

The European Dioxin Emission Inventory Stage II

Volume 1

EXECUTIVE SUMMARY

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Background

1. Background

Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/PCDF) have been of considerable public concern for more than twenty years now since large amounts 2,3,7,8-Tetrachlorodibenzo-*p*-dioxin (TCDD, „Seveso dioxin“) from a chemical production site located in Seveso, Italy were released accidentally in 1976. At this time already known to chemists as one of the most toxic artificial chemical compound for the first time „dioxin“ became a real threat for health and environment in the public’s and environmentalist’s opinion. However, most probably it would not have become as important as it is today without the discovery of PCDD/PCDF in the fly ash of waste incineration facilities suggesting a continuous and ubiquitous contamination of the environment by stack emissions [1].

Although the possibility of dioxin formation in any combustion process suited with the elements needed (chlorine, oxygen, carbon) was discussed quite early [2] for more than ten years municipal waste incinerators almost exclusively remained in the focus of research as well as of public and political concern. But, at the end of the eighties growing information indicated that other industrial and non industrial processes also may cause dioxin emissions. Comprehensive emission measurement programmes were conducted in several European countries during the first years of the 90s [3-11] leading to additional dioxin emission abatement measures in a variety of industrial sectors.

During the EU Council Conference June 1993, the German Delegation presented a memorandum to the council covering the difficulties of dioxin emissions from industrial sources with the objective of compiling the knowledge available in the member states, evaluating it and demonstrating the possibilities of limiting the emissions.

The European Commission took up the subject and implemented the project „Identification of Relevant Industrial Sources of Dioxins and Furans; Quantification of their Emissions and Evaluation of Abatement Technologies“, also called “The European Dioxin Project”. Due to the fact that important data on dioxin emissions from metallurgical and other processes had been gained from the dioxin emission testing programs of the German State of North Rhine-Westphalia [11], the State Environment Agency (Landesumweltamt, LUA NRW) was committed to carry out this project. Starting in 1995, Stage I of the project was finished in November 1997 with release of the a ca. 900-page report covering the information on dioxin emissions available from 17 European Countries (EU 15 + Norway and Switzerland) and an

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evaluation of these data to estimate the annual emissions of these countries on a comparable basis [12].

According to that study, which reflects the emission situation of the period 1993 to 1995, still the emissions from municipal solid waste incinerators appeared to be the main fraction of the overall European dioxin emission freight. Further, the emissions from iron ore sintering plants were identified as emission sources contributing to a similar extent. Considerable emissions - of the industrial facilities covered by the report - were 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, traffic and dioxin releases from PCP treated wood were assessed to be important, but less quantifiable emission sources for PCDD/PCDF.

However, it became obvious quite early in the course of Stage I that considerable data gaps still existed for a number of potential and relevant dioxin emission sources even in those countries where the problem of dioxin emissions had already been addressed. Moreover, at that time several countries in Europe had no or only few related information on their dioxin emission sources.

In view of this lack of information DG Environment decided to prolong the project in order to collect additional data which should reduce identified uncertainties, extend the scope of the project to yet less regarded aspects and to „catalyse“ a better perception of the problem. The results of this Stage II of the project are presented in the current report.

Objectives and Approach

2. Objectives and Approach

For Stage II of the European Dioxin Project the following objectives were set:

- Amendment of existing emission data collected for most relevant emission sources in order to reduce uncertainties of emission estimates
- Collecting first emission data from countries not yet performing dioxin emission measurement programs
- Extending the inventory of dioxin emissions to ambient air produced in Stage I by a complementary study on emissions to land and water
- extending the regional scope of data collection to countries in Central Europe

Addressing these objectives the approach for Stage II had to be different from that of Stage I which was carried out as a pure desk-top study involving only two parties, LUA and DG Environment. 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.

Similar to Stage I, LUA served as central co-ordinating institute being the only direct contractor of DG Environment. In Stage II, 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.

To implement the sub-projects a 1-day-workshop was held at the start-up of Stage II in November 1997. Invitations were sent to the environmental ministries of the considered countries and to known research centres or private scientific companies working in the area of dioxin testing. With regard to the demands of the European Commission also delegates from Poland, Latvia and Estonia participated in this workshop.

Among several agreements made on the workshop it was recognised that

- no further efforts should be undertaken within Stage II with respect to municipal waste incineration due to the broad range of data yet available

Structure of Report

- the main emphasis with respect to emission measurements at industrial sources thus should be put to iron ore sintering plants, hospital waste incineration, processes in the non-ferrous metal industries and cement production.
- a desk-top study should be carried out aiming at presentation of a proposal for an experimental research program designed to improve the emission data for domestic solid fuel combustion in Europe
- a further desk-top study should be made concerning PCDD/F emissions to land and water

3. Structure of Report

The report on Stage II of the European Dioxin Project is presented in 3 Volumes.

Volume 1 – this one – contains an overview on the background and approach of different activities carried out and on the results obtained. These results are put into a broader view regarding the dioxin reduction measures in Europe leading to conclusions and recommendation for future work.

Volume 2 of the report contains a detailed presentation of the sub-projects carried out. The chapters of Volume 2 are structured in a similar manner and start with a short summary in order to allow for a fast cross-reading. In the case of the desk-top studies an overview of the main results or statements is given. Regarding emission measurements details on the experimental set-up and the facilities being investigated are presented.

Volume 3 contains a re-evaluation of the dioxin emission inventory presented for the most relevant sources types in the Stage I report. New data gathered from the projects of Stage II as well as from independent activities in the European countries are considered for a revision of the 1995 emission estimates. Additionally, based on current trends and activities the PCDD/F emissions for the years 2000 and 2005 are estimated. Finally, an attempt is made to evaluate the PCDD/F emission reduction rates which might be possible to achieve by the year 2005 compared to 1985.

Despite being related to each other, any of these volumes can be read and understood as a stand-alone document. This could only be achieved by somehow redundant data presentation. For example, the summarising descriptions of the sub projects in this volume are widely identical with the short summaries provided at the head of each chapter of volume 2. Also the

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country related chapters in volume 3 in most cases give a brief summary of volume 2 results rather than just referring to the other volume.

4. Summary of Results covered by Volume 2

4.1. Desktop studies

4.1.1. Dioxin Emissions to Land and Water

The study on PCDD/F emissions to land and water was initiated when it became obvious that - compared to information on emissions to ambient air - far less data had been compiled in the national dioxin programs. Within Stage I the extension of the inventory to the other pathways of dioxin emissions was impossible; hence for Stage II a consultant was searched for who could be capable to carry out the requested evaluation. AEA Technology plc, UK, was committed finally as this company previously had prepared a similar national report for the UK environmental authorities.

The report on dioxin emissions to land and water in Europe [13] was completed by 1999 and has meanwhile been made available to the public via the internet on the DG Environment dioxin homepage¹. The source structure of the report is similar to that chosen in the Stage I report which was adapted to the source code developed for the CORINAIR² inventory. Since the estimates given in the “land and water” report are calculated for the year 1994 it is complementary to the Stage I air emission inventory.

Following, a brief summary of the main results is given:

There are 5 sources comprising nearly 90 % of the total release of dioxins to land (c.f. Table 1). The highly uncertain emissions from pesticide production (13000 g I-TEQ, 1994) and pesticide use (1600 g I-TEQ; 1994) will probably be decreasing because of improved control and reductions in the use of production of chlorinated pesticides and intermediates. Emissions from incineration of municipal solid waste of 7200 g I-TEQ (1994) are decreasing because of improved combustion and control technology. The trends for the highly uncertain emissions from accidental fires 7950 g I-TEQ (1994) and disposal of MUNICIPAL SOLID WASTE to landfill 4000 g I-TEQ (1994) are unpredictable. Other sources of dioxin releases

¹ <http://europa.eu.int/comm/environment/dioxin/>

² CORINAIR = Air module of the CORINE (CO-ordination d'INformation Environnementale) inventory program

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to land, predominantly from processes in the non-ferrous metal and in the steel industry, are less important but not negligible.

Source	Release to land (best estimate) g I-TEQ (1994)	% of Total Release	Uncertainty (High/ Medium/ Low)	Likely trends in release from 1994
Pesticide Production	13000	34 %	High	-
Accidental Fires	7950	21 %	High	?
Incineration of municipal solid waste	7200	19 %	Medium	-
Disposal of municipal solid waste to Landfill	4000	10 %	High	?
Pesticide Use	1600	4,2 %	High	-
Secondary Lead Production	1200	3,2 %	Medium	?
Combustion of Wood - Domestic	650	1,7 %	High	?
Secondary Copper Production	390	1,0 %	Medium	?
Electric Furnace Steel Plant	350	0,9 %	Medium	?
Secondary Aluminium Production	310	0,8 %	Medium	?

Table 1 dioxin and furan sources with a high potential for release to land

There are far less data available to enable releases to water to be estimated thus for most sources only an indication of whether the source is likely to release a high, medium or low emission to water is possible. The production and use of pesticides, chemical production, accidental fires (releases to water through fire fighting practices), disposal of municipal solid waste to landfill are deemed to be the highest potential sources for dioxin emissions to water.

4.1.2. Domestic solid fuel combustion

As mentioned above it became clear during the start-up workshop of Stage II that a comprehensive investigation regarding the various types of domestic solid fuel combustion in Europe was far beyond the scope and possibilities of the Stage II project. However, as all

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workshop participants noted that emissions from domestic heating appliances using wooden or coal-based fuels belong to the most important sources it was decided to carry out a desk-top study in order to provide a proposal for a related comprehensive research program. The study was conducted in international co-operation including experts from Austria, The Netherlands, Norway and Poland –countries having major interest in such a research program due to the high extent of solid fuel consumption in their households.

In the following the main content of this research proposal - as being finally submitted to DG Environment – is briefly summarised:

The proposal addresses heating facilities with central boilers (< 50 kW) and at single stoves. All together 60 on-site samplings (at stoves and stacks of residential buildings) are estimated to be necessary in order to provide a reliable data base for the assessment of dioxin emissions from domestic heating. According to the proposal, this is the minimum data base required for an improvement of the emission factors for domestic solid fuel combustion (wood, coal). Besides on-site emission measurements in households supplementary measurements at test facilities (18 samplings) are regarded as essential in order to get data on special fuels. The total cost of the outlined research project are assessed to 385.000 EURO; each 50% of this sum shall be paid by the participating institutions/national authorities and the European Commission, respectively. A period of 18 months is estimated to be necessary for the completion of the entire project.

However, during the Stage II period new evidence for the potential importance of domestic coal combustion was presented by the Austrian Federal Environment Agency (A-UBA). These results stimulated a further research activity of LUA regarding the emission factors from small, coal-fired single room heating stoves (see below and Volume 2, chapter 12). As a result of this additional measurement program a part of the research program covered by the research proposal described before already has been realised. Therefore it appears necessary to update the proposal taking the new results into account. It is anticipated that such an update will lead to a reduction of the measurements still needed. Probably, the updated research proposal may address domestic wood combustion mainly; the extent of measurements regarding domestic coal combustion presumably could be reduced considerably and thus the overall costs for the proposed research program could be lower than assessed in the original proposal.

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4.1.3. France — The French Dioxin Emission Inventory

In the Stage I report the data available on dioxin emissions in France were assessed to be scarce. Hence the French environmental authorities were invited to participate in a Stage II sub-project and a delegate from ADEME attended the start-up workshop. However, at that time the French ministry of environment already had decided to set up a national fund for a separate comprehensive emission testing program; therefore, no contract was made with the French institution ADEME within this project. Nevertheless, it was agreed that data gathered within the French measurement program may be reported in the framework of the European Dioxin Project as well. This data has been made public on the internet homepage of the ministry of environment. Further information could be gathered on occasion of a national dioxin symposium held in France in 1998. As no measurements were carried out as part of the Stage II project, the survey on the French results is considered here as a further “desktop study“ which results are summarised briefly in the following.

4.1.3.1. *Municipal solid waste incineration in France*

For municipal solid waste incineration ADEME estimated an annual emission of around 400 g I-TEQ/year from the arithmetic mean value of the emission factors. Using the published concentrations and multiplying them with a typical specific flue gas volume of 5000 m³/ton waste and with a yearly operation time of 8000 hours the annual emissions obtained are: 435 g I-TEQ/year for 1997, 350 g I-TEQ/year for 1998 and 227 g I-TEQ/year for 1999. Abatement measures as well as closures of plants have reduced the emissions from municipal solid waste incinerators in France considerably and the actual situation in 2000 is likely to improve further. For the year 2000, annual emissions are assessed to be around 200 g I-TEQ/year. It appears unlikely that all existing plants will be retrofitted until 2005 to comply with the emission limit value of 0,1 ng I-TEQ/m³, but in case the trend observed during the recent years will continue a further emission reduction by 50% seems to be realistic.

4.1.3.2. *Dioxin emissions from the French metallurgical industries*

The results of the measurements carried out at iron ore sintering plants and secondary non-ferrous metal producing facilities within the French dioxin program were published in terms of annual emission freights only. These values can be converted to emission concentrations if the specific production values of the investigated plants are known. For the six French sintering plants emissions of 93 g I-TEQ/year are reported by the French dioxin survey. Thus,

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as in other countries with integrated steel works the French iron ore sintering process constitutes a considerable emission source, too.

A number of electric arc steel works revealed to be rather strong dioxin emitting sources as well. Overall, 36 g I-TEQ/year were calculated mainly from the measurement results of 6 plants. Another 11,5 g I-TEQ were assigned to secondary aluminium production, whereas copper and lead production facilities were estimated to cause emissions of little more than 2 g I-TEQ/year each. Slightly higher emissions of 3,4 g I-TEQ/year are released from iron foundries; coke ovens and some other installations in the metallurgical industries appeared to be of minor importance. One particular facility of the non-ferrous metal sector revealed to be by far the most relevant single source, it was estimated to emit about 200 g I-TEQ/year. According to the company a three step abatement program was carried out to reduce the emissions to less than 1ng I-TEQ/m³. Measurements confirmed an emission reduction by more than 90%.

4.1.3.3. *Other French dioxin emission sources*

Regarding other emission sources a considerable lack of knowledge still exists with respect to clinical waste incineration. There are 3 incinerators especially designed for hospital waste incineration. These installations presumably apply a pyrolysis reactor combined with a post-combustion unit. Additionally, one plant for hazardous waste incineration is mentioned. About 20 plants for municipal solid waste incineration are used for co-combustion of hospital waste. Besides these plants also on-site combustion facilities might still be in operation. The number of these small facilities was decreasing considerably during the last decade from about 1400 installations in 1991 to about 400 after 1994. How many on-site facilities are operated actually seems to be uncertain. No data were published yet on the distribution of hospital waste to the different types of plants and on measurement results from on-site incinerators. Dioxin emissions from co-incinerated hospital waste were already considered within the estimate for municipal solid waste incinerators. For 1995 a revised dioxin emission level between 10 and 50 g I-TEQ/year was estimated, the value from the lower end of this range being more probable due to continuing closures of small on-site incinerators and abatement measures at the co-incineration plants.

