DIOXIN AND FURAN INVENTORIES
National and Regional Emissions of PCDD/PCDF

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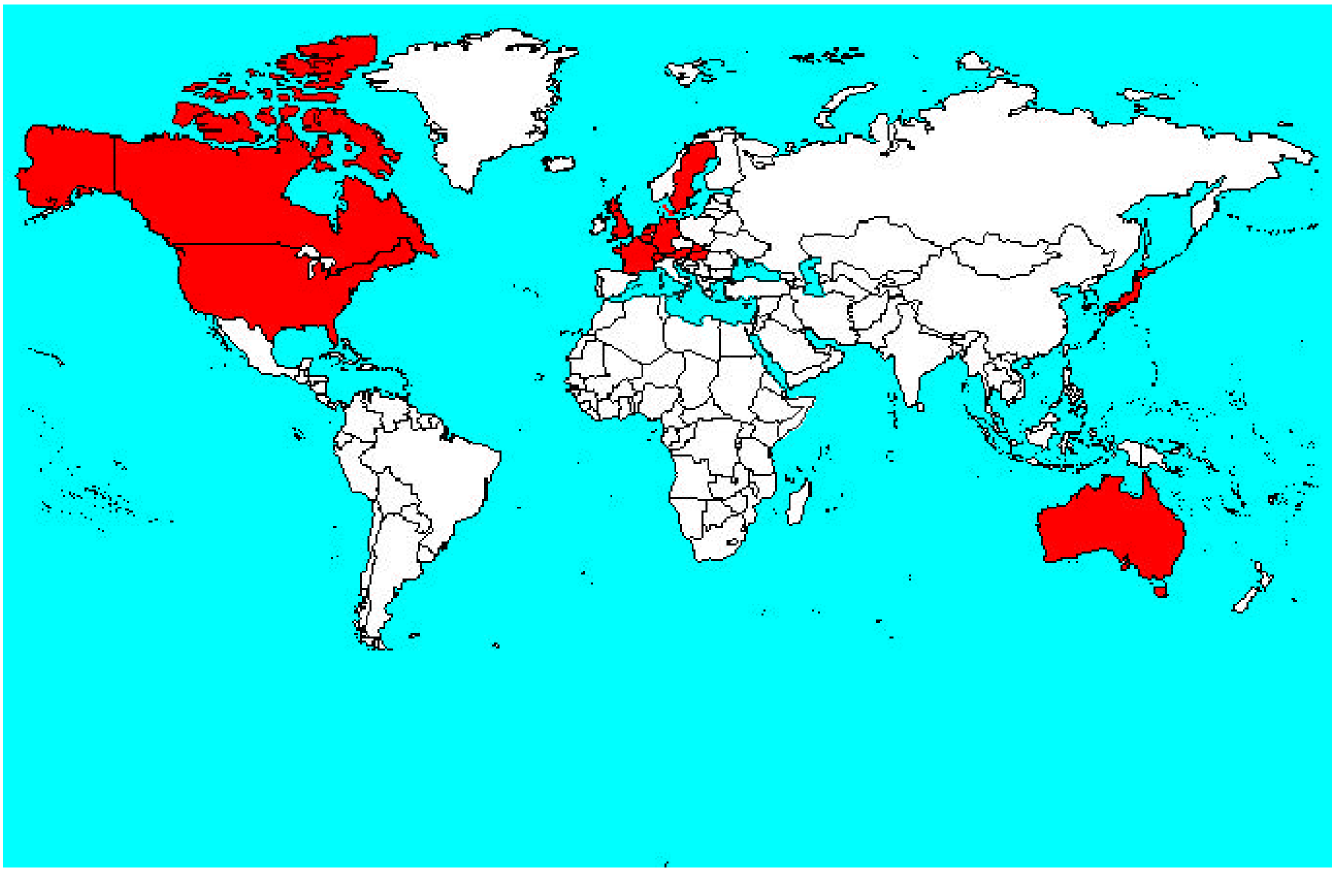

May 1999

Prepared by UNEP Chemicals
Geneva, Switzerland

IOMC
INTER-ORGANIZATION PROGRAMME FOR THE SOUND MANAGEMENT OF CHEMICALS
A cooperative agreement among UNEP, ILO, FAO, WHO, UNIDO, UNITAR and OECD

The publication is intended to serve as the first guide on available dioxin and furan inventories established for countries or regions. The information contained in this report was taken from the published scientific literature, from government reports as well as from the Internet and through personal communication. While the information provided is believed to be accurate, UNEP disclaims any responsibility for possible inaccuracies or omissions and consequences which may flow from them. Neither UNEP nor any individual involved in the preparation of this report shall be liable for any injury, loss, damage or prejudice of any kind that may be caused by any persons who have acted based on their understanding of the information contained in this publication.

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This publication was developed under contract with Dr. Heidelore Fiedler, University of Bayreuth, Germany.

On the cover: The countries in red have established dioxin and furan inventories.

This publication is produced within the framework of the Inter-Organization Programme for the Sound Management of Chemicals (IOMC)

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UNEP CHEMICALS

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Preface

In its decision 19/13 C of February 7, 1997 on Persistent Organic Pollutants (POPs), the Governing Council of UNEP requested the Executive Director of UNEP to convene an Intergovernmental Negotiating Committee with a mandate:

1. to prepare an international legally binding instrument for implementing international action on twelve specified POPs; and

2. to establish an expert group for the development of science-based criteria and a procedure for identifying additional POPs as candidates for future international action.

With this mandate to facilitate a convention on reduction and elimination of releases of POPs, UNEP Chemicals initiated several activities to help the countries to prepare for the negotiations of the convention. As one of these immediate actions, UNEP Chemicals will "…assist countries in the identification of national sources of dioxin and furan releases by promoting access to the information on available sources of these compounds…".

