quite high uncertainty regarding the latter. Further, also dioxin emission from power plants could be decreased considerably where substantial abatement of dust, SO₂ and NO_x emissions had been achieved. As described before, the large emissions from two secondary zinc recovery plants have been eliminated; presuming that the remaining three plants are less relevant also this sector achieved reductions by more than 90%. Finally, reclamation of metals by open-field burning of cables, a practice still frequently carried out in the 1980s (Bröker and Schilling, 1986; Hagenmaier et al., 1986; Wijnen et al., 1992), has widely been stopped.

6. Final conclusions

The case studies thus confirmed the conclusions of the Stage I report and highlight the necessity to establish a more comprehensive picture about the European emissions from sintering plants and hospital waste incinerators. To achieve this, the sinter plants not yet investigated should be subjected to emission measurements; with respect to their overall importance and in order to initialise abatement measures an obligation to monitor dioxin emissions frequently, for instance by applying semi-continuous sampling, should be established.

Regarding hospital waste incinerators an inventory of existing facilities accompanied with emission testing of installations with probable inadequate technology should be carried out.

Non-industrial emission sources will relatively become more important the near future and soon may dominate the overall annual emissions of PCDD/F in Europe. Besides educational efforts in order to inform the public about consequences when using inappropriate fuels also technical improvements, particularly with respect to solid fuel domestic heating appliances, may help to achieve further reductions.

Clearly, the strategies to reduce dioxin emissions to air developed by the EU member states and the Commission achieved appreciable success; nevertheless, a number of challenges remain and certainly some new will appear in connection with the enlargement of the European Community.

Acknowledgements

Financial support by the European Commission, DG Environment, and by the Portuguese Ministry of Environment is gratefully acknowledged. The authors further wish to thank all the people involved in the case studies carried out throughout Europe for their substantial support and flexibility which were essential for the realisation of this project.

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