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4.1.4. Dioxin data from Central European Countries (Poland, Estonia, Latvia and Czech Republic)

In view of the scheduled extension of the countries belonging to the European Community the Commission expressed strong interest in getting information about the dioxin emission situation in central European countries. Poland, Estonia and Latvia were included in the project right from the beginning as delegates from these countries participated in the start-up workshop. In the course of the project further contact was made to research institutes in the Czech Republic. The main results of these contacts are presented here as the third “desktop study” since most information was taken from related papers and documents. Only in the case of Estonia some experimental work was carried out.

4.1.4.1. Poland

In case of Poland, the main emphasis is put on hospital waste incinerators. About 45 % (ca. 85.000 t/year) from the total annual amount of hospital waste in Poland (about 187.000 t in 1997) is disposed off in landfills. The remaining 102.000 t/year are incinerated in power plants, local hospital heating facilities and waste incinerators. Around 75.000 t/year are incinerated in about 300 old obsolete incinerators build between 1960 and 1980. The 300 old incinerators contribute at an average of 15 ng I-TEQ/m³ to the total atmospheric dioxin pollution in Poland. The 25 modern hospital waste incinerators emitted about 0,08 ng I-TEQ/m³ of dioxins if huge, fresh carbon filters were used. Just before replacing of used-up carbon filters the dioxin concentrations increased up to 25 ng/m³.

4.1.4.2. Estonia

A special question was tried to answer in case of the co-operation with Estonia. The Estonian thermal power station is the world largest thermal power station burning low-grade local oil shale. In preparation of a potential measuring campaign filter ash and raw oil shale dust samples were analysed first. If dioxins are formed during the combustion of oil shale they should be found in filter ash samples of the plant concerned. The analyses revealed that the PCDD/F concentrations were near the lower end of the range covered by PCDD/F filter dust contents analysed in samples from German hard coal and brown coal combustion plants (0,3 - 21 ng I-TEQ/kg). The found filter dust concentrations correspond with flue gas concentrations of well below 0,1 ng I-TEQ/m³. Therefore the results obtained for the oil shale samples neither did indicate a considerable input of polychlorinated organic compounds nor their

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formation and emission during the combustion process. Based on the results obtained it is unlikely that the oil shale combustion plant in Estonia is a relevant emission source for dioxins and furans. Therefore, no further activities were scheduled.

4.1.4.3. Latvia

From Latvia only a compilation of industrial activities which presumably may cause dioxin emissions to air was received. These include some potentially relevant sources municipal and hospital waste incinerators and (illegal) non-ferrous scrap processing installations facilities. Unfortunately, no measurements of handled materials or emissions could be carried out.

4.1.4.4. Czech Republic

In the Czech Republic comprehensive investigations took place in the recent years which led to the establishment of an emission inventory. Data are presented for the period 1990-1998 showing a steady decrease of emissions from 2,200 g I-TEQ/a down to 1.380 g I-TEQ/year, respectively. Another study [14], at least partly using the same measurement data, reports different estimates with 1990 emissions of 1.252 g I-TEQ/a and a decrease down to 770 g I-TEQ/a in 1998. Most relevant sources in both inventories are iron ore sinter plants (>70 g I-TEQ/a in 1998) and domestic burning (ca. 390 g I-TEQ/a).

4.2. Emission measurement programmes

Measurement programs were carried out in the following plants in the countries listed below:

- in **Belgium** at iron ore sintering plants located in the Walloon region,
- in **Denmark** at a municipal waste incinerator municipal solid waste with co-combustion of hospital wastes,
- in **Germany** at cold air cupola furnaces (iron/steel foundries), at various facilities suspected to emit dioxins and related compounds, at heavy duty diesel engines and regarding the problem of dioxin emissions from coal-fired domestic single room heating stoves.
- in **Greece** at an electric arc furnace steel plant, a bio-waste (from olive production) drying installation and a hospital waste incinerator,

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- in **Portugal** at hospital waste incinerators, an electric arc furnace steel plant and a secondary aluminium smelter.

In the following the results of the measurement programmes are summarised sorted by countries.

4.2.1. Belgium — Emissions from Iron Ore Sintering Plants

Emissions of dioxins and furans from two iron ore sintering plants located in the Walloon part of Belgium have been measured by a Belgian institute. One plant recently was reconstructed introducing a separate cooler and thus increasing its production capacity. The other plant, approximately half the size of the first, was measured under conditions which may be considered representative for the recent year's operation. Average flue gas concentrations of 0,7 ng I-TEQ/m³ and 6.8 ng I-TEQ/m³, respectively, were found. Overall annual releases from both plants is calculated to be 28 g I-TEQ/year. Thus, together with the emissions from the two sinter plants in the Flemish part of Belgium, a release of more than 100 g I-TEQ can be estimated for the year 1995. Meanwhile this emission has declined by approximately two thirds due to primary abatement measures at the most important Flemish facility.

4.2.2. Denmark — Co-incineration of Hospital Waste with Municipal Solid Waste

In some EU countries, e.g. Denmark, hospital waste is co-incinerated with municipal solid waste; therefore a comparative emission measurement was carried out by the Danish National Environmental Research Institute NERI in order to see if the co-combustion of hospital wastes leads to an increase of PCDD/F emissions.

Under normal operation, about 15% of hospital wastes are mixed up with industrial and municipal waste. The results of the 6 measurements for dioxins and furans showed emission concentrations slightly exceeding the 0,1 ng I-TEQ/m³ value. The average PCDD/F-concentration for co-incineration was 0,29 ng/m³ with a standard deviation of 0,12 ng/m³. The average concentration without co-incineration was 0,36 ng/m³, with a standard deviation of 0,05 ng/m³. Based on the above figures, no significant difference was detected between co-incineration (i.e. with hospital waste) and incineration of pure municipal solid waste.

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4.2.3. Germany — Measurement programs**4.2.3.1. *Dioxin Emissions from Cold wind Cupola Furnaces in Iron Foundries***

In the Stage I report several emission sources were identified — belonging to the basic metal industry — which contribute relevant amounts PCDD and PCDF to the total emission of dioxins and furans in Europe. According to available data and aside from the very important iron ore sintering plants, the less important electric furnaces used in steel production and a number of various processes in the non-ferrous metal industries play a remarkable role. All metallurgical processes emitting polychlorinated dibenzodioxins and dibenzofurans (PCDD and PCDF) in non-negligible amounts appear to use not only input materials composed of ores and additives from natural resources but also of secondary, differently contaminated scrap materials, residual matters from preceding process steps and production residues.

Another process in the iron and steel industry which fits into this criterion is the smelting process in cupola furnaces for the production of cast iron and cast steel. To this process various amounts of scrap are added depending on the desired product and quality; this practice of which purpose it is to save raw materials by re-circulating waste materials has certainly beneficial effects on the environment. As only few data on dioxin emissions from this process were available it was decided to carry out a comprehensive investigation at plants located in North Rhine Westphalia. This investigation was carried out on behalf of LUA by the Institute for Casting Technology (IfG).

The investigation comprised PCDD/F analyses of filter dusts stemming from 25 facilities; PCDD/F concentrations ranged from 0,03 to 12,4 µg I-TEQ/kg dry mass. According to the dioxin contamination of filter dusts the plants were assigned to three classes (class I: < 0,1; class II: 0,1-1; class III > 1 µg I-TEQ/kg) and six plants were selected for emission measurements of which 2 belonged to class I, 3 to class II and one to class III.

PCDD/F concentrations in the flue gases were found to be quite variable and ranged from 0,003 and 0,184 ng I-TEQ/m³. A comparison between contamination of dust collected during the emission measurements and flue gas concentrations suggested a linear correlation for 5 of the six plants. However, at the facility with the highest dust contamination the PCDD/F flue gas concentrations were much lower than expected from this correlation.

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From an extrapolation of the measurement results to all facilities being in operation in the State of North Rhine-Westphalia an overall annual emission to air of less than 1 g I-TEQ/a and thus minor relevance of cold-wind cupola furnaces was revealed

4.2.3.2. Emissions from a titanium dioxide production plant

Dioxin emission measurements were carried out at the waste gas duct of a factory producing titanium dioxide. Using a modified cooled-probe method for sampling due to extraordinary high flue gas temperatures only blank values were found by the GC/MS analysis. Thus no dioxin emission to ambient air has been found to occur at this plant.

4.2.3.3. Case study on “PCDD/F sulphur analogue compounds” (PCDTs/PCTAs)

A plant located in the Germany producing sulphur dioxide by combustion sulphur-containing waste materials (spent sulphuric acid, acidic tars and oils, organic sulphur compounds, miscellaneous coal and coke products) was suspected to cause emissions of PCDD/Fs and possibly their sulphur-analogue compounds polychlorinated dibenzothiophenes (PCDTs) and polychlorinated thianthrenes (PCTAs).

A comprehensive literature research revealed that data on these compounds is quite scarce; however, the following statements could be derived:

- Sulphur-analogues of dioxins and furans have been found in different environmental compartments; in most cases only PCDTs were detected.
- Toxicological information is equivocal; some experiments reveal dose-effect relationships similar to those observed with PCDD/Fs, others indicate that the sulphur analogue compounds are metabolised much more rapidly and therefore exhibit considerably less toxicity. From precautionary considerations the sulphur analogues shall be treated as being of similar toxicological relevance as the PCDD/Fs
- From laboratory experiments, structural considerations and from the observation, that in samples where they could be detected the level of PCDT concentrations appears to be correlated with the level of PCDD/Fs similar formation reactions and formation conditions as known for PCDD/Fs may be assumed.

Summary of Results covered by Volume 2

- No validated sampling method is available for the emission measurement of the sulphur analogues of the dioxins. Thus the methods developed for classical dioxins and furans have to be used (e.g. EN 1948).
- Analytical detection of PCDTs is difficult due to the small mass difference of these compounds compared to the dibenzo-p-dioxins of same chlorination degree; the mass difference between 1 sulphur atom (PCDTs) and 2 oxygen atoms (PCDDs) is only 0,0177 amu. Hence mass spectrometry with increased resolution (>18.000) is needed. As no ¹³C-labelled congener standards are available, only semi quantitative analysis is possible relating the sulphur analogues to standards of classical dioxins and furans. Chromatographic separation of PCDTs from PCDFs is poor; improvement seems to be possible by pre-chromatographic derivatisation to the corresponding sulfones.

To evaluate whether such emissions might be possible the company's process was analysed in detail with respect to conditions which might favour a de-novo synthesis of PCDD/Fs and PCDTs/PCTAs. Two process steps were identified which are operated at relevant temperatures between 200 and 900 °C. Hence possible emissions of PCDD/Fs and their sulphur analogue compounds could not be precluded.

Analyses were made of production residues (coke dust, fly ash and sludge); very low PCDD/F concentrations were found (< 0,1, <1, and <10 ng I-TEQ/kg, respectively). Only the sludge sample contained detectable amounts of PCDTs (11-42 ng/kg as sum of homologue concentration).

Further, stack gases sampling and analyses revealed no significant emissions of PCDD/Fs compared to a sampling blank which would correspond to 5 pg I-TEQ/m³. As well, no sulphur analogue compound could be detected in the flue gases.

4.2.3.4. *Dioxin emissions from Diesel Engines*

This part of the work had already been carried out in the interim period between Stage I and Stage II of the European Dioxin Project and was released as part of the interim progress report submitted to DG Environment covering the year 1997. The results of the study has been published meanwhile in a scientific journal [15]; nevertheless, as this work was part of the project it appears reasonable to include it in the Stage II report with respect to completeness.

Summary of Results covered by Volume 2

Starting point of the work on PCDD/F emissions from diesel engines was an equivocal assessment of this matter in the scientific literature. For each of the principle approaches - test rig measurements and tunnel experiments - results existed showing minor or high potential relevance of diesel engines for the environmental dioxin load.

In the work presented here, PCDD/F emission measurements have been carried out at two different diesel engines, the first one being a stationary engine for emergency power supply and the other being a modern heavy duty vehicle engine. It is shown that the emissions of dioxins and furans in both cases are nearby or lower than the overall detection limit for sampling and analysis ($< 10 \text{ ng I-TEQ/m}^3$). From these results a minor importance of PCDD/F emissions from diesel engines is indicated. Meanwhile, this assessment has got further support by more recent tunnel studies made in the United States and in Austria.

4.2.3.5. *Dioxin emissions from small single room domestic heating stoves*

A comprehensive investigation was made to measure the PCDD/F emissions from small single room heating stoves fuelled with different types of coal. Two stoves, typical for heating facilities of the years around 1960 and 1980 were used which operated according to the through-burning and the under-burning principle, respectively. The stoves were fed with Anthracite, hard coal briquettes, coke and brown coal from Germany, hard coal from Poland and Czech brown coal briquettes, respectively.

PCDD/F concentrations in the flue gases ranged from around 100 up to 10.000 pg I-TEQ/m^3 (at 0 % O_2). With the same fuel type, the elder through-burning oven emitted less (30-50%) than the more modern one. Lowest emissions were achieved with Czech brown coal, highest with Polish hard coal which previously had been shown to cause very high emissions in a test conducted by the Austrian Umweltbundesamt. However, the PCDD/F concentrations found in our experiments never reached the Austrian results.

4.2.4. *Greece — Measurements EAF steel plant, rotary kiln, hospital waste incinerator*

The very first PCDD/F emission measurements at industrial installations in Greece are reported. Sampling and analysis was done by an experienced German research institute in co-operation with a Greek company. Of the facilities investigated, an electric arc furnace (EAF) steel plant proved to have the highest annual emission of about 1 g I-TEQ/year. By extrapolation of this emission using the production rate of the measured plant and the

Summary of results covered by Volume 3

statistically reported entire Greek production it may be assumed that 4-5 g I-TEQ will be released to air from Greek EAF furnaces annually. A lower annual emission, but much higher flue gas concentrations exceeding the scheduled European emission limit of 0,1 ng I-TEQ/m³ by more than a factor 3000 were found at a hospital waste incinerator. This finding is a confirmation of the Stage I assessment regarding this process. Annual emissions of about 35 g I-TEQ/year can be estimated from statistical data. Finally, a rotary kiln process for drying residual materials from olive oil production was found to be negligible with regard to emissions of dioxins and furans.

4.2.5. Portugal — Emission measurements at various industrial facilities

Within a co-operative subproject being equally funded by the Stage II project and the Portuguese government and involving a German and a Portuguese laboratory a number of potential PCDD/F sources in Portugal were investigated. As far as known, the emission measurement reported here are the very first PCDD/F emission tests carried out in Portugal. These included 3 hospital waste incinerators (two old ones and one new state-of-the-art installation), an electric arc furnace steel plant, a secondary aluminium smelter and a secondary copper smelting facility. Very high PCDD/F concentrations in the flue gas of about 100 ng I-TEQ/m³ were found at one of the old hospital waste incinerators, while the second old facility had surprisingly low emissions (~2 ng I-TEQ/m³). As expected, the new facility emitted far less than 0,1 ng I-TEQ/m³. With respect to the PCDD/F mass flow rate the electric arc furnace proved to be the worst installation accounting for annual emissions of about 1 g I-TEQ. All together the contribution of the 6 plants to the annual dioxin emission in Portugal is assessed to be nearly 2 g I-TEQ.