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF) are environmental contaminants detectable in almost all compartments of the global ecosystem in trace amounts. PCDD/PCDF have never been produced intentionally and have never served any useful purpose unlike other POPs, *e.g.* polychlorinated biphenyls (PCB) or DDT. PCDD/PCDF are formed as unwanted by-products in many industrial and combustion processes. Consequently, it is impossible eliminate dioxins and furans by prohibition of production and use. Indirect measures have to be taken to reduce the emissions of PCDD/PCDF into the environment and to minimize human exposure. Such work can be done successfully only if the sources of PCDD and PCDF are identified and quantified. Only then can technologies and measures be put in place to minimize or eliminate these sources.

Conducting inventories is an essential step in implementing an international instrument on POPs. Besides the fact that there is little information available regarding POPs produced as byproducts, technical assistance is needed for many developing countries to establish dioxin inventories. At the second session of the Intergovernmental Negotiating Committee for an International Legally Binding Instrument on POPs it was requested that the Secretariat gather and synthesize national inventory information.

Present knowledge about sources of PCDD and PCDF is based on experiences from a few industrialized countries of the Northern Hemisphere. This report is a first step towards filling the knowledge gap on existing dioxin and furan inventories. The report summarizes the information on releases of PCDD/PCDF from known sources available for far from the published scientific literature, from government reports, and based on personal communication.

UNEP is fully aware that further intensified efforts are needed to better identify and quantify sources of dioxin and furan emissions before a global dioxin inventory will be available. The information compiled here should help countries to establish their own dioxin inventories as asked in the decisions adopted by UNEP's Governing Council.

Klaus Töpfer
Executive Director
United Nations Environment
Program

DIOXIN AND FURAN INVENTORIES

National and Regional Emissions of PCDD/PCDF

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Acronyms

2,4,5-T	2,4,5-Trichloroacetic acid
AhR	Aryl hydrocarbon receptor
BC	British Columbia
Bw	Body weight
CR	Confidence rating
d.m.	Dry matter
EDC	Ethylene dichloride
EPA	Environmental Protection Agency
EU	European Union (Member States: Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Italy, Luxembourg, The Netherlands, Portugal, Spain, Sweden, United Kingdom)
FAO	Food and Agricultural Organization
HCl	Hydrochloric acid, hydrogen chloride
I-TEQ	International Toxic Equivalents
IARC	International Agency for Research on Cancer
IFCS	International Forum on Chemical Safety
IOMC	Inter-Organization Programme for the Sound Management of Chemicals
IPCS	International Programme on Chemical Safety
Koc	Partition coefficient: octanol/carbon
Kow	Partition coefficient: octanol/water
M(S)WI	Municipal (solid) waste incineration
MW	Megawatt
N-TEQ	Nordic Toxic Equivalents (scheme adopted by the Scandinavian countries)
NATO/CCMS	North Atlantic Treaty Organization/Challenges of Changes in Modern Society
ND	Not determined
ONT	Ontario
PCB	Polychlorinated biphenyls
PCDD	Polychlorinated dibenzo-*p*-dioxins
PCDF	Polychlorinated dibenzofurans
PCN	Polychlorinated naphthalenes
PCP	Pentachlorophenol
PCT	Polychlorinated terphenyls
POPs	Persistent Organic Pollutants
PVC	Polyvinyl chloride
t	Ton (metric)
TCDD	Tetrachlorodibenzo-*p*-dioxin
TDI	Tolerable Daily Intake
TEF	Toxicity Equivalency Factor
TEQ	Toxicity Equivalent
TJ	Terajoule (10^{12} Joule)
UNEP	United Nations Environment Programme
VC(M)	Vinyl chloride (monomer)
WHO	World Health Organization

Units

mg	Milligram	10^{-3} g
µg	Microgram	10^{-6} g
ng	Nanogram	10^{-9} g
pg	Picogram	10^{-12} g
fg	Femtogram	10^{-15} g

DIOXIN AND FURAN INVENTORIES

NATIONAL AND REGIONAL EMISSIONS OF PCDD/PCDF

0 SUMMARY

In its decision 19/13 C of 7 February 1997, the Governing Council of UNEP [*] requested the Executive Director of UNEP to convene an Intergovernmental Negotiating Committee with a mandate to prepare an international legally binding instrument for implementing international action on twelve specified POPs, and to establish an expert group for the development of science-based criteria and a procedure for identifying additional POPs as candidates for future international action. With this mandate to facilitate a convention on reduction and elimination of releases of POPs, UNEP Chemicals initiated several activities to help the countries to prepare for the negotiations of the convention. As one of these immediate actions UNEP Chemicals will "…assist countries in the identification of national sources of dioxin/furan releases by promoting access to the information on available sources of dioxins/furans…".

0.1 General

Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF) have never been produced intentionally but are unwanted byproducts of many chemical industrial processes and of all combustion processes. Almost all possible 210 congeners are released from these sources and, due to chemical, physical, and biological stability and long-range transport, are ubiquitous and have been detected in all environmental compartments. Due to the persistence of the 2,3,7,8-substituted congeners and the lipophilicity of these compounds, PCDD/PCDF accumulate in fatty tissues and in carbon-rich matrices such as soils and sediments.

PCDD/PCDF exhibit biological effects commonly associated with chlorinated organic chemicals. Dioxin exposures are associated with an increased risk of severe skin lesions, altered liver function and lipid metabolism, general weakness associated with drastic weight loss, changes in activity of various liver enzymes, depression of the immune system, and endocrine and nervous system abnormalities. 2,3,7,8-Cl_4DD is a potent teratogenic and fetotoxic chemical in animals and a potent promoter in rat liver carcinogenesis; it also causes cancers of the liver and other organs in animals

In 1997, the International Agency for Research on Cancer (IARC) classified the most toxic congener of the PCDD/PCDF as a Group 1 compound as carcinogenic to humans. All other 2,3,7,8-substituted PCDD/PCDF were classified as non-carcinogenic to humans (Group 3).

In May 1998, the World Health Organization (WHO) evaluated the risks which dioxins may cause to health. In Bilthoven 1990, WHO experts established a tolerable daily intake of 10 pg TCDD/kg bw. Since then, new epidemiological data have emerged, especially on neurotoxicological development and the endocrine system. Finally, WHO agreed on a new tolerable daily intake (TDI) of 1 to 4 picogram per kilogram body weight and day. The experts however recognized that subtle

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[*] United Nations Environment Programme

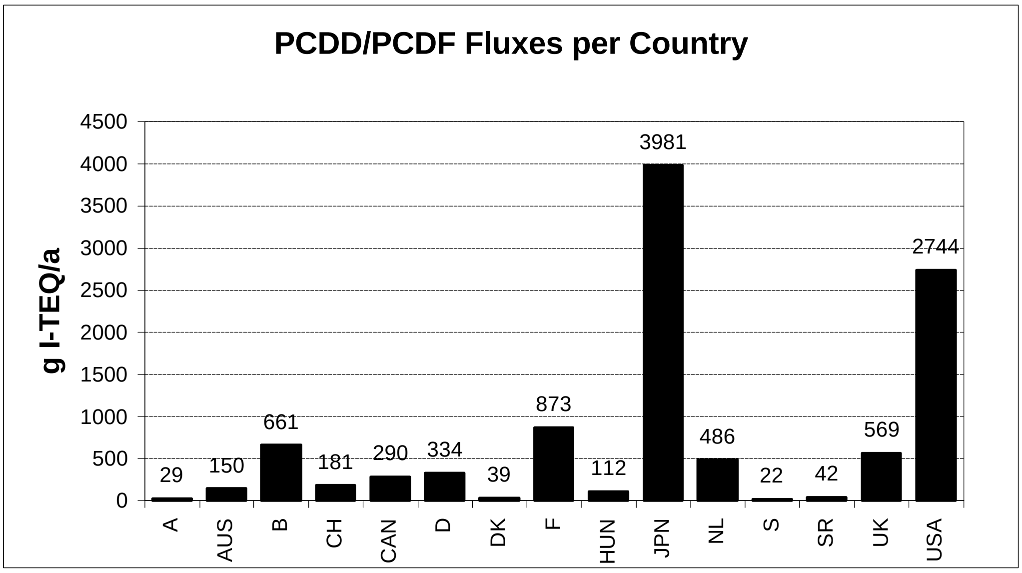
effects occur in the general population in developed countries at background levels of 2 to 6 picograms per kilogram body weight and day. They therefore recommended that: every effort should be made to reduce exposure to the lowest possible level and the range established for 2,3,7,8-Cl_4DD should be applied to a TEQ including the 17 2,3,7,8-substituted PCDD and PCDF as well as the coplanar and *mono-ortho* substituted PCB (WHO 1998).

For the risk assessment of complex mixtures of PCDD/PCDF, Toxicity Equivalency Factors (TEFs) were established. They were developed to rank the toxicity of the 17 individual 2,3,7,8-substituted congeners in relation to the toxicity of 2,3,7,8-Cl_4DD based on the evidence that there is a common, receptor-mediated mechanism of action for these compounds. Although there exist limitations of the approach due to a number of simplifications, TEFs allows conversion of quantitative analytical data for individual PCDD/PCDF congeners into a single Toxic Equivalent (TEQ), and thus order of magnitude estimates of toxicity.

0.2 Dioxin and Furan Inventories

In this report, initial findings obtained from national inventories of releases of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF) are presented and evaluated. Most countries have estimated PCDD/PCDF emissions to air and only a few findings are available for emissions to water and soil.

Today’s major emissions of PCDD/PCDF into the environment come from combustion processes. Based on the presently available data and a reference year around 1995, the central estimate of total annual PCDD/PCDF emissions is approximately 10,500 g I-TEQ. The lower estimate is around 8,300 g I-TEQ/a and the upper estimate approximately 36,000 g I-TEQ/a. The high PCDD/PCDF emissions reported by a few countries for the early 1990s mainly drive these numbers. It should be noted that Japan has updated its dioxin inventory; for the year 1998, a total emission of 5,300 g I-TEQ was estimated. This new number will add another 1,300 g I-TEQ to the “global” inventory. The estimate for a year around 1995 is shown in Figure 1. For France, the estimate for 1998 is included.



A	Austria
AUS	Australia
B	Belgium
CH	Switzerland
CAN	Canada
D	Germany
DK	Denmark
F	France
HUN	Hungary
JPN	Japan
NL	The Netherlands
S	Sweden
SR	Slovak Republic
UK	United Kingdom
USA	United States

Figure 1: PCDD/PCDF annual fluxes into air (g I-TEQ/a); reference year 1995.
Total emissions from known sources = 10,500 g I-TEQ/a

The numbers in Figure 1 can be compared with an inventory established for the Member States of the European Union and the TNO inventory which both cover broader geographical areas.

The EU report covers 17 countries and estimates approximately 5,800 g I-TEQ/a to be emitted from known sources. However, the authors assume a large margin of uncertainty and cannot exclude an upper estimate of approximately 20,000 g I-TEQ being released into the air from the 17 European countries.

The TNO study is less specific to PCDD/PCDF sources and estimates an annual release of 1,300 g I-TEQ for the reference year 1990. Finally, based on relatively few assumptions and no detailed evaluation of emission inventories, Brzuzy and Hites estimate a global PCDD/PCDF release of 50,000 g I-TEQ/a.