5. Summary of results covered by Volume 3

Since the Stage I report was prepared the availability of dioxin emission data (to ambient air) improved considerably regarding some countries and certain emission source types. A number of the new data filled in gaps that had been identified in the Stage I report and thus could be taken to check the assessments and estimates presented therein. This comparison finally led to the feeling that an revision and update of the 1995 inventory presented in the Stage I report should be included in Stage II. Moreover, the Commission demanded to make an assessment on the question whether the aim of a 90% reduction (until 2005 compared to 1985) of dioxin releases is likely to be reached or not. Also this request caused an urge to revise the 1995

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assessment in order to set up a better data base for emission estimates for the years 2000 and 2005.

5.1. Revised PCDD/F emission inventories

The 1995 emission inventory is revised for the main sources identified as presented in the Stage I report. The revision is made country by country including the national information being available to date. However, for some source types more general considerations could be made being valid for all countries. These considerations are presented in detail in Volume 3. From the revised 1995 national inventories estimates for the years 2000 and 2005 are derived by taking into account trends of sector activity rates on the one hand and yet known upcoming abatement measures (country specific or on European scale) on the other hand. The country specific inventories then are integrated into an European inventory (as in Stage I the EU 15 and CH, N) which is shown below in table 2.

5.2. Development of European Dioxin emissions from 1985-2005

To get an impression whether the aim of 90% emission reduction might be achievable until the year 2005 data was generated which is intended to indicate the extent of the emissions of the year 1985. As reliable and comparable data on dioxin emission measurements for this reference year is very scarce, the emission situation can be characterised only roughly. It was attempted as far as the necessary effort was justifiable to obtain information on the activity rates for 1985; in case of uncertainties always an upper estimate was used. The same approach was applied for the emission factors which mostly were taken from the Stage I report with improvements or changes taken into account if revealed from related literature. Overall, the 1985 inventory used here for comparison with the 2005 emission forecasting must be considered as not reflecting the real situation. Hence, there is a significant likelihood that the actual emissions in 1985 were lower than those shown in table 3.

6. Discussion

6.1. Dioxin emissions to air, land and water

Already in the Stage I report - despite only scarce data was available then - it was stated that the annual dioxin emissions to air do not cover the largest fraction of the total PCDD/F emissions if all environmental compartments are taken into account. The study on emissions to land and water which was carried out as part of the Stage II project confirmed that probably

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several times the mass of PCDD/F emissions to air is contained by solid materials of different origin. Large parts of these solids must be considered as waste and consequently find their way into waste treatment processes. A large fraction of these wastes presumably is incinerated in specially designed facilities with operation conditions that guarantee dioxin destruction. Some part, however, will enter the environment when being disposed off in landfills. Further, waste treatment processes being non-destructive with respect to PCDD/Fs may lead to dioxin emissions via other pathways (dust emissions, contaminated water). Since for most waste materials no sufficient data on the different fractions exist the input of dioxins and furans to land and water can be estimated with high uncertainty only.

Thus, the inventory of dioxin emissions to land and water developed within the Stage II project can only serve as an initial step on the way to a better knowledge about the different ways PCDD/Fs enter the environment and finally the food-chain. The importance of better information about these subjects is highlighted by recent cases of food contamination which always were caused by the use of industrial waste materials in animal feedstuff.

6.2. Emission measurement results compared to the EPER limit

In **figure 1** the annual PCDD/F emissions to air are shown for those Western European plants/emission sources which have been investigated within the Stage II sub-projects or on which data could be obtained from independent work in France and Belgium, respectively. For reason of clarity not all single plant emissions available from the French data are shown; in some cases the maximum and mean emissions of the measured installations are listed. Further, the results of measurements carried out at the French municipal solid waste incinerators are not considered in the graph.

The graph is divided into two fields separating sources with emissions > 1 g I-TEQ/year from those with lower emissions. The 1 g I-TEQ threshold was chosen here according the criterion set for the European Pollutant Emission Register (EPER [16]).

All plants with higher emissions than 1 g I-TEQ/year belong to the metal industries. As outlined above, municipal solid waste incineration was not considered for measurements within the Stage II project. Based on the data published by the French ministry of environment in 1998 further 78 municipal solid waste incineration plants exceeded the EPER limit. At the time of writing this number is likely to be lower due to closures and realised abatement measures.

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Thus, the highest emission displayed in **figure 1** was found at the French zinc recovery plant; however, meanwhile significant abatement measures have been installed which reduce the emissions to a few g I-TEQ/year.

Among the facilities which most probably will fulfil the EPER criterion are the iron ore sintering plants. Of the plants listed, only the Walloon installations were measured within the framework of Stage II; data on the Flemish and French plants were taken from the programs carried out by the national authorities. All attempts to get the permission for measurements in Spain and Italy failed; in case of the Portuguese plant a measurement could have been done in the year 2000 but was rejected because this plant will be closed in 2001. In Italy a dioxin emission measurement program at industrial facilities started in 1999; however, in its first phase this program is directed to smaller installations of the steel and non-ferrous metal industries. No concrete schedule has become known about emission measurements of the Spanish sintering plants, too.

All other facilities investigated within the Stage II project do not appear to fulfil the EPER criterion. However, there is a group of sources which have calculated emissions between 0,1 and 1 g I-TEQ/a of which a number could be EPER-relevant for following reasons:

- in case of the Portuguese electric arc steel plant the two separate stacks were listed separately, too; put together, the annual emissions of this plant is nearly 0,9 g and thus reaching the EPER limit
- The actual mean flue gas concentrations might be higher than indicated by the single measurement campaigns
- several of the plants operated below their maximum capacities when the measurements were made. Extension of the operation time and/ or throughput could lead to emissions which exceed the EPER limit value.

One further aspect should be mentioned in connection with the EPER. As the ranking shown in **figure 1** demonstrates, small on-site hospital waste incinerators frequently will be disregarded as their annual emissions usually do not reach 1 g I-TEQ/year. Nevertheless these facilities must be considered as highly relevant emission sources since their flue gases are emitted near ground level. Flue gas concentrations as high as found by the Stage II measurements may cause elevated ambient air concentrations and depositions in the vicinity of the emission source. Moreover, since hospitals are often located in urban environments it is

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not unlikely that the emissions may find their way into sewage sludge after being deposited on the ground and washed out by rainfalls. In case the sludge is agriculturally used the emissions from health care waste incinerators could enter the food chain via this indirect pathway.

6.3. Actual and future developments of dioxin emissions**6.3.1. Waste incineration****6.3.1.1. Municipal solid waste incineration**

With respect to the already established abatement measures in most countries, to the general awareness of the problem and to the upcoming integrative directive on waste incineration no special project was carried out within Stage II concerning municipal solid waste incineration. Nevertheless, new information had become available from a number of countries which are worth to mention.

When finishing Stage I of the European Dioxin Project the largest “white area” on the waste incineration map was France. In this country more than 300 incinerators were operated which emissions had been investigated only on basis of a few tests at that time. Meanwhile, all larger facilities have been measured several times and not unexpectedly this sector proved to be most relevant for the overall dioxin emissions to air in France. A number of high emissive installations were shut down, others will have to upgrade or close in the near future. This presumably also applies to the numerous small incinerators which still no measurement data have been published for.

As in the case of France, new measurement results have also been published on PCDD/F emissions from municipal solid waste incinerators in Spain and Italy. Accordingly, most of these plants already comply with the 0,1 ng I-TEQ/m³ limit; thus in these countries, too, the relevance of this sector has decreased considerably.

Improvements of the emission situation can also be found in most other countries that had been assigned to considerable emissions in the Stage I report. Since 1996, all German plants must comply with the emission limit of 0,1 ng-TEQ/m³; as the annual emission measurements show this limit in fact is mostly undercut. In Belgium, too, the emission limit has set to the same value. Moreover, all plants are obliged to install a semi-continuous emission monitoring. Thereby, long-term samples covering 1 month each are taken and analysed. In case the average concentration exceeds the limit, additional short-period sampling and analyses must

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be carried out. Clearly, the Belgian regulations to date are the most stringent that can be found in Europe.

From the evaluation of current and near future emissions which is presented in detail in volume 3 of this report it follows that in the year 2005 the European emissions from municipal solid waste incineration might be dominated by the facilities located in France and in the UK (c. f **figure 2**). For both countries compliance with the 0,1 ng I-TEQ/m³ limit is not expected until 2005; in France the introduction of abatement measures started only recently while in UK still a guide value of 1 ng I-TEQ/year is in force which might prevent the emissions from decreasing more rapidly. However, in both cases the emission forecast appears more probable to be a worst-case than an optimistic estimation.

In Central Europe (at least in the countries considered in this report) municipal solid waste incineration only plays a minor role yet. Most municipal waste still is landfilled; but there are also a few incinerators in operation. PCDD/F emissions have been measured so far only at the Czech facility and with 0,5 g I-TEQ/m³ this plant is not a relevant source.

6.3.1.2. *Hospital and health care waste (hcw) incineration*

This source sector is one of the main topics the Stage II project focused at. From the Stage I evaluation considerable data gaps was revealed concerning the actual situation regarding hospital and health care waste incineration. Generally, three ways of incineration exist:

- on-site combustion of waste carried out at the location of the hospital/health care institution; usually simple furnaces without flue gas cleaning and low stack heights are used.
- Co-incineration of health care waste in installations mainly used for municipal solid waste incineration; these facilities usually are large capacity incinerators having at least some flue gas treatment systems (dedusting, scrubber)
- health care waste combustion in special purpose-made incinerators or in incinerators of hazardous waste.

From emission measurements which had been carried out in Germany and the Netherlands in the late 80's it was known that small on-site incinerators may have very high PCDD/F flue gas concentrations leading to emission factors (per tonne of waste) of more than 2000 µgI-

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TEQ. In these countries such facilities were already closed when the European Dioxin Project started; concerning the situation in other countries information was scarce.

Unfortunately, it was not possible to get a comprehensive overview on the practice of health care waste incineration in Europe. Even the working group on hospital waste belonging to the International Solid Waste Association (ISWA) did not have information covering all EC countries. Nevertheless, within Stage II a considerable improvement of the state of information could be achieved.

Of the 5 measurement projects carried out in Stage II 3 included measurements at health care waste incineration facilities. Further information about this industrial sector could be obtained for France, Norway, Poland and the Czech Republic. According to the results following statements may be done:

- Small, on-site incineration of health care waste is still a common and wide-spread practice in some European countries. This was shown for Greece, Portugal, Poland and (to a much lower extent) is the case also in France and Norway.
- PCDD/F flue gas concentrations observed at these on-site facilities may range from below 0,1 ng I-TEQ/m³ (for a facility equipped with active carbon injection) to up to 300 ng I-TEQ/m³. Most frequently concentrations between 10 and 50 ng I-TEQ/m³ were measured in the related independent testing programs.
- Despite usually low annual emissions (due to low throughput and intermittent operation) these facilities may cause problems in their vicinity because of their low stack heights.

Co-Incineration of health care waste in municipal solid waste incinerators is the most common way of health care waste treatment in some countries like France and Denmark. Some German installations also use this technology. From the measurements made at the Danish plant it is indicated that this practice may be a favourable way if high capacity municipal solid waste incinerators already exist and the fraction of co-combusted health care waste can be held on a level of some percent.

It seems clear that a large improvement regarding PCDD/F emissions from health care waste incineration took place in the late 80's and early 90's through the closure of hundreds of small on-site facilities. Due to the diversity of treatment methods, the inhomogeneous definitions of the term "hospital waste" and the lack of specific statistical data the current PCDD/F emissions from health care waste incineration is hardly to assess with certainty.

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6.3.2. Iron and Steel industry

Next to municipal solid waste incinerators installations of the iron and steel industry have been identified to be highly relevant PCDD/F emission sources. This in particular concerns the iron ore sintering plants; moreover, electric arc furnaces and foundries used for steel production from scrap were considered within Stage II.

6.3.2.1. Iron ore Sintering

It was one of start-up objectives of this project to initiate measurements at those sinter plants which no data were available from during Stage I. However, for different reasons this goal could not be reached and only the two Walloon facilities were measured within the Stage II project. Fortunately, the French and the Flemish plants were investigated in simultaneous national programs; thus data are still missing for the plants located in Spain, Italy, and Portugal. Regarding the latter, information was obtained that this plant, having the lowest capacity of all European plants, will be shut down in 2001.

In other countries, too, changes of the number or capacity of sintering plants took place during the recent years. For instance, despite successful implementation of abatement measures the German installation which in 1993 was identified as the facility with the highest PCDD/F emission has meanwhile stopped operation. Also from Spain the closure of one installation and a regional movement of another was reported. By contrast, one of Belgian plants measured within this project has been upgraded recently with a separate sinter cooler and thereby increased its production capacity by ca. 30%.

At most of the plants which already have been subject to emission measurements measures were taken to reduce the release of dioxins and furans. As an example, by means of primary measures (optimisation regarding strand speed, chloride input, bed height, millscale oil content and air-tightness of the production line) the flue gas concentrations of the British sintering plants could be decreased to values around 1 ng I-TEQ/m³ [17]. Similar concentrations were reached at a German plant by using up-to date electrostatic precipitators. Two plants in Europe located in Austria and the Netherlands [18] were equipped with a high-performance scrubbing system; long-term concentrations below 0,4 ng I-TEQ/m³ are achievable. Also in Germany two plants have been equipped with special abatement systems, the first consisting of adsorbent injection/fabric filter units installed after split of the flue gas and the second one being a prototype installation using coke injection and a consecutive

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catalytic reactor [19]. No measurement results have been made public yet for the latter design; according to publications with the adsorbent/fabric filter system the striven limit of 0,1 ng I-TEQ/m³ could not be guaranteed every time.

However, the emission measurement results obtained through the French, Flemish and EU programs revealed that the assessment of sinter plants as very important emission sources for dioxins and furans is not out of date. PCDD/F concentrations in the flue gas were found to range from below 1 to up to 20 ng I-TEQ/m³; a calculation of the annual emissions for these 10 plants yielded nearly 200 g I-TEQ/a. At the most relevant Belgian facility immediate primary measures achieved a reduction of emissions by more than 90%; whether similar success will be possible at the other installations remains to be proved.

Thus, as shown in figure 4, there will be considerable emission reduction between 1995 and 2005 in Belgium, Germany, Luxembourg, Norway, The Netherlands and Portugal while emissions are assessed to be nearly constant in Italy and France and –on a quite low level – in the United Kingdom.