To compare the most important emission sources from national inventories, the PCDD/PCDF sources were categorized into nine major sectors which are listed below:

Iron and Steel:	Iron and steel plants including foundries, sinter and coke plants (as in many countries, coke is exclusively produced for use in steel plants)
Non-ferrous Metals:	Primary and secondary plants for the generation of copper, aluminum, zinc, lead
Power Plants:	Fuelled with coal, gas, crude oil, wood
Industrial Combustion Plants:	Industrial units fuelled with coal, gas, crude oil, sewage sludge, and biomass for use on-site
Small Combustion Units:	Mostly domestic stoves and chimneys fired with coal, oil, and gas
Waste Incineration:	Includes incineration of municipal solid waste, hazardous waste, sewage sludge, hospital waste, waste wood, and crematoria
Road Transport:	Passenger cars, busses, trucks run on leaded or unleaded gasoline or Diesel
Mineral Products Production:	Generation of cement, lime, glass, brick, *etc.*
Others:	Shredder plants, asphalt mixing, drying of green fodder, wood chips, chemical industry, accidental fires and prescribed burnings

The percentage share of the above sectors within a country is illustrated in Figure 2. It can be seen that the sector of waste incineration as defined above is still the major emitter in many countries. Exceptions are Austria, Australia, and Germany for the reference year 1995. For the “global” inventory based on these fifteen countries the share of the waste incineration is almost 50 % (see right bar in Figure 2).

In absolute numbers, the main part of PCDD/PCDF emissions from the sector of waste incineration is due to the municipal waste incinerators located in Japan. This source sector alone is responsible for almost 40 % of the total PCDD/PCDF inventory to air from identified sources so far. On the other hand, in countries with stringent regulation of PCDD/PCDF emissions from municipal and hazardous waste incineration, *e.g.* the member states of the European Union that started legislation in the early 1990, incineration of waste is a minor contributor to any national inventory.

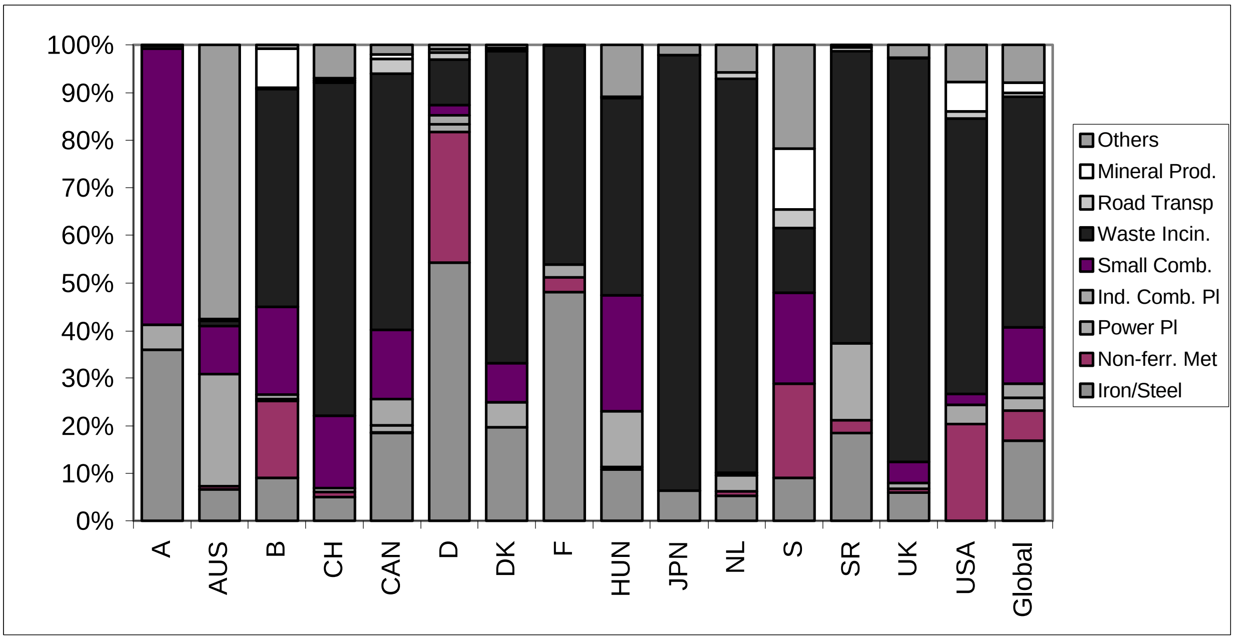


Figure 2: Percentage contribution per sector and country to the overall PCDD/PCDF air emission inventory; reference year 1995

The present situation of PCDD/PCDF inventories in terms of geographic coverage and methodology can be summarized as follows:

- The present number of national PCDD/PCDF emission inventories is very small (15 based on national data).
- Almost exclusively, the existing inventories only address PCD/PCDF emissions to air. There may be considerable amounts of dioxins and furans being contained in reservoirs containing “old” chemicals such as 2,4,5-trichlorophenoxy acetic acid (2,4,5-T), pentachlorophenol used for wood treatment, polychlorinated biphenyls (PCB) used in transformers and capacitors. Other reservoirs include landfills/dumps, contaminated soils, and sediments.
- Most data is available for countries from Western Europe and Northern America. However, the inventory for the United States is still in a draft stage. From Asia, there is only one inventory for Japan, which covers a few source categories, and the estimate for emissions from waste incinerators from South Korea. From the Southern hemisphere, so far, only Australia has estimated annual emissions based on emission factors from the literature. From Africa, Central and Southern America, there are no data at all.
- Some countries have based their inventories on emission factors generated outside the own country. For some sources, there are no emission factors determined, *e.g.* open garbage burning, landfill fires, *etc.*

From the existing inventories the following conclusions can be drawn:

- Amongst the source sectors, the best coverage exists for municipal solid waste incineration for both stack emission measurements and activity rates. As this sector undergoes the most dramatic changes in technology, emission factors and PCDD/PCDF emissions change rapidly. As a consequence, strong downward trends are recognized in countries with modern technology or rigid legislation.