With regard to Central European countries data are available on the emissions from sintering plants in the Czech Republic. Emission factors ranging from 0,05 to 20 µg I-TEQ/tonne are reported from measurements made in 1997-99. These findings fit well to the results described before and underline the importance of these industrial sources.

With respect to these results there still appears to be an urgent need for determining the PCDD/F emissions from the yet untested plants in Spain and Italy.

6.3.2.2. *Electric Arc Furnaces (EAF)*

Compared to sintering plants the electric arc furnaces are much less important emission sources. Two installations located in Greece and Portugal have been investigated within the Stage II project. PCDD/F flue gas concentrations of the primary dedusting unit of both plants were below 1 ng I-TEQ/m³; including the secondary dedusting units the corresponding annual releases can be estimated to ca. 1g I-TEQ/a. These results can be regarded as typical for electric arc furnaces. Higher emissions had been observed previously if the scrap is preheated prior to the smelting process. However, this practice appears to be of decreasing importance.

Nevertheless, electric arc furnaces are very interesting for two reasons:

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- Firstly, the capacity for this process of steel production has increased significantly in the recent years from about 30 million tonnes in 1985 to 50 million tonnes in 1995. Less striking, the number of plants increased from ca. 220 (1988) to 250 (1995) [17]. Presuming that the possibilities for emission reduction are limited this increase of activity rates leads to an increase of the estimate for PCDD/F emissions from this process. This is a trend opposing to the results obtained for all other sources considered in this project.
- Secondly, there is remarkable interconnection of electric arc steel production to the non-ferrous metal industry and in particular to zinc recovery plants. Due to increasing extent of galvanising for steel protection the filter dusts stemming from scrap recycling contain considerable amounts of zinc. Usually, these dusts are contaminated by dioxins and furans, too. Big packs filled with the filter dusts are partly used for filling cavities of closed coal mines [20]; a large fraction, however, is transported to some specialised plants for zinc recovery. These secondary zinc production sites also have been identified to be considerable sources for dioxins and furans (c.f. section on non-ferrous metal installations).

6.3.2.3. *Iron foundries*

Cold-air cupola furnaces used for production of iron and steel castings are the third type of emission sources belonging to the iron and steel industry which major attention was paid to within the Stage I project. Filter dust analyses demonstrated a wide range of PCDD/F contamination with highest value of about 12 µg I-TEQ/kg dust. Surprisingly low dioxin concentrations (up to ~0,2 ng I-TEQ/m³) were found in the stack gases of the 6 plants which were subjected to emission measurements. From these values annual emissions well below 1 g I-TEQ/a were derived for the facilities located in North Rhine Westphalia. Even without knowing the actual number of cold-wind cupola furnaces operated in Germany the contribution of iron foundries to the national dioxin emissions appears very low. However, if the dioxin contents of filter dust may be considered as a mirror of stack gas releases (which is indicated by 5 of the 6 emission measurements made) there might be some furnaces having considerably higher emissions than the majority of plants. This assumption is supported by the results reported from the French dioxin emission program. Only four of the 14

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investigated plants caused more than 90% (3,3 g I-TEQ/a) of the overall emissions (3,6 g I-TEQ/a).

Summarising, on a EU-wide scale iron foundries most probably are of minor importance for the overall annual releases of dioxins and furans to ambient air. Some particular facilities may have elevated emissions which even could exceed the EPER limit of 1 g I-TEQ/year. To identify these plants an analysis of filter dust could be a useful first step.

6.3.3. Non-ferrous metal industry

Production processes of the non-ferrous metal industry frequently were found to cause emissions of dioxins and furans. Among these, a sintering plant and a facility for zinc recovery from filter dusts (see previous section), both located in Germany, revealed to have the highest PCDD/F flue gas concentrations [11]. In Stage I, the highest annual emissions were assigned to the sinter plant, followed by secondary copper and secondary aluminium production.

This view has to be revised today because the French measurement program identified a zinc recovery facility (operating with similar processes as the German plant mentioned before) to have caused annual emissions of nearly 130 g I-TEQ/a until 1997. This emission value was based on measurements which revealed flue gas concentrations significantly above 100 ng I-TEQ/m³ (c. f. Vol. 2, chapter 4). According to operator's information, these high concentrations were not typical; mean values are said to be about 20 ng I-TEQ/m³. Meanwhile, several additional abatement installations reduced the emissions by more than 90%.

However, these results indicate that in the non-ferrous metal industry single plants or processes can be found which might cause unexpected high dioxin emissions. With respect to the zinc recovery facilities there are hints that some further plants exist in Europe. For instance, according to the Danish dioxin report [21] filter dusts from the Danish electric arc furnaces are shipped to Spain for zinc recovery. Another hint to this plant can be found in the IPPC BREF document on non-ferrous metals [22], where in chapter 5.3.7.1 a Spanish Waelz kiln is mentioned as an example for application of Waelz oxide leaching. To carry out emission measurements at this plant within the Stage II project unfortunately proved impossible and remains as an important task for the future.

Some further aspects appear to be noteworthy in this context:

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- The German zinc recovery plant also has installed flue gas cleaning systems reducing the stack gas concentrations to below 0,1 ng I-TEQ/m³. Despite this effort, PCDD/F deposition rates in the vicinity of the plant remained to be significantly higher than at reference locations [23]. This appears to be mainly due to fugitive emissions of dioxin contaminated dusts (input materials and carbon from dioxin scrubber system) which to a part are handled at the production site at open air. The actual emissions through this pathway cannot be assessed; measures to reduce them are currently explored.
- Investigations on the dioxin formation processes in electrofilters of iron ore sintering plants recently led to the result that dioxin breeding may be inhibited considerably by an increase of alkalinity of the materials. Further tests using the input material for zinc recovery confirmed this effect [24].

Besides these particular facilities, there is a larger number of installations in the European non-ferrous metal industry using a variety of production processes [22]. Each of these plants may cause low or moderate dioxin emissions which in total result in a considerable environmental load. An example is the secondary aluminium smelter that has been investigated in the Portuguese sub-project of Stage II. On the other hand the low emission found at the secondary copper production facility in Portugal demonstrates that the application of generalised emission factors may also lead to considerable overestimation of the actual situation.

Thus, with respect to most countries there is still considerable lack of information regarding PCDD/F emissions from the large variety of installations of the non-ferrous metal industry (metal foundries, dressing facilities). There is a non-negligible probability that many emission sources in the European non-ferrous metal have not yet been identified. To account for this, in the emission inventories presented in Vol. 3 a standard PCDD/F maximum emission estimate of 5 g I-TEQ/a per each country has been assigned to the source “special sintering/dressing (SNAP 04 03 09). A graphical display of the emission estimates calculated for the years 1995/2000/2005 is shown in **figure 5**; the data shown is the sum of all estimates which can be assigned to the non-ferrous metal industry. As the figure reveals the overall emissions were dominated by the plants located in Germany and France which meanwhile have reduced their PCDD/F emissions significantly. Further hints about actual or potential dioxin releases from the various installations and processes as well as an comprehensive evaluation of feasible abatement technologies are presented in the NFM-BREF document [22].

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Regarding Central Europe almost no PCDD/F emission data have been published so far for the non-ferrous metal sector. In spite of being quite comprehensive even the Czech dioxin inventory appears to contain no data from non-ferrous metal production facilities. The compilation of potential emission sources in Latvia comprised “illegal lead smelters” which recover this metal from scrap. Many small, presumably “barrel smelting” plants are told to exist. These plants might be comparable in their environmental impact to the small cable-burning companies that existed in Western Europe until the 80’s. It appears reasonable to assume such practices to be widespread in Central and Eastern Europe. Therefore, a huge data gap exists with respect to this industrial sector.

6.3.4. Non-industrial sources

6.3.4.1. Domestic solid fuel combustion

Among the non-industrial sources for dioxins and furan emissions to ambient air domestic combustion of solid fuels plays an important role. This was taken into account within the Stage II project through the preparation of a research proposal aiming at a more exact determination of basic emission factors as well as through the measurement program on coal burning in small single room heating stoves.

The results of the emission measurements in connection with similar experiments carried out in Austria lead to a reconsideration of domestic coal burning as generally elevated emissions were found to occur with the small stoves compared to larger central heating facilities. The revision reveals that despite decreasing extent in most Western European countries still domestic coal burning may cause emissions of 100 to 300 g I-TEQ/a (c.f. **table 2**). For the year 1985 an upper estimate of ca. 900 g I-TEQ was derived (c.f. **table 3**). Taken into account that the use of single room heating stoves already was declining at that time it appears likely that even much higher emissions from domestic coal burning may have occurred in the past. In the future the emissions from coal burning are likely to decline further because of increasing usage of oil and natural gas for domestic heating purposes (c.f. **table 2** and **figure 6**).

However, the actual emissions cannot be estimated with certainty because the emission factors strongly depend on the type of coal used and there is still a number of fuels which has not been investigated. Moreover, a detailed statistical survey on the use of coal with

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differentiation for provenience of fuel and type of combustion facility is not available for the EU.

The same limitations apply to the usage of wood and wooden materials. Without a better knowledge on the actual amounts of contaminated wood (like plastic coated furniture wood and wood wastes preserved by pesticides) neither an exact statement on the current emissions nor on the likely time trend could be made. Thus the approach chosen in the Stage I report was maintained and no substantial change of the emission estimate derived thereby is forecasted here (c.f. **table 2** and **figure 7**).

6.3.4.2. Traffic

For nearly 20 years there has been a debate on the relevance of PCDD/F emissions from internal combustion and diesel engines. Evidence was found that combustion of leaded gasoline fuel caused elevated emissions for polychlorinated and polybrominated dioxins and furans; this was traced back to the so-called “scavenger-compounds” which are added to the fuel to prevent the engine from being soiled by lead precipitates. Consequently, the use of scavengers was prohibited and programs to reduce the usage of leaded fuel down to zero were started in some countries. Therefore, PCDD/F emissions from internal combustion engines have experienced a steady decrease during the last decade.

In the case of diesel combustion the situation was more complicated as test rig and tunnel measurements conducted in various countries led to contradictory results, some indicating a high potential for PCDD/F generation while others showing diesel engines to be of minor relevance (for details c. f Vol. 2, chapter.11). The results of the measurements made within the Stage II support the latter view which meanwhile was confirmed further by new tunnel studies made in the United States and in Austria, which found a measurable, but comparatively low emission to occur.

It can therefore be expected that PCDD/F emissions from traffic will further decrease as the usage of leaded fuel is declining. When leaded fuel will have been faded out in Europe the remaining PCDD/F emissions from traffic will depend on the extent of diesel combustion; taken the trend of increasing on-road transportation into account also PCDD/F emissions might again increase, however contributing only by some percent to the overall “background emissions” caused by the non-industrial sources.

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6.3.4.3. Other sources

The role of fires, whether occurring accidentally or - as in the case of bonfires, stubble burning and landfill fires - being started intentionally still remains a matter of concern. As outlined in Vol. 3 (chapter 1) a reliable assessment of the dioxin releases to air stemming from accidental fires is difficult to achieve. There are indications that the approach chosen for the Stage I report has led to overestimation; for the revised inventory presented in this report therefore the emission value given in the Stage I study is taken as an upper estimate. However, as these estimates are derived by capita-related emission factors a more exact evaluation of fire statistics (if available) might possibly reveal that even the lower estimate is too high.

On the other hand, the dioxin emissions caused by intentional fires appear to be still considered inadequately. "Illegal" waste combustion (also called backyard burning), may occur mainly in rural areas to considerable extent throughout the year and especially on occasion of bonfire events which frequently are good opportunities to get rid of materials (like contaminated wood waste) which otherwise had to be disposed off at higher costs [25, 26]. Very high PCDD/F emission factors have been demonstrated in the United States for the common practice of "Barrel burning" [27]; despite no statistical information is available about, it appears reasonable that similar activities may occur also in Europe, maybe predominantly in the southern countries. Moreover, landfill fires might be an even more important emission source for certain European areas [28]

Finally, a considerable annual PCDD/F emission has been assigned to volatilisation of dioxin and furans from in-use wood materials which had been impregnated with PCP or PCP salts when sold. Despite scientific evidence exist for this process [29] the extent of PCDD/F emissions to ambient air is almost impossible to assess exactly. No improvement of the data basis occurred since the Stage I report was released; therefore still the estimate given for the Netherlands can be taken as the only indicator for the EU wide PCDD/F emissions. Accordingly, as shown in table 2 they might be in the order of 200 g I-TEQ/year, slowly decreasing due to reservoir depletion.

Conclusion and Recommendations

7. Conclusion and Recommendations

Stage II of the European Dioxin Project being executed between the years 1997 and 2000 comprised of a number of sub-projects which aimed to bring light into several data gaps that had been identified in Stage I. In the following, main conclusions and recommendations for further investigation or actions shall be given.

7.1. Emission Inventories

Inventories on PCDD/F emissions to air to date have been presented from most Western European countries. Importantly, comprehensive inventories based on emission measurements are still missing for Greece and –except for municipal solid waste incineration – for Spain and Italy. First measurement data regarding the Italian metal industries can be expected for the year 2001 [30]. Also Ireland, most likely having minor PCDD/F releases compared to the more industrialised countries, is going to start a multi-media inventory project soon [31].

A first inventory on dioxin emissions to land and water in Europe was prepared as part of this project and made available to the public recently [13]. Much less information than for emissions to air is available which causes high uncertainties of the emission estimates. In the case of emissions to water only a qualitative assessment could be made. However, the main potential sources have been identified; at this stage further work pointed to these sources is needed to get more reliable estimates.

Recommendations:

- Any support needed should be given to the aforementioned countries to achieve good progress in their emission inventory programs
- In order to assure comparability the PCDD/F emission inventories should use a standardised source structure, e .g as used for CORINAIR, with well defined sub-structures. Application of the EMEP/UNECE Guidebook on emission inventories [32] is recommended.
- Newly prepared or upgraded inventories would be of increased worth if at least assessments of dioxin emissions to land were derived additionally. The report on dioxin emissions to land and water in Europe could be used as a starting point and template for corresponding national studies.

Conclusion and Recommendations

- When using the emission factor approach (top-down-inventory) problems commonly arise regarding the availability of suitable activity statistics. To facilitate future work a special European statistic on industrial and non industrial emission-relevant activities (clearly assigned to the standard source structure) would be helpful. To achieve this national statistical agencies, Eurostat and people responsible for preparing emission inventories in the European countries should be linked to each other.

7.2. Waste incineration

7.2.1. Municipal solid waste incineration

The incineration of municipal solid wastes has experienced a rapid decrease of PCDD/F emissions to air due to abatement measures and plant closures during the last decade. Actually, municipal solid waste incineration no longer is the most important source type. Of the western European countries only France still has considerable emissions from these facilities, but here, too, the emissions are declining. There are some countries (Ireland, Portugal, Greece) which are going to build up their first incineration plants in the near future. It is expected that these plants will have state-of-the-art flue gas cleaning and therefore will not create significant new emissions of dioxins and furans to air. Nevertheless, depending on the type of abatement measures installed dioxin emissions via residues and waste water streams are likely to occur. These emissions (to land and water) may further increase in future if the capacities of municipal solid waste incineration were enlarged as a result of decreasing space suitable for land-filling.

Some few incineration plants also exist in Central European countries; future expansion of the incineration capacity is very probable.

Recommendations

- The emission limit of 0,1 ng I-TEQ/m³ for municipal solid waste incineration plants should be set into force as soon as possible to be applicable to all plants which currently are in the layout stage.
- As far as possible the use of dioxin-destroying flue gas cleaning devices (catalytic filters [33-35] should be considered for new installations or in case of upgrading in order to minimise the emissions to land caused by transfer of PCDD/Fs into fly ashes and filter dusts.

Conclusion and Recommendations

7.2.2. Hospital waste incineration

The very first emission measurements carried out in Portugal and Greece clearly showed that diverse industrial installations in these countries must be considered as relevant emission sources. In particular small on-site hospital waste incinerators have proven to be an inadequate technology with respect to PCDD/F formation and emissions. As the measurement at a modern on-site incinerator shows upgrading of these facilities - where feasible - with a dioxin retarding or destroying abatement system may lead to considerable improvement. The measurement carried out in Denmark indicates that also co-combustion of hospital wastes in large incinerators for municipal solid waste appears to be a convenient way avoiding additional releases of PCDD/Fs.

Small on-site incinerators for hospital wastes presumably exist in considerable numbers in Greece, Portugal, Italy and Spain; some facilities might also be operated in other Western European countries. Considerable numbers of such facilities are also known to be found in Poland and presumably further installations exist in other central European countries.

Recommendations:

- As these facilities in most cases do not exceed the EPER emission threshold value of 1 g I-TEQ/year a comprehensive inventory on these facilities including their main operation data should be generated in order to assess the actual impact such installations may have for the environment.
- Those countries still relying on the on-site incineration of hospital wastes should be supported to change to other, less emissive waste management systems and treatment methodologies. The EU is encouraged to implement a working group about this subject involving representatives of the national and local environmental authorities, hospital operators, and members of related industrial associations.

Conclusion and Recommendations

7.3. Iron and steel industry

7.3.1. Iron ore sintering

Iron ore sintering was confirmed to be one of the major sources for emissions of dioxins and furans to ambient air. The emission measurements made within the Stage II project in the Walloon part of Belgium as well as the simultaneous tests performed in the national Flemish and French programs clearly indicate that there is considerable variance of emissions between the installations. Still data from the Spanish, Italian and Portuguese plants are missing.

As revealed by the updated inventories iron ore sintering already is or soon will become the most relevant industrial sector. The importance of this source will be further enhanced by the facilities located in accession countries in Central Europe (Czech Republic, Poland).

Recommendations:

- Emission measurements at the plants still not tested should be carried out in order to be able to decide whether immediate actions for reduction of emissions would be necessary. These measurements should be carried out by experienced, independent institutes according to standardised procedures. National environmental authorities should be encouraged by the EU to convince their iron and steel companies accordingly.
- As the examples of British and some German sintering plants show PCDD/F emissions from sintering plants may be reduced considerably by primary measures (optimisation regarding strand speed, chloride input, bed height, millscale oil content and air-tightness of the production line [17]). The EU and the national authorities involved should help to spread this knowledge to the respective contacts in the iron and steel industry.
- Since small variations in flue gas PCDD/F concentrations may cause high fluctuation of the (calculated) annual emissions the usual punctual emission measurement is considered insufficient to give a reliable picture of annual emissions. Therefore an improvement of emission control should be considered. It is recommended hence to apply a semi-continuous dioxin monitoring method to control PCDD/F emissions from all sintering facilities like already done at the Belgian municipal solid waste incinerators.

7.3.2. Electric arc furnaces

Electric arc furnaces (EAFs) used for steel production from scrap is the second considerable source for PCDD/F emissions to air which belongs to the iron and steel industry. Despite

Conclusion and Recommendations

much lower annual emissions per plant than in the case of sinter plants they should not be neglected due to their larger and still increasing number. EAFs with scrap preheating were shown to have a higher probability of enhanced emissions. Effective emission reduction has become known for some plants but does not yet appear to be achieved on a broad scale yet. As revealed by the revised inventory presented in Vol. 3 production increase is likely to compensate for the emission reductions known so far. Thus, this source type might be the only relevant industrial source with constant or increasing emissions to air. However, through application of suitable abatement technologies which have already been developed to reach flue gas concentrations below 0,1 ng I-TEQ/m³ [36] this trend could be stopped in the future.

EAF filter dusts frequently are contaminated with PCDD/F; if disposed in landfills or mines they cause an emission to land; in case of (EU wide) transport to zinc recovery plants (see below) fugitive emissions during handling and transport may occur.

Recommendations:

- EAFs, at least those with scrap preheating, should be obliged to carry out PCDD/F emission measurements.
- At least for new EAFs an obligation to install best available dioxin reduction technologies should become part of the license.
- Strategies to minimise dioxin emissions through the further processing of EAF filter dust should be developed.

7.4. Non-ferrous metal industry

7.4.1. Zinc recovery from EAF filter dusts

Among the facilities of the non-ferrous metal industry those for zinc recovery from electric arc furnace (EAF) filter dusts have proven to be major PCDD/F emission sources. Facilities in France and Germany are known; further plants are likely to exist at least in Spain and Italy. However, it is not known whether the processes applied at these installations are identical to those of the French and German plants. Thus there still is considerable uncertainty about the potential emissions. Moreover, while the stack emissions of the French and the German installations actually are reduced fugitive emissions may cause high PCDD/F deposition rates in the vicinity of the plant as experiences at the German facility show. These fugitive emissions are at least in part due to open-air handling of EAF filter dusts. Since these dusts

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are shipped from the steel plants to the recycling installations, further emissions may occur during transport in case of big-pack leakage.

Recommendations

- In view of the high potential relevance an identification of all facilities for zinc recovery from EAF dusts and similar materials is urgently needed as well as the determination of dioxin emissions from these installations.
- Regarding EAF filter dust see above

7.5. Miscellaneous industrial sources

There is a vast number of miscellaneous industrial installations with small PCDD/F releases per each facility but together contributing considerably to the annual dioxin and furan emissions in Europe. Among these are secondary smelters for non-ferrous metals (aluminium, copper), iron foundries (cupola furnaces), cement production (particularly in case of wet technology and co-incineration of hazardous waste). For these emission source types generalised emission factors can be applied to assess the overall PCDD/F emissions which are to be expected. However, some individual plants with extraordinary high emissions have also been found in the past and further facilities with “outlying” emissions are likely to exist.

Recommendations:

- An evaluation of possible dioxin emissions from these “low emissive” installations can be made only by local institutions (licensing authorities) in a case-by-case consideration.
- Screening of filter dusts for PCDD/F, eventually applying recently developed, cost saving immunoassays for TEQ determination [37] could be helpful to identify installations with higher emissions
- Wherever feasible, upgrading of the abatement systems with catalytic active fabric filters [34] is recommended in order to avoid shift of dioxin emissions from air to land or water.

Conclusion and Recommendations

7.6. Non-industrial emission sources

7.6.1. Domestic solid fuel combustion

A better knowledge of the PCDD/F releases from domestic solid fuel combustion still is needed if the emission reduction potential of this sector shall be assessed. From the results of measurements on coal combustion presented in this report it is revealed that dioxin emissions from this process might have been underestimated previously. This view is supported by recent findings of UK researchers who stated that decreasing ambient air concentrations observed might have been achieved “accidentally” through changes of the domestic heating sector than by intentional abatement of industrial sources [38].

Still high uncertainties exist with respect to domestic wood combustion since the fraction of wood contaminated with chlorine containing compounds or in the worst case wood preservatives is hardly to assess. Moreover, for countries like Norway where wood used for heating purposes is exposed to air with high inorganic chlorine even burning of “clean” wood may exhibit a dioxin emission problem.

Recommendations:

- The proposal presented by FTU, Austria, which has been developed by an international working group, should be realised - after a revision for actuality - at least with respect to wood combustion
- To obtain a more detailed insight an emission inventory following the example of the recently published German’s UBA research report [39, 40] should be set up for all EU and accession countries.
- Screening methods [41] could be possibly applied to ashes and soot collected during regular chimney cleaning in order to assess the extent of (illegal) use of contaminated wood and household wastes in solid fuel ovens and open chimneys
- Better information of the consumers (e.g. through solid fuel trading companies and chimney sweepers; warnings printed on solid fuel packs) about the environmental effects the abuse of non-licensed materials as fuels for heating purposes may have could help to improve the situation.

Emission trend 1985 – 2005

7.6.2. Other sources

While future emission reduction appears likely and feasible with regard to domestic heating this is much less the case for emissions caused by **fires** and releases of PCDD/F from reservoir compartments like **PCP treated wood**. Clearly, the assessment of emissions to air from these source categories are highly uncertain and presumably will never be estimated more reliably. Fortunately, in the case of emissions from reservoirs the rate of release will decrease as the reservoirs are depleted. Regarding PCDD/F emissions from fires equivocal opinions exist if a reduction of chlorinated plastics used in buildings and products could lead to a substantial emission abatement. A high standard of precautionary fire prevention measures – in particular with respect to public and industrial buildings – probably helps to reduce the formation of dioxins and furans.

8. Emission trend 1985 – 2005

According to the assessment presented in Volume 3 of this report for those industrial processes which are considered as the most relevant emission sources the 5th EU action programme aim of a 90% reduction appears to be nearly realised. This is to a large part due to the successes regarding particular emission sources which

- already by 1985/1990 were targets of active PCDD/F-abatement policy (like power plants, municipal, hazardous and hospital waste incineration, open air cable burning, scavenger-reduction)

or

- were represented by only some individual plants with high PCDD/F-emissions where meanwhile appropriate abatement measures have been installed (as the zinc recovery plants in Germany and France, the German special sinter plant).

Thus, considerable improvement of the general situation concerning emissions to air during the last decade can be stated which is – at least to a large part - due to comprehensive abatement measures carried out in the most industrialised member states. This improvement is reflected by decreasing PCDD/F concentrations in ambient air, generally declining depositions and reduction of food, human blood and mother's milk contamination. But, with respect to the remaining industrial sources which in most cases belong to metallurgical industries considerable effort is required to further minimise dioxin and furan emissions.

Emission trend 1985 – 2005

Still, as the evaluation of PCDD/F emissions to ambient air presented in Volume 3 of this report reveals the overall emission reduction until 2005 compared to the year 1985 is not likely to reach the 90 % value. According to the data shown in table 3 the PCDD/F emissions from domestic burning of solid fuels alone make up ca. 10% of the total emission estimate for 1985; thus, even in case that there is considerable overestimation a general decrease of emissions to ambient air to below 10% of the 1985 emissions until 2005 is improbable.

There are two main reasons for missing this goal :

- PCDD/F emissions from non-industrial sources are much more difficult to assess and to regulate than emissions from industrial sources. On the basis of the limited available data it can be concluded that the rate of emission reduction achieved so far is much lower regarding non-industrial emission sources than those observed for industrial facilities.
- A number of highly relevant industrial emission sources have been identified only in the early 90s; even to date, not all European countries which have such emission sources did at least characterise the emissions.

Moreover, with regard to emissions to land and water it is even more difficult to say whether the trend is down- or upwards. Clearly, dioxin containing hazardous waste materials from industrial production which formerly was disposed off (sometimes on inadequate landfills) today mostly are incinerated. Further, production methods – particularly regarding certain chemicals like pesticides and dyes - have been altered to reduce PCDD/F formation. But, as the study on emissions to land and water shows, reliable data on the current contamination and amounts of materials entering the environment is not available. Additionally, the emission reduction probably achieved by these abatement measures might be countered by increases of solid waste materials from other industries as a consequence of improved air emissions (e.g. shift of emissions from stack gases to collected filter dusts or to waste water sludge). It should be noted, however, that in view of the pathway for dioxins to enter the food chain and the human body in most cases any shift –even if uncontrolled - of emissions from gaseous to solid and/or liquid waste media principally can be appreciated. Of course, the better way is to avoid PCDD/F formation or, if this is not possible, and to destroy these compounds by appropriate means before they enter the environment.

Tables and figures

9. Tables and figures

SNAP			Revised for 1995			Actual data 2000			Projection 2005			Change 1995/2000	Change 1995/2005
			min	prob.	max	min	prob.	max	min	prob.	max		
01	Power plants	fossil fuels	59		122	55		72	50		67	-30%	-35%
0202	Res. combustion: Boilers, stoves, fireplaces	wood	544		989	532		971	523		969	-2%	-3%
0202	Res. combustion: Boilers, stoves, fireplaces	coal/lignite	92		408	86		370	82		337	-9%	-16%
0301	Combustion in Industry/boilers, gas turbines, stationary engines		32		83	34		81	39		78	0%	2%
030301	Sinter plants		671		864	447		554	383		467	-35%	-45%
030308	Secondary zinc production		242		245	22		25	20		20	-90%	-92%
030309	Secondary copper production		31		33	15		17	15		17	-50%	-50%
030310	Secondary aluminium production		41		82	27		72	21		60	-20%	-34%
30311	Cement		14		50	13		49	14		50	-2%	0%
030326	Other: metal reclamation from cables		42		52	40		50	40		50	-3%	-3%
040207	Electric furnace steel plant		115		162	120		153	141		172	-1%	13%
040309	Other: Non ferrous metal foundries		36		78	40		74	38		72	0%	-4%
040309	Other: sintering of special materials and dressing facilities		115		200	1		86	1		86	-72%	-72%
060406	Preservation of wood		145		388	131		349	118		310	-10%	-20%
0701	Road transport		57		138	37		82	41		60	-39%	-48%
090201	Inc. of Dom. or municipal wastes	legal combustion	973		1213	412		506	178		232	-58%	-81%
090201	Inc. of Dom. or municipal wastes	illegal (domestic) combustion	129		221	126		200	116		187	-7%	-13%
090202	Inc. of Industrial wastes	hazardous waste	149		183	131		166	16		45	-10%	-81%
090207	Inc. of hospital wastes		133		530	96		392	51		161	-27%	-68%
090901	Cremation: Inc. of Corpses		11		46	9		19	13		22	-51%	-40%
1201	Fires		54		382	60		371	60		371	-1%	-1%
Total of sources considered (g I-TEQ/year)			3685 — 6470			2435 — 4660			1959 — 3834			-30%	-43%
industrial sources			2793 — 4165			1589 — 2516			1135 — 1786			-41%	-58%
non-industrial sources			892 — 2305			846 — 2144			824 — 2048			-6%	-10%

table 2 PCDD/F emission estimates (in g I-TEQ/year) for the years 1995, 2000 and 2005 as revealed from the country-related evaluations (c.f. Vol. 3)