- The sector of hazardous waste incineration is relatively homogeneous and does not present a major source in any country.

- There is only limited information available from the iron and steel-producing sector. Some European countries have identified this sector as the major contributor to national dioxin inventories. The United States and Canada are aware of these sources, but so far, not a single measurement was performed. Here, generation of reliable data is urgently required.

- From the few examples on dioxin and furan emissions to water and land or with products it can be concluded that emissions to water only cover wastewaters from the pulp and paper industry. Contamination in products largely is limited to pentachlorophenol (PCP) and the PCDF in polychlorinated biphenyls (PCB) are being ignored.

- For some countries, the inventory should be updated to improve estimates of the present situation, especially when more stringent regulation has been established.

- Presently there exist no harmonized methods on generating and evaluating data for national PCDD/PCDF inventories. In addition, the coverage of sources varies from country to country. Some countries such as the United Kingdom and the United States include releases of PCDD/PCDF to landfills and land to a certain extent. Some countries give ranges of lower and upper estimates whereas other countries use mean/median values to calculate the annual dioxin emissions for a given source. Harmonization of data acquisition and evaluation is an obvious need and will help in comparing national dioxin and furan inventories.

- Finally, harmonization of protocols for sampling stack emissions, water, soil, *etc.* and for analyzing these samples is highly recommended.

The present report should only be seen as a snapshot on PCDD/PCDF emissions and estimates of total releases of these compounds into the environment. The major sectors of PCDD/PCDF releases into the air have been identified. Nevertheless, PCDD/PCDF sources may exist which have not yet been identified or quantified, especially in geographic areas with no data. Nevertheless it has to be taken in mind that emission factors established in industrialized countries cannot be assumed to be representative for industries in less developed countries.

Presently, the coverage is not sufficient to estimate global emissions of PCDD/PCDF. There are several efforts underway to identify dioxin sources in parts of the world where so far, there is no information available. In addition, existing inventories will be updated, as it is obvious that measures were taken by many countries to reduce emissions of PCDD/PCDF into the environment. For some industrialized countries in Europe and North America, strong downward trends have been observed during the last years. Implementation of dioxin abatement technologies in industrial sectors and advanced combustion technology will help to reinforce such trends.

1 PURPOSE OF THE STUDY

The Governing Council of the United Nations Environment Programme (UNEP), in its decision 18/32 of May 25, 1995, invited international organizations (IOMC, IPCS, IFCS) to develop recommendations and information on international action for twelve persistent organic pollutants (POPs). In June 1996, sufficient evidence was achieved for IFCS (Intergovernmental Forum on Chemical Safety) to recommend a global action plan on POPs. In its decision 19/13 C of 7 February 1997, the Governing Council of UNEP requested the Executive Director of UNEP to convene an Intergovernmental Negotiating Committee (INC) with a mandate

1) To prepare an international legally binding instrument for implementing international action initially beginning with the twelve specified POPs and

2) To establish an expert group for the development of science-based criteria and a procedure for identifying additional POPs as candidates for future international action.

With this mandate to develop a convention on reduction and elimination of releases of POPs, UNEP Chemicals, initiated under the direction of UNEP's Executive Director, Dr. Klaus Töpfer, several activities to help the countries to prepare for the negotiations of the convention. Among the immediate actions, UNEP Chemicals will

"…assist countries in the identification of national sources of dioxin/furan releases by promoting access to the information on available sources of dioxins/furans…".

This report summarizes findings in dioxin research and technology and is a first attempt to compile national emission inventories for polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF). The information compiled here will help countries to establish their own dioxin inventories as asked in the decisions adopted by the Governing Council (GC) at its 19th Session (19/13C).

For this study, most information is taken directly from the original reports of a given country or from summarizing reports without harmonization or classification of *e.g.* source categories or industrial processes. This makes the present findings somewhat heterogeneous in terms of source categories. Further complication to compare dioxin inventories is due to the fact that the estimates are given for different reference years and thus, data cannot be compared directly. In addition, countries took different approaches to generate their estimates. This report will be updated, as more information will become available. At a later stage, emission factors can be included as well as comparative evaluations.

2 CONTENT OF THIS REPORT

In order to establish a dioxin inventory, some general information on this class of compounds is needed to give a rationale for such inventory. Thus, the report starts with “Background Information on Toxicity of PCDD/PCDF” in Section 3 to give a brief summary on the present state of knowledge in toxicology and the classification of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in terms of carcinogenicity. It also addresses the recommendation of WHO for a tolerable daily intake (TDI) for humans which is the starting point for actions taken to reduce exposure of humans towards dioxins and furans.

Findings on sources and emissions of dioxins and furans are given in Chapter 4 “Background Information on Sources of PCDD/PCDF” summarized from the literature and experiences mostly from industrialized countries. Chapter 5 briefly addresses technical abatement technologies that where successfully applied in order to reduce emissions of PCDD/PCDF into the environment. Most of the technologies referenced here attempt to minimize emissions into air. The methodology how the national inventories were generated is given in Chapter 6.

In the early 1990s, many industrialized countries initiated national emission inventories. As a consequence, countries commissioned national research institutes or agencies to identify and quantify PCDD/PCDF emissions from known sources. For some countries, updates of the original inventories were performed. National PCDD/PCDF emission inventories were established for most European countries: these are summarized in Chapter 8 “European Dioxin Inventories”. A detailed regional dioxin mass balance is available for the city of Hamburg, Germany, which published a report where almost all sources of PCDD/PCDF could be quantified; the results are given in section 8.6.2. So far, only the European Commission undertook efforts to quantify dioxin emissions for a multitude of countries using the same methodological approach when it launched a project to establish a dioxin emission inventory. The results are shown in section 8.12.2. As part of the UN ECE LRTAP protocol, TNO published a report on emissions of heavy metals and organic pollutants for the reference year 1990 (UBA 1997). The dioxin-related results are summarized in Chapter 8.14.