Tables and figures

S N A P			1985		2005		Reduction/ Increases %		Trend	90% reduction likely?
			upper estimate		min	max	max	min		
01	Power plants	fossil fuels	666		50	67	-92	-90	↓↓↓↓	Y E S
0202	Res. combustion: Boilers, stoves, fireplaces	wood	989		523	969	-47	-2	↓	N O
0202	Res. combustion: Boilers, stoves, fireplaces	coal/lignite	900		82	337	-91	-63	↓↓↓	N O
0301	Combustion in Industry/boilers, gas turbines, stationary engines		238		39	78	-84	-67	↓↓↓	N O
030301	Sinter plants		1650		383	467	-77	-72	↓↓↓	N O
030308	Secondary zinc production		450		20	20	-96	-96	↓↓↓↓	Y E S
030309	Secondary copper production		29		15	17	-49	-40	↓↓	N O
030310	Secondary aluminium production		65		21	60	-68	-7	↓↓	N O
30311	Cement		21		14	50	-32	+137	↔	N O
030326	Other: metal reclamation from cables		750		40	50	-95	-93	↓↓↓↓	Y E S
040207	Electric furnace steel plant		120		141	172	+17	+43	↑	N O
040309	Other: Non ferrous metal foundries		50		38	72	-25	+44	↔	N O
040309	Other: sintering of special materials and dressing facilities		200		1	86	-100	-57	↓↓↓	N O
060406	Preservation of wood		390		118	310	-70	-20	↓↓	N O
0701	Road transport		262		41	60	-84	-77	↓↓↓	N O
090201	Inc. of Dom. or municipal wastes	legal combustion	4000		178	232	-96	-94	↓↓↓↓	Y E S
090201	Inc. of Dom. or municipal wastes	illegal (domestic) combustion	200		116	187	-42	-6	↓	N O
090202	Inc. of Industrial wastes	hazardous waste	300		16	45	-95	-85	↓↓↓	N O
090207	Inc. of hospital wastes		2000		51	161	-97	-92	↓↓↓↓	Y E S
090901	Cremation: Inc. of Corpses		28		13	22	-55	-23	↓↓	N O
1201	Fires		382		60	371	-84	-3	↓↓	N O
Total of sources considered (g I-TEQ/year)			13690		1959	3834	-86	-72	↓↓↓	N O
Industrial sources			10539		1007	1577	-90	-85	↓↓↓	N O
non-industrial sources			3151		952	2257	-70	-28	↓↓	N O

table 3 1985 upper emission estimate compared to 2005 emission forecast (g I-TEQ/year)

reduction indicators: ↓↓↓↓↓ >90%; ↓↓↓↓ 60-90%; ↓↓ 30-60%; ↓ 0-30%; ↔ :min/max reduction with opposite trend; ↑: min/max both indicating increases of emission

Tables and figures

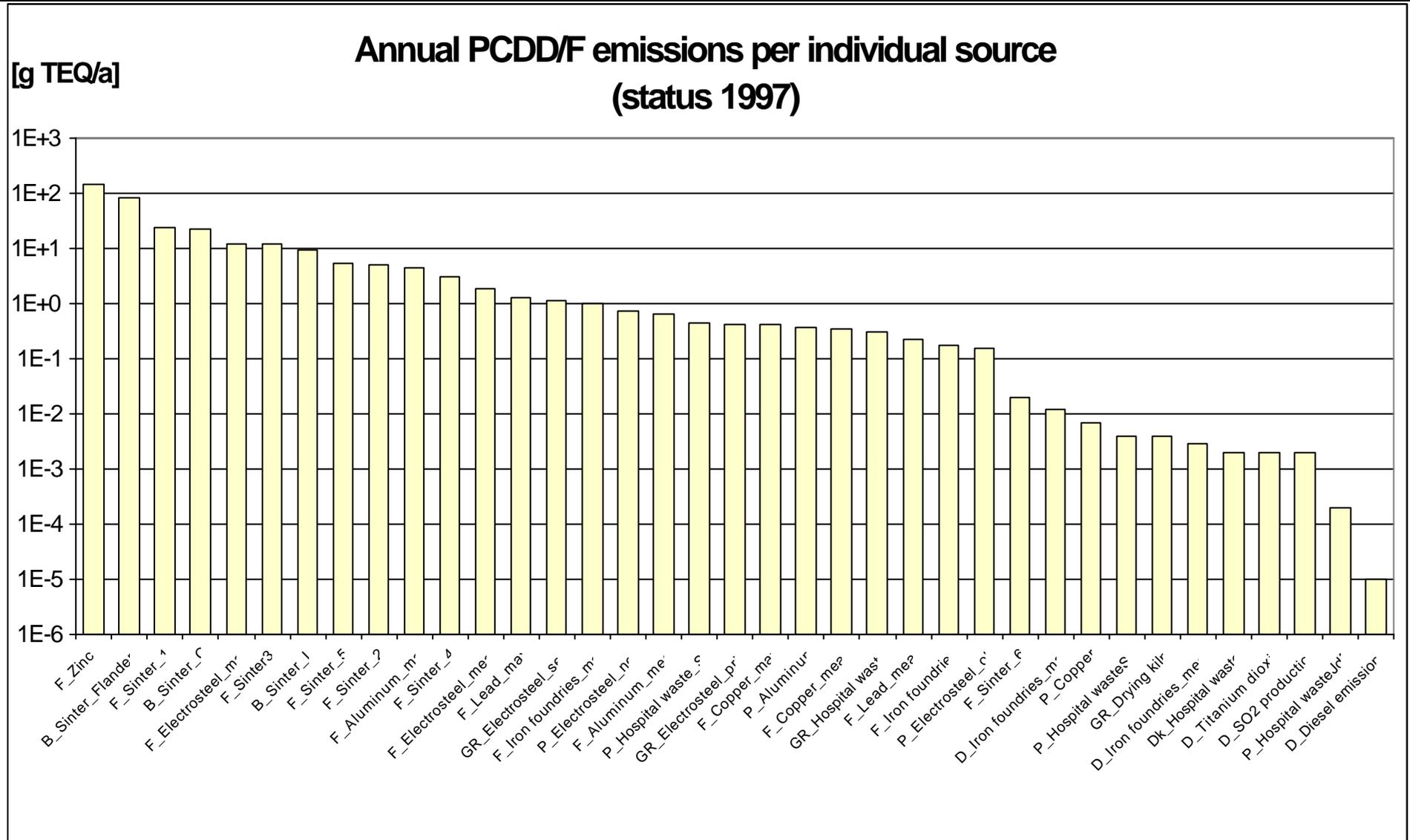


figure 1 Ranking of the Western European plants considered in this report according to their annual PCDD/F emissions to ambient air

Tables and figures

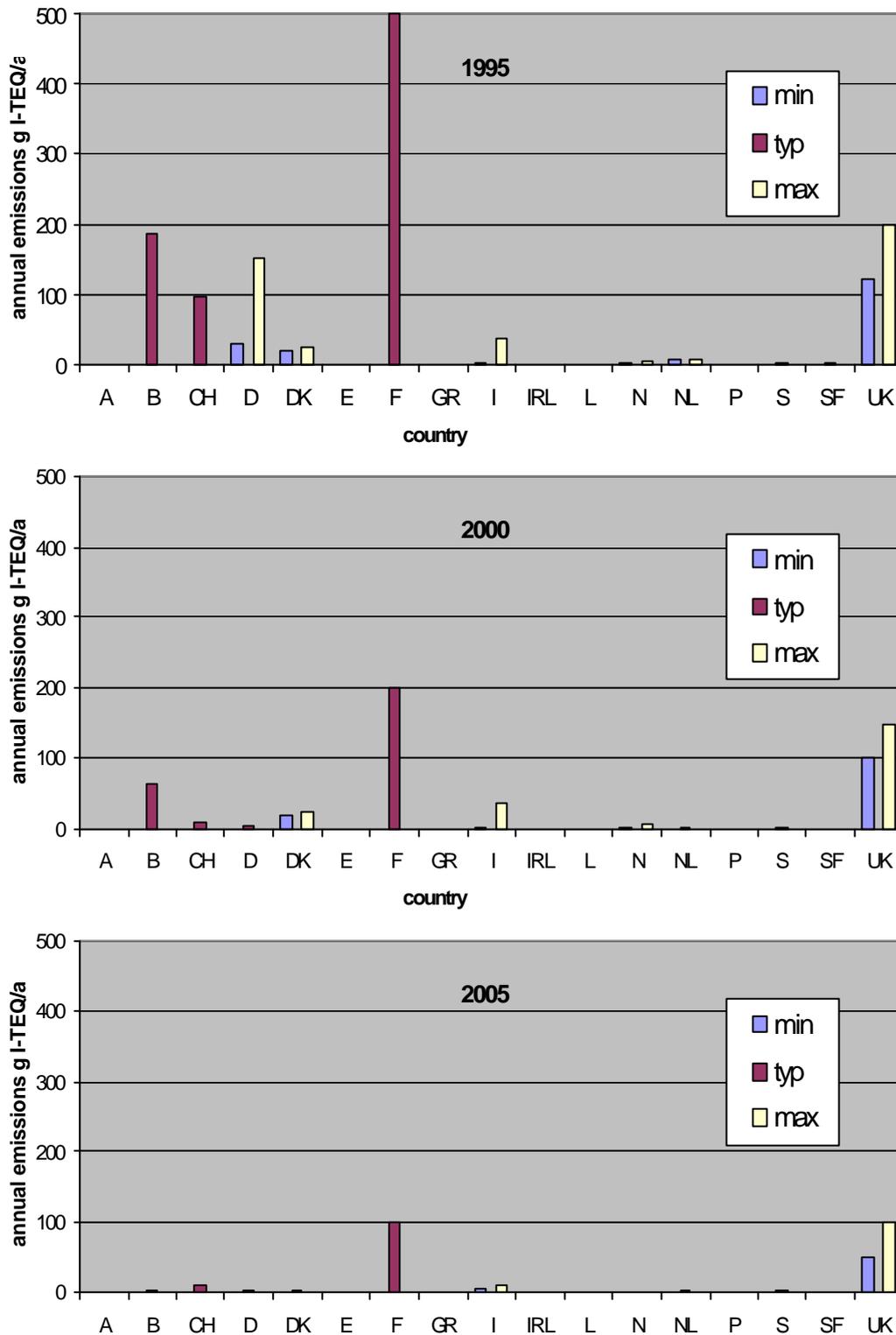


figure 2 trend of PCDD/F emissions from municipal solid waste incineration in Western Europe (as estimated in Vol. 3)

Tables and figures

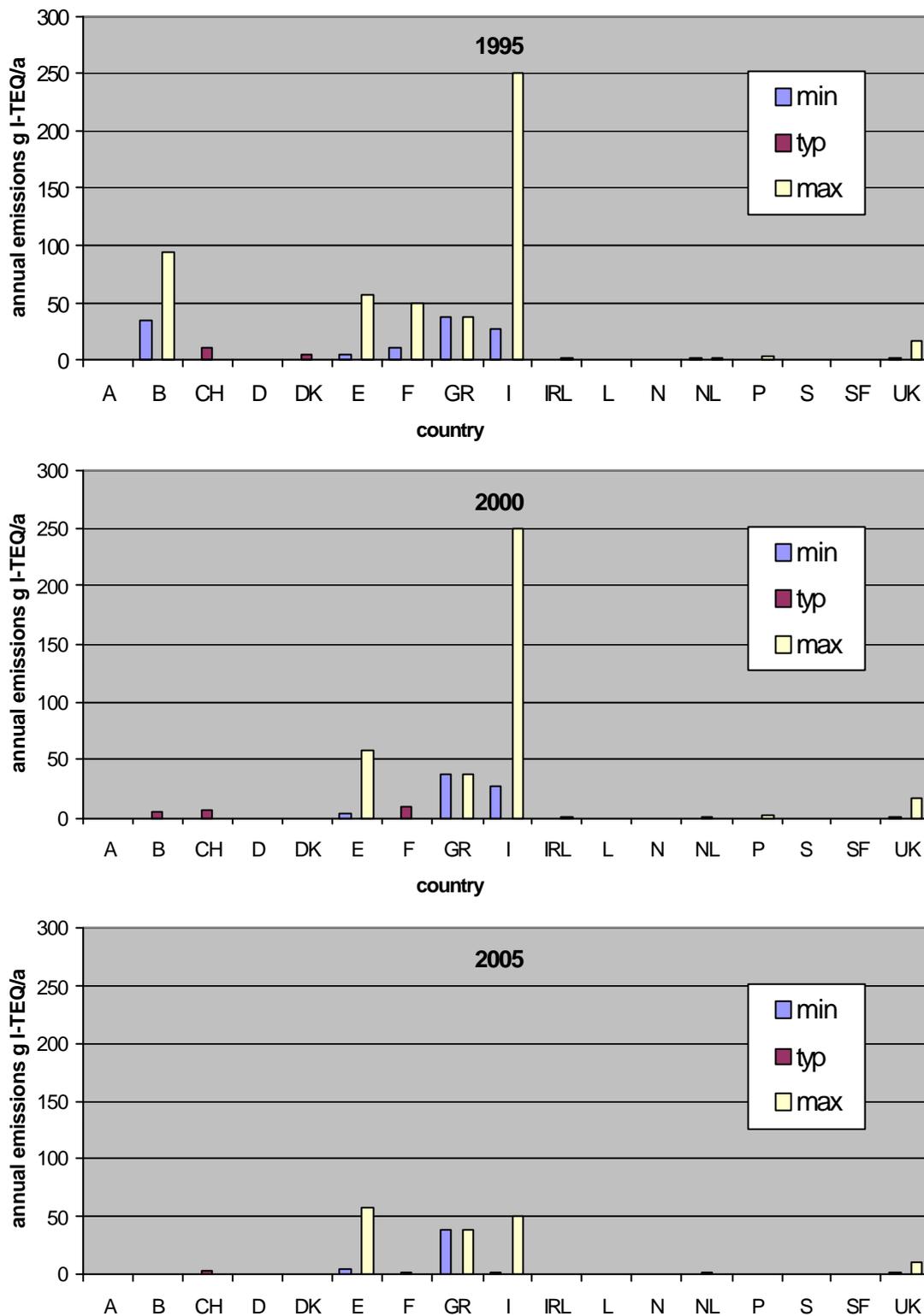


figure 3 trend of PCDD/F emissions from hospital waste incineration in Western Europe (as estimated in Vol. 3)

Tables and figures

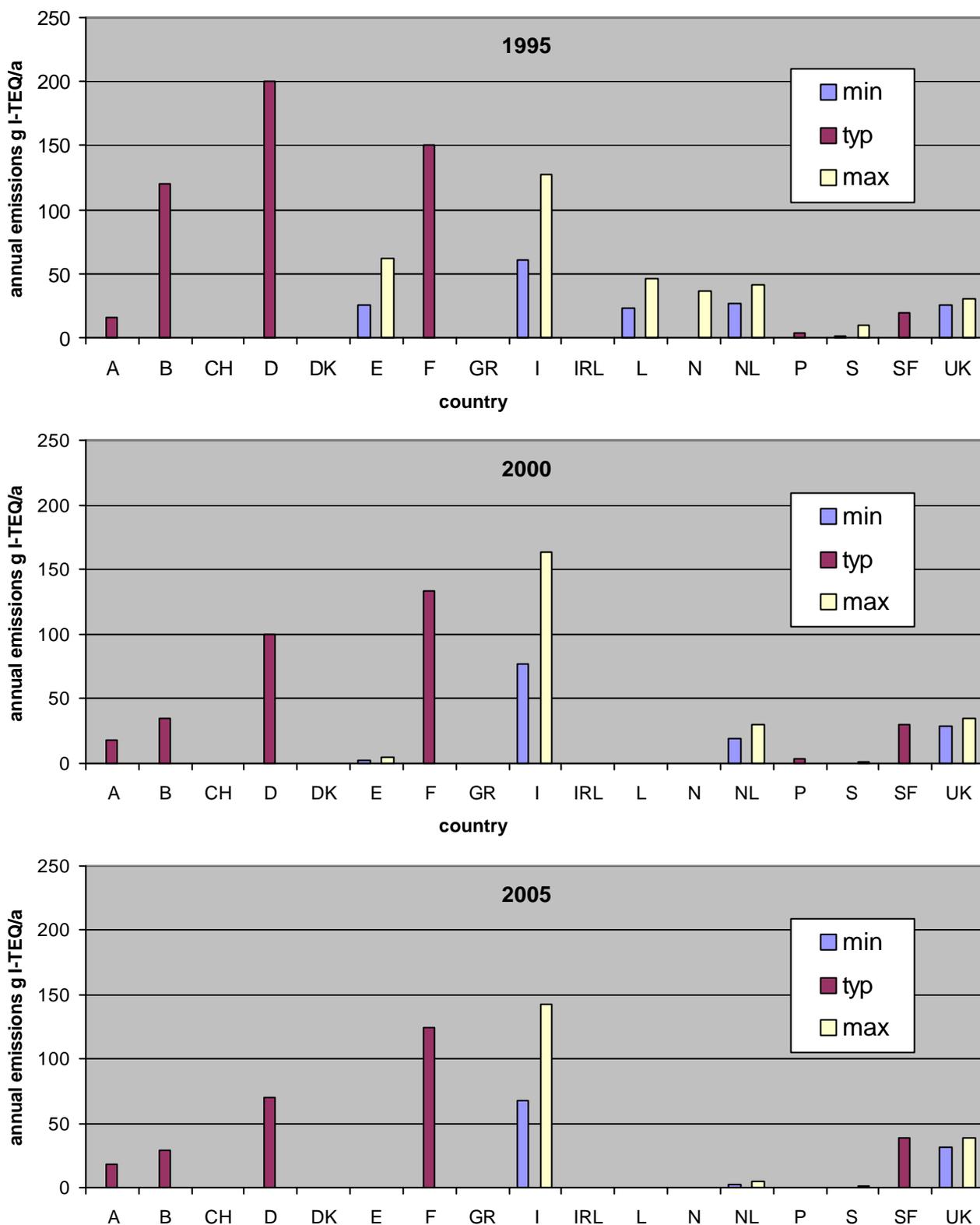


figure 4 trend of PCDD/F emissions from iron ore sintering in Western Europe (as estimated in Vol. 3)

Tables and figures

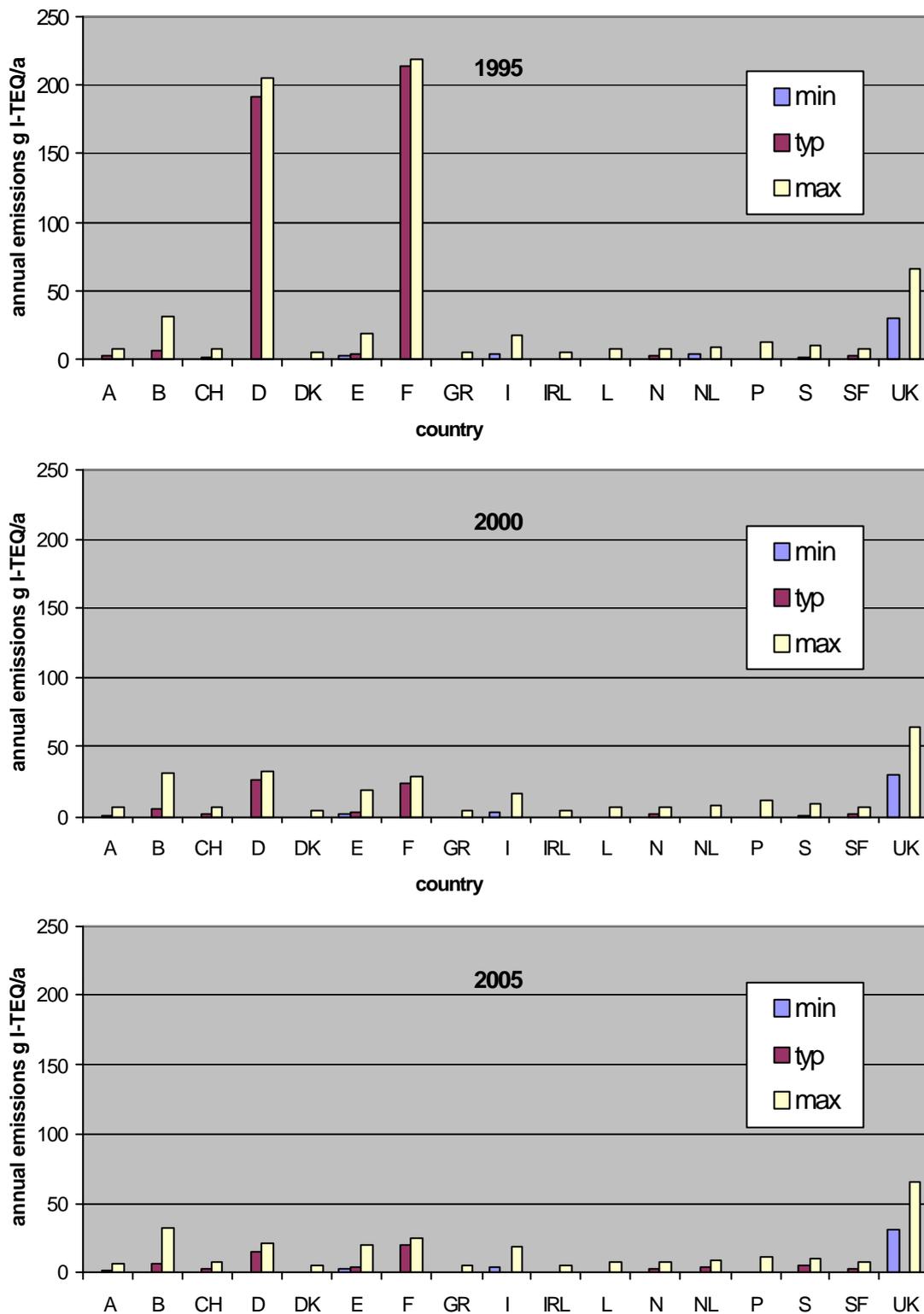


figure 5 trend of PCDD/F emissions from facilities of the non-ferrous metal industry in Western Europe (SNAP 03 03 08-10;04 03 09 as estimated in Vol. 3)

Tables and figures

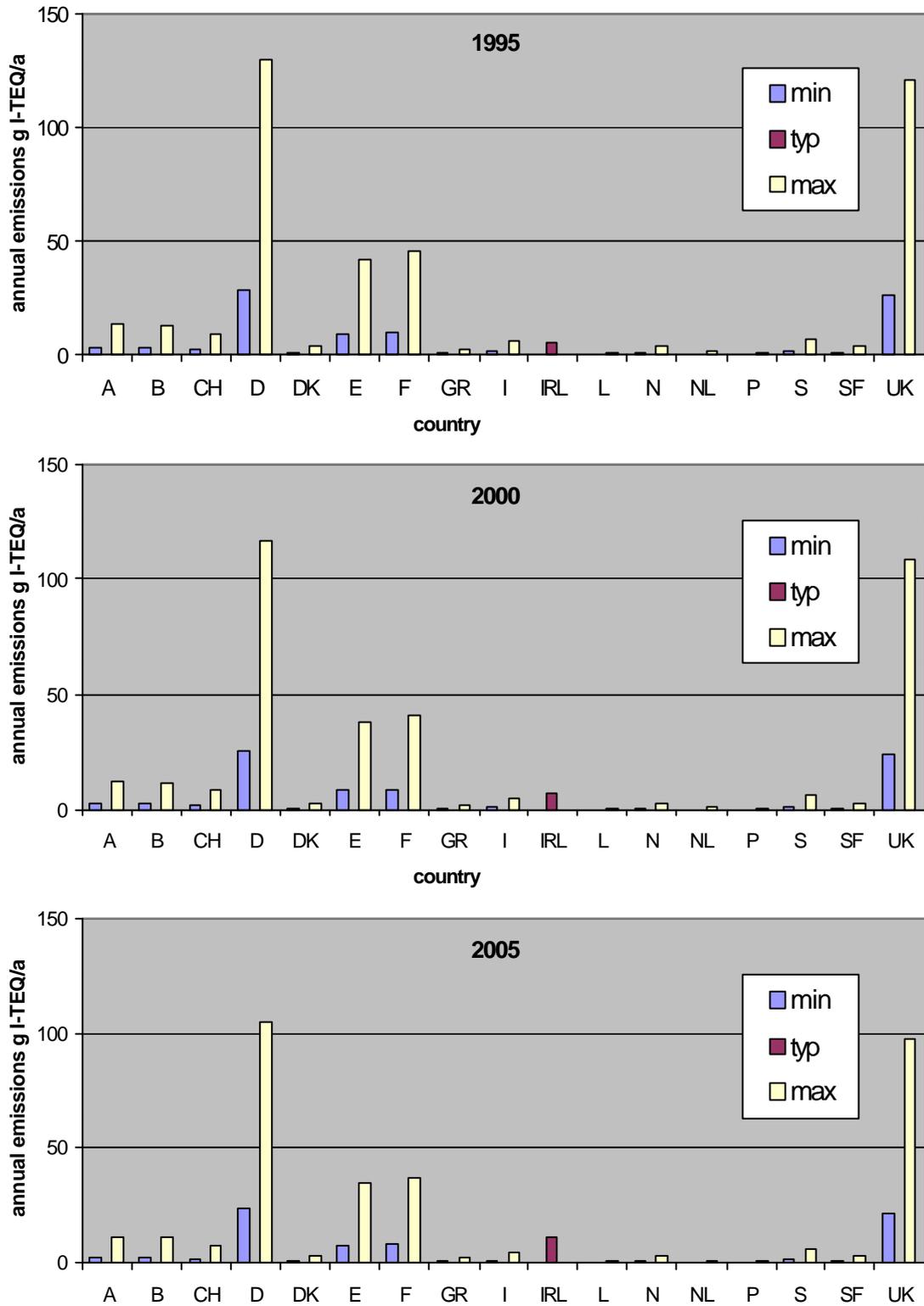


figure 6 trend of PCDD/F emissions from domestic coal combustion in Western Europe (as estimated in Vol. 3)

Tables and figures

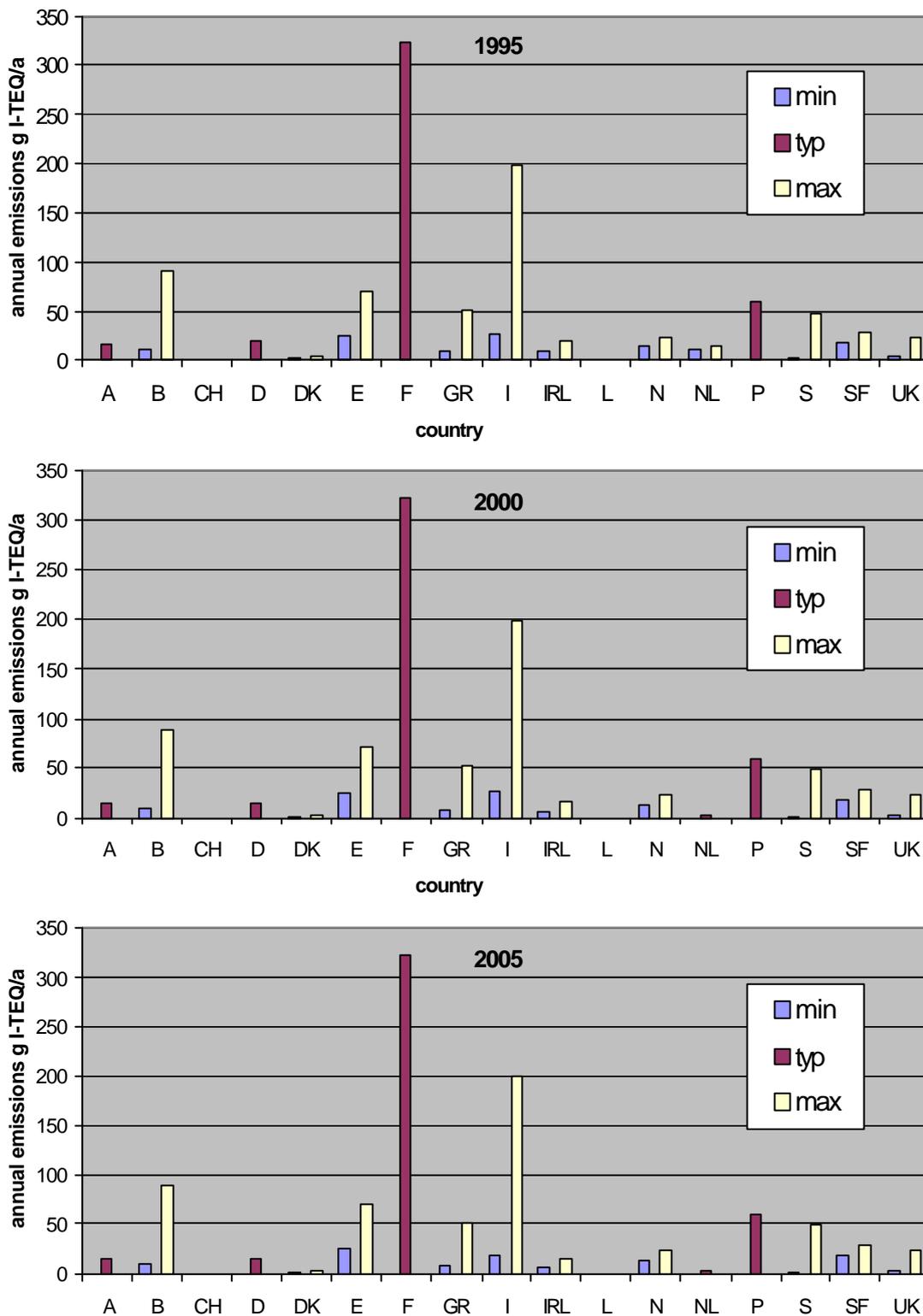


figure 7 trend of PCDD/F emissions from domestic wood combustion in Western Europe (as estimated in Vol. 3)