Outside Europe, there are only two national emission inventories available from the Americas, namely for Canada and the United States of America. Both inventories are still in a draft stage and not finalized. The results are summarized in Chapter 9 “Dioxin Inventories from the American Continents“. The inventories for Australia and Japan are presented in Chapter 10 “Dioxin Inventories in Asia and the Pacific”.

In most cases, information was compiled on a nation-wide basis. However, for some areas, only one source category has been addressed so far, *e.g.* for Korea where only the sector of municipal solid waste incineration was investigated (see Section 10.4).

Attempts for a global dioxin emission inventory were undertaken by Ron Hites and coworkers who based their emission estimates on deposition measurements from samples collected all over the globe (see Chapter 11).

The responses on questionnaires sent by UNEP Chemicals to the POPs focal points in 1997 are summarized in Appendix B. For three countries new quantitative information was given which was not found in the published literature and thus not contained in the main part of the report (Chapters 1-13). On the other hand, there exist estimates on emissions of PCDD/PCDF from identified sources in the publications that were not addressed in the questionnaires.

3 BACKGROUND INFORMATION ON TOXICITY OF PCDD/PCDF

Polychlorinated dibenzo-*p*-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) are environmental contaminants detectable in almost all compartments of the global ecosystem in trace amounts. These compound classes in particular have caused major environmental concern. In contrast to other chemicals of environmental concern such as polychlorinated biphenyls (PCB), polychlorinated naphthalenes (PCN), and polychlorinated pesticides like DDT, pentachlorophenol (PCP) or others, PCDD/PCDF never were produced intentionally. They are formed as by-products of numerous industrial activities and all combustion processes (Fiedler *et al.* 1990).

The term “dioxins“ [†] refers to 75 congeners of polychlorinated dibenzo-*p*-dioxins (PCDD) and 135 congeners of polychlorinated dibenzofurans (PCDF). Amongst these 210 compounds, 17 congeners can have chlorine atoms at least in the positions 2, 3, 7, an 8 of the parent molecule. These 17 2,3,7,8-substituted congeners are toxic to many laboratory animals, resistant towards chemical, biological, and physical attack, and thus many accumulate in the environment and in organisms, such as animals and humans. The 2,3,7,8-TCDD (2,3,7,8-Cl_4DD) also named “Seveso-dioxin“ is considered to be the most toxic man-made compound. Besides the anthropic sources, an enzyme-mediated formation of PCDD and PCDF from 2,4,5- and 3,4,5-trichlorophenol has been demonstrated *in vitro* (Öberg *et al.* 1990, Wagner *et al.* 1990).

First risk assessments only focused on the most toxic congener, the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-Cl_4DD = 2,3,7,8-TCDD). Soon it was recognized, though, that all PCDD/PCDF substituted at least in position 2, 3, 7, or 8 are highly toxic and thus, major contributors to the overall toxicity of the dioxin mixture. In addition, despite the complex composition of many PCDD/PCDF containing “sources“, only congeners with substitutions in the lateral positions of the aromatic ring, namely the carbon atoms 2, 3, 7, and 8, persist in the environment and accumulate in food-chains.

Almost all 210 individual congeners have been identified in emissions from thermal and industrial processes and consequently PCDD/PCDF are found as mixtures of individual congeners in environmental matrices such as soil, sediment, air, and plants and lower animals. PCDD/PCDF, particularly the higher chlorinated, are poorly soluble in water, have a low volatility, and adsorb strongly to particles and surfaces (high K_{OC}). Thus, PCDD/PCDF can hardly be identified in water and are immobile in soils. Especially, the 2,3,7,8-chlorine substituted PCDD/PCDF are extremely stable in the environment and bioaccumulate in fatty tissues (high K_{OW}) of animals and humans.

3.1 Toxicity of PCDD/PCDF and Risk Assessment

PCDD and PCDF produce a spectrum of toxic effects in animals; however, most information is available on 2,3,7,8-Cl_4DD (TCDD) only. Most toxicity data on TCDD result from high dose oral exposures to animals. There is a wide range of difference in sensitivity to PCDD lethality in animals. The signs and symptoms of poisoning with chemicals contaminated with TCDD in humans are similar to those observed in animals. Dioxin exposures to humans are associated with an increased risk of severe skin lesions (chloracne and hyperpigmentation), altered liver function and lipid metabolism, general weakness associated with drastic weight loss, changes in activity of

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[†] In this report, where the term “dioxin” or “dioxins and furans” is used alone, it should be interpreted as including all polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans, unless specifically stated otherwise.

various liver enzymes, depression of the immune system, and endocrine and nervous system abnormalities. 2,3,7,8-TCDD is a potent teratogenic and fetotoxic chemical in animals and a potent promoter in rat liver carcinogenesis. TCDD also causes cancers of the liver and other organs in animals (see below).

3.1.1 IARC Evaluation of Human and Animal Carcinogenicity Data

The most important epidemiological studies for the evaluation of 2,3,7,8-Tetrachlorodibenzo-*p*-dioxin (2,3,7,8-Cl_4DD or 2,3,7,8-TCDD) are four cohort studies of herbicide producers (one each in the United States and the Netherlands, two in Germany). These studies involve the highest exposures to 2,3,7,8- Cl_4DD. The cohort of residents in a contaminated area from Seveso, Italy is well known, but the exposures at Seveso were lower and the follow-up shorter than those in the industrial settings. Most of the four industrial cohorts include analyses of sub-cohorts considered to have the highest exposure and/or longest latency. Overall, the strongest evidence for the carcinogenicity of 2,3,7,8-Cl_4DD is for all cancers combined, rather than for any specific site. On the basis of these studies, the International Agency for Research on Cancer (IARC) concluded that there is limited evidence in humans for the carcinogenicity of 2,3,7,8-Cl_4DD. There was inadequate evidence in humans for the carcinogenicity of PCDD other than 2,3,7,8- Cl_4DD.

For PCDF, two incidents, each involving about 2,000 cases occurred in which people were exposed to sufficient PCB and PCDF to produce symptoms (Yucheng and Yusho accidents). Fatal liver disease is 2-3 times more frequent than national rates in both cohorts. In the Yusho cohort from Japan, at 22 years of follow-up, there is a three-fold excess of liver cancer mortality in men, which was already detectable and even higher at 15 years of follow-up. In the Yucheng cohort, Taiwan, after 12 years of follow-up, there is no excess of liver cancer mortality. Based upon these data, IARC concluded that there is inadequate evidence in humans for the carcinogenicity of PCDF.

PCDD: In a number of experiments with rats and mice in which 2,3,7,8-Cl_4DD was administered, increases in the incidence of liver tumors were consistently found in both males and females. In addition, tumors were increased at several other sites in rats, mice and Syrian hamsters, but these effects were dependent upon the species, sex and route of administration of 2,3,7,8-Cl_4DD. Although the doses resulting in increased tumor incidence in rodents are extremely low, they are very close to doses that are toxic in the same species. These data led to the conclusion that there is sufficient evidence in experimental animals for the carcinogenicity of 2,3,7,8- Cl_4DD. Evaluation of much smaller databases led to the conclusion that there is limited evidence in experimental animals for the carcinogenicity of a mixture of 1,2,3,6,7,8- and 1,2,3,7,8,9-Cl_6DD and that there was inadequate evidence for the carcinogenicity in experimental animals of 2,7-Cl_2DD, 1,2,3,7,8-Cl_5DD and 1,2,3,4,6,7,8,-Cl_6DD.

PCDF: There are no long-term carcinogenicity studies on PCDF, but some tumor promotion studies were evaluated in which rats and mice were exposed to some of the congeners following short duration exposure to known carcinogens. It was concluded that there is inadequate evidence in experimental animals for the carcinogenicity of 2,3,7,8-Cl_4DF, but there is limited evidence in experimental animals for the carcinogenicity of 2,3,4,7,8-Cl_5DF and 1,2,3,4,7,8-Cl_6DD.

The toxicity of 2,3,7,8-Cl_4DD segregates with the cytosolic aryl (aromatic) hydrocarbon receptor (AhR), and the relative toxicities of other PCDD and PCDF congeners is associated with their ability to bind to the receptor, which occurs in all rodent and human tissues. The AhR binding affinities of 2,3,7,8-Cl_4DF, 1,2,3,7,8- and 2,3,4,7,8-Cl_5DF are in the same order of magnitude as that observed for 2,3,7,8-Cl_4DD. PCDD with at least three lateral chlorine atoms bind with some affinity to the AhR. Current evidence is that most, if not all, biological effects of 2,3,7,8-Cl_4DD

and other PCDD arise from an initial high affinity interaction with the AhR and it appears that the biochemical and toxicological consequences of PCDF exposure are the result of a similar mode of action. The limited carcinogenicity data available for congeners other than 2,3,7,8-Cl_4DD indicate that carcinogenic potency is also proportional to AhR affinity. Based on this evidence, all PCDD and PCDF are concluded to act through a similar mechanism and require an initial binding to the AhR. Binding of 2,3,7,8-Cl_4DD to the AhR results in transcriptional activation of a battery of 2,3,7,8-Cl_4DD-responsive genes, but currently no responsive gene has been proven to have a definitive role in its mechanism of carcinogenesis.

Overall evaluation

Taking all of the evidence into consideration, the following evaluations were made by IARC (IARC 1997):

- 2,3,7,8-Tetrachlorodibenzo-*p*-dioxin (2,3,7,8-Cl_4DD) is carcinogenic to humans (Group 1).
- Other polychlorinated dibenzo-*p*-dioxins are not classifiable as to their carcinogenicity to humans (Group 3).
- Dibenzo-*p*-dioxin is not classifiable as to its carcinogenicity to humans (Group 3).
- Polychlorinated dibenzofurans are not classifiable as to their carcinogenicity to humans (Group 3).

3.1.2 Toxicity Equivalency Factors (TEF)

Many regulatory agencies have developed so-called Toxicity Equivalency Factors (TEF) for risk assessment of complex mixtures of PCDD/PCDF (Kutz *et al.* 1990). The TEF are based on acute toxicity values from *in vivo* and *in vitro* studies. This approach is based on the evidence that there is a common, receptor-mediated mechanism of action for these compounds. However, the TEF approach has its limitations due to a number of simplifications. Although the scientific basis cannot be considered as solid, the TEF approach has been developed as an administrative tool and allows to convert quantitative analytical data for individual PCDD/PCDF congeners into a single Toxic Equivalent (TEQ). TEF particularly aid in expressing cumulative toxicities of complex PCDD/PCDF mixtures as one single TEQ value. Today, almost all literature data are reported in I-TEQ.

It should be noted that TEF are interim values and administrative tools. They are based on present state of knowledge and should be revised as new data gets available. Today's most commonly applied TEFs were established by a NATO/CCMS Working Group on Dioxins and Related Compounds as International Toxicity Equivalency Factors (I-TEF) (NATO/CCMS 1988a). However, in 1997, a WHO/IPCS working group re-evaluated the I-TEFs and established a new scheme. The two schemes are found in Appendix A. The re-evaluation concluded to include *non-ortho* and *mono-ortho*-substituted polychlorinated biphenyls (PCB) into the TEF scheme for dioxin-like toxicity as well.