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References

11. References

1. K. Olie, P. L. Vermeulen and O. Hutzinger, *Chemosphere* **6**, 455-459 (1977).
2. R. R. Bumb, W. B. Crummett, S. S. Artie, J. R. Gledhill, R. H. Hummel, R. O. Kagel, L. L. Lamparski, E. V. Luoma, D. L. Miller, T. J. Nestrick, L. A. Shadroff, R. H. Stehl and J. S. Woods, Trace Chemistries of Fire: A Source of Chlorinated Dioxins., *Science* **210**, 385-390 (1980).
3. H. J. Bremmer, L. M. Troost, G. Kuipers, J. de Koning and A. A. Sein, Emissions of Dioxins in the Netherlands, 770 50 1018, National Institute of Public Health and Environmental Protection (RIVM) - Bilthoven, Netherlands Organization for Applied Scientific Research (TNO) - Apeldoorn (1994).
4. C. Ehrlich, W. D. Kalkhoff and W. Albrecht, Industrial Emissions of PCDD/F and of Dioxin-like PCBs in Saxony-Anhalt, *Dioxin '96, Organohalogen Compounds* **27**, pp. 50-55 (1996).
5. S. J. Harrad and K. C. Jones, A Source Inventory and Budget for Chlorinated Dioxins and Furans in the United Kingdom Environment, *The Science of the Total Environment* **126**, 89-107 (1992).
6. LAI, Determination of Requirements to Limit the Emissions of Dioxins and Furans, UBA-Texte 58/95, (1995).
7. O. H. Manscher, N. Z. Heidam, J. Vikelsoe, P. Nielsen, P. Blinksbjerg, H. Madsen, L. Pallesen and T. Tiernan, The Danish Incinerator Dioxin Study. Part 1., *Chemosphere* **20**, 1779-1784 (1990).
8. A. Milhaut and H. Pernin, Preliminary Dioxin Emission Inventory in France, ADEME (1996).
9. R. Orthofer and A. Vesely, Abschätzung von toxischen Emissionen (PCDD, PCDF, PAH, BaP) aus Verbrennungsprozessen in Österreich, 4554, Österreichisches Forschungszentrum Seibersdorf (ÖFZS) (1990).
10. F. Wurst and C. Hübner, Erhebung des PCDD/F-Emissionspotentials für Österreich, Forschungsgesellschaft Technischer Umweltschutz FTU (1997).
11. G. Bröker and H. Gliwa, Results of Measurements of the Emissions of Dioxins by Industrial Plants in North-Rhine-Westphalia - Dioxins Measurement programme North Rhine-Westphalia Final Report 1996, LUA-Materialien No 30, p. 106 pgs. Landesumweltamt Nordrhein-Westfalen (1997).
12. U. Quaß, M. Fermann and G. Bröker, Identification of Relevant Industrial Sources of Dioxins and Furans in Europe, LUA Materialien Nr. 43, Landesumweltamt NRW (1997).
13. M. Wenborn, K. King, D. Buckley-Golder and J. A. Gascon, Releases of dioxins and furans to land and water in Europe, AEAT-4703/20591001/Final Report -Issue 2, p. 141. (1999) available from: <http://europa.eu.int/comm/environment/dioxin/>.
14. I. Holoubek, J. Kohoutek, P. Machalek, I. Dvorakova, B. Bretschneider, J. Mitera, V. Bures and M. Fara, The Emission Inventory of POPs (PAHs, PCBs, PCDD/Fs,HC) in the Czech Republic, *Dioxin 2000*, Monterey, CA, 13.-17.08.2000. *Organohalogen Compounds* **46**, pp. 51-54 (2000).

References

15. K.-J. Geueke, A. Gessner, U. Quass, G. Bröker and E. Hiester, PCDD/F Emissions from Heavy Duty Vehicle Diesel Engines, *Chemosphere* **38**, 2791-2806 (1999).
16. Commission Decision of 17 July 2000 on the implementation of a European pollutant emission register (EPER) according to Article 15 of Council Directive 96/61/EC concerning integrated pollution prevention and control (IPPC). In *Official Journal*, 2000; Vol. L 192, pp 0036, 28/07/2000.
17. European_IPPC_Bureau, Integrated Pollution Prevention and Control (IPPC); Best Available Techniques Reference Document on the Production of Iron and Steel, European Commission, Directorate General Joint Research Center, Sevilla (1999).
18. A. Smit, T.H.P. Leuwerink, A. L. J. v. d. Panne, W. Gebert, C. Lanzerstorfer, H. Riepl and K. Hofstadler, Reduction of Dioxin Emission from Hoogovens Sinter Plant with the AIRFINE System, *Dioxin '99*, Venice, Italy, Sept. 12-17. *Organohalogen Compounds* **40**, pp. 441-444 (1999).
19. W. Schüttenhelm, R. Wemhöner, H.-U. Hartenstein and K. Werner, Reduction of PCDD/F Emissions from Iron Ore Sintering Plants - The First Full-Scale SCR Installation, *Dioxin 99*, Venedig, 12.-17.9.1999. *Organohalogen Compounds* **40**, pp. 453-458 (1999).
20. anonymus, Wider die Schwarz-Weiß-Malerei, *Umwelt-Magazin*, 40/42 (1999).
21. A. A. Jensen, Dioxins, Report No 50, 1997, DK-Teknik (1997).
22. Integrated Pollution Prevention and Control (IPPC); Draft reference document on best available techniques in non-ferrous metals industry, European Commission, Directorate General Joint Research Center, Sevilla (1999).
23. E. Hiester and L. Radermacher, Immissionsmessungen und Pflanzenuntersuchungen auf Dioxine und Furane im Umfeld einer Metallrecyclinganlage, LUA Jahresbericht '96, pp. 37-48. Landesumweltamt Nordrhein-Westfalen (1996).
24. K. Hell, L. Stieglitz, E. Dinjus, P. Segers and A. Buekens, Inhibition of PCDD/F 'De NOVO' Formation by addition of basic compounds to dust from metallurgical plants: Experimental results and discussion of inhibition mechanisms, *Dioxin 2000*, Monterey, CA, *Organohalogen Compounds* **46**, pp. 252-255 (2000).
25. R. Akkermann, Verbrennen pflanzlicher Abfälle - Brauchtuumsfeuerunbd Auswirkungen auf vegetation, Tire, Luft und Gesundheit, *natur&kosmos NVN/BSH Merkblatt* **61** (2000).
26. P. Dyke and P. Colrman, Dioxins in Ambient Air, Bonfire Night 1994, *Dioxin '95*, Edmonton, Alberta, Canada, *Organohalogen Compounds* **24**, pp. 213-216 (1995).
27. B. K. Gullett, P. M. Lemieux, C. K. Winterrowd and D. L. Winters, PCDD/F Emissions from Uncontrolled, Domestic Waste Burning, *Dioxin 2000*, Monterey, CA, 13.-17.08. *Organohalogen Compounds* **46**, pp. 193-196. University of california, Davis, CA (2000).
28. S. Psomas, Personal Communication., 2000.
29. R. Blessing, Polychlorierte Dioxine und Furane in öffentlichen Gebäuden, *Gefahrstoffe - Reinhaltung der Luft* **57**, 305-309 (1997).
30. A. M. Ghezzi, Personal Communication Nov. 2000.

References

31. P. M. L. Pat Swords, Personal Communication., 2000.
32. G. E. McInnes, EMEP/CORINAIR Atmospheric Emission Inventory Guidebook, (1996).
33. H. C. Choi, C. G. Park, H. S. Seung and T. Y. Oh, Catalytic destruction of PCDDs/DFs by the SCR units, *Dioxin 2000*, Monterey, CA, *Organohalogen Compounds* **45**, pp. 387-391 (2000).
34. R. Weber, M. Plinke and Z. Xu, Dioxin destruction efficiency of catalytic filters - evaluation in laboratory and comparison to field operation, *Dioxin 2000*, Monterey, CA, *Organohalogen Compounds* **45**, pp. 427-430 (2000).
35. M. Plinke, K. Frisky, C. P. Ganatra, M. Wilken, H. Gass, R. Weber and Y. Mogami, Catalytic dioxin/furan removal from flue gases, *Dioxin 2000*, Monterey, CA, *Organohalogen Compounds* **45**, pp. 452-455 (2000).
36. K. Hofstadler, A. Friedacher, W. Gebert and C. Lanzerstorfer, Dioxin at sinter plants and electric arc furnaces - emission profiles and removal efficiency, *Dioxin 2000*, Monterey, CA, *Organohalogen Compounds* **46**, pp. 66-69 (2000).
37. D. Brown, Y. Kishimoto, O. Ikeno, M. Chu, J. MNomura, T. Maurakami and H. Murata, Validation study for the use of the dioxin responsive CALUX (TM) assay for analysis of japanese ash and soil samples, *Dioxin 2000*, Monterey, CA, Aug. 13-17. *Organohalogen Compounds* **45**, pp. 200-203 (2000).
38. K. C. Jones, R. E. Alcock, N. J. L. Green, J. Jones, R. G. M. Lee, R. Lohmann and A. J. Sweetman, Diffuse and secondary sources of atmospheric PCDD/Fs: are they significant, *Dioxin 2000*, Monterey, CA, Aug. 13-17. *Organohalogen Compounds* **46**, pp. 39-43 (2000).
39. P. Blank, B. Wickert, A. Obermeier and R. Friedrich, Erstellung eines Emissionskatasters für Feuerungsanlagen in Haushalt und Kleinverbrauch, FB 104 02 830, Universität Stuttgart, Stuttgart (1999).
40. F. Pfeiffer, M. Struschka and G. Baumbach, Ermittlung der mittleren Emissionsfaktoren zur darstellung der Emissionsentwicklung aus feuerungsanlagen im Bereich Haushalte und Kleinverbraucher, UBA-FB 295 46 364, p. ca. 400. Universität Stuttgart, Institut für Verfahrenstechnik und Dampfkesselwesen, Stuttgart (2000).
41. D. Noger and E. Pletscher, Der EMPA-Ascheschnelltest, *Schornsteinfegerhandwerk* **3**, 19-23;34-37 (2000).

Glossary and list of abbreviations for Volumes 1-3

12. Glossary and list of abbreviations for Volumes 1-3

µg	microgram (10^{-6} g)
2,3,7,8-TCDD	2,3,7,8- Tetrachlorodibenzo-para-dioxin (“Seveso dioxin”)
a	Annum
Ahh	Aryl hydrocarbon hydroxylase , enzyme used in bioassays as an indicator for Ah-receptor activity
Airfine	Emission abatement system for iron ore sintering plants sold by the company Voest-Alpine, Austria
Alicyclic compounds	Chemical compounds with a cyclic structure build only from carbon atoms
Aroclor®	Trade name of technical mixtures of polychlorinated biphenyls
CO	Carbon monoxide
CO ₂	Carbon dioxide
CYP1A1	Specific cytochrome P 450 enzyme, induced by dioxin-like compounds through affection of the Ah receptor
CYP1A2	Specific cytochrome P 450 enzyme, induced by dioxin-like compounds through affection of the Ah receptor
d	Day
d.s.	dry substance
DNA	Desoxyribonucleic acid
Dibenzofurans	Class of chemical compounds; often used as a short form for polychlorinated dibenzofurans
Dibenzo-p-dioxins	Class of chemical compounds; often used as a short form for polychlorinated dibenzo-p-dioxins
EC	European Community
EDC	Ethylene dichloride (1,2-Dichloroethane)
EPA	Environmental Protection Agency
EROD	7-ethoxyresorufin- O -diethylase, enzyme used in bioassays as an indicator for Ah-receptor activity
ESP	Electrostatic Precipitator
Eurostat	Statistical office of the European Community
fg	femtogram (10^{-15} g)
HCW	Health Care Waste
Heterocyclic compounds	Chemical compounds with a cyclic structure build from carbon atoms and at least one other element
HWI	Hazardous waste incineration
I-TEF	International Toxic Equivalence Factor according to the NATO/CCMS list
I-TEQ	International Toxic Equivalent ; unit based on the I-TEFs and used to express the toxicity of a mixture of PCDDs and PCDFs compared to the so-called Seveso dioxin
Keramsit	Product from natural zeolithes; used e.g. for water filtration
kg	Kilogram
L.D.	Lethal dose
mg	Milligram (10^{-3} g)
Mpa	Mega Pascal , pressure unit, (10^6 Pascal)
MSW or msw	Municipal Solid Waste
n. d.	“not detectable”; “no data”, “not determined”

Glossary and list of abbreviations for Volumes 1-3

NATO/CCMS	North Atlantic Treaty Organisation/Commission for Challenges of Modern Society
Ng	nanogram (10^{-9} g)
Nm ³	Normalised cubic meter, volume of a gas at 1013 hPa and 0° Celsius
NOAEL	Non observable adverse effect level
N-TEQ	Nordic Toxic Equivalent, commonly used in Scandinavian countries to express the toxicity of a mixture of PCDDs and PCDFs compared to the so-called Seveso dioxin
PCDD	Polychlorinated dibenzo-para-dioxins
PCDD/F	PCDD and PCDF
PCDF	Polychlorinated dibenzofurans
PCDT	Polychlorinated dibenzothiophene (sulphur analogue compound of PCDF)
PCP	Pentachlorophenol
PCTA	Polychlorinated thianthrene (sulphur analogue compound of PCDD)
pg	Picogram (10^{-12} g)
POPs	Persistent organic pollutants, group of different chemicals known to accumulate in the environment; include PCDDs and PCDFs
PVC	Polyvinylchloride
Seveso dioxin	2,3,7,8 – tetrachlorodibenzo-p-dioxin
SNAP	Selected Nomenclature for Air Pollution
SO ₂	Sulfur dioxide
Sorbalit®	class of adsorbents based on lime and activated coal components
TEF	Toxic Equivalence Factor, in general
TEQ	Toxic Equivalent, in general
Thianthrene	Heterocyclic compound with structure similar to the alicyclic anthracene but with 2 sulphur atoms bridging the outer carbon rings
Thiophene	Sulphur analogue compound of furan
throughburner	Operation principle of solid fuel heating stoves; flue gases generated from the burning layer at the bottom of the fuel load flow through the fuel load before entering the chimney
Underburner	Operation principle of solid fuel heating stoves; flue gases generated from the burning layer at the bottom of the fuel load are drawn away and do not flow through the fuel load before entering the chimney
Vol.	Volume
WHO	World Health Organisation