3.2 Exposure Data

Human exposure to background contamination with PCDD/PCDF is possible *via* several routes:

- Inhalation of air and intake of particles from air
- Ingestion of contaminated soil
- Dermal absorption
- Food consumption

In 1990, a WHO working group concluded that 90 % of the daily dioxin intake (from background contamination) results from ingestion. Especially, foodstuffs of animal origin are responsible for the daily intake of approximately 2 pg TEQ/(kg bw·d). All other foodstuffs, especially the „non-fatty“ ones, are of minor importance in terms of PCDD/PCDF intake. They are either of plant origin or do not have a high potential for bioaccumulation of lipophilic compounds. Due to many measures to reduce emissions of PCDD/PCDF into the environment, reduction of PCDD/PCDF contamination in food was observed. As a consequence, the daily intake *via* food decreased. Whereas in Germany in 1991, the average daily intake was 127.3 pg TEQ/d, the present daily intake for an average German adult is estimated to 69.6 pg TEQ/d. The strongest decline was observed for fish. In 1991, fish contributed for ca. 30 % of the daily intake (same percentage as for dairy and meat products): today only 10 % of the daily intake is due to fish.

Although no adverse health effects could be causally linked so far with background exposures of PCDD/PCDF in human milk, for reasons of preventive health care, the relatively high exposure of breast-fed infants must still be considered a matter of concern. Analyses of more than 1,000 individual human milk samples from nursing mothers in North-Rhine Westphalia (Germany) revealed that the mean PCDD/PCDF concentration decreased from 34 pg I-TEQ/g milkfat in 1989 to 14.2 pg I-TEQ/g milkfat in 1996. Despite this decline of 60 %, the PCDD/PCDF daily intake for babies is 68 pg I-TEQ/kg bw·d), which is almost 70-fold above the TDI of 1 pg TEQ/(kg bw·d) for an adult.

In May 1998, WHO evaluated the risks which dioxins may cause to health. During a previous meeting in Bilthoven 1990, WHO experts established a tolerable daily intake of 10 pg TCDD/kg bw. Since then, new epidemiological data has emerged, especially on neurotoxicological development and the endocrine system. Finally, WHO agreed on a new tolerable daily intake (TDI) of 1 to 4 picogram per kilogram body weight and day. The experts however, recognized that subtle effects occur in the general population in developed countries at background levels of 2 to 6 picograms per kilogram body weight and day. They therefore recommended that every effort should be made to reduce exposure to the lowest possible level. In addition, the range established for 2,3,7,8-Cl_4DD should be applied to a TEQ including the seventeen 2,3,7,8-substituted PCDD and PCDF as well as the coplanar and *mono-ortho* substituted PCB (WHO 1998, van Leeuwen and Younes 1998).

4 BACKGROUND INFORMATION ON SOURCES OF PCDD/PCDF

Since the first overview on formation and sources of PCDD/PCDF was published in 1980 (Esposito et al. 1980), several updates are available in the international literature. The findings can be summarized as follows (Hutzinger and Fiedler 1993):

- PCDD/PCDF were never produced intentionally but occur as trace contaminants in a variety of industrial and thermal processes.

- Due to their chemical, physical and biological stability PCDD/PCDF are able to remain in the environment for long times. As a consequence dioxins from so-called "primary sources" (once formed in industrial or combustion processes) can be transferred to other matrices and enter the environment. Such "secondary" sources are sewage sludge/biosludge, compost, or contaminated areas (*e.g.* Kieselrot in Germany).

- Enzymatic reactions can dimerize chlorophenols to PCDD/PCDF. However, compared to chemical-industrial and combustion sources, biological formation seems to be negligible.

4.1 Primary Sources of Dioxins

4.1.1 Industrial-Chemical Processes

Primary sources of environmental contamination with PCDD/PCDF in the past were due to production and use of chloroorganic chemicals, including the pulp and paper industry. In wet-chemical processes the propensity to generate PCDD/PCDF during synthesis of chemical compounds decreases in the following order:

Chlorophenols > Chlorobenzenes > Aliphatic chlorinated compounds > Inorganic chlorinated compounds

Factors favorable for the formation of PCDD/PCDF are high temperatures, alkaline media, presence of *UV*-light, and presence of radicals in the reaction mixture/chemical process (Hutzinger and Fiedler 1991, 1993). An overview on dioxin concentrations in chemicals is given in Table 1. As can be seen the concentrations can vary by several orders of magnitude.

Table 1: PCDD/PCDF concentrations in chemical products

Substance	Concentration µg I-TEQ/kg
PCP	up to 2,320
PCP-Na	up to 450
PCB - Clophen A 30	11
PCB - Clophen A 60	2,179
2,4,6-Trichlorophenol	680
Trichlorobenzene	0.023
p-Chloranil (old)	376
o-Chloranil (old)	63
Hostaperm Violet RL	1.2
Violet 23	19
Blue 106	56

Changes in the industrial processes have resulted in reduction of PCDD/PCDF concentrations in the products: *e.g*. an estimate for Germany says that until 1990 about 105 g I-TEQ have been introduced through use of the dye pigment Violet 23 (chloranil produced by old process as intermediate). Application of a new process *via* hydroquinone will reduce the annual input to about 3 g I-TEQ (BGA/UBA 1993).

Emissions of PCDD/PCDF into the environment *via* water and to soils occur from kraft pulp and paper mills. The US-EPA inventory estimates annual emissions from kraft pulp and paper mills in the range of 20 g I-TEQ. In addition, PCDD/PCDF were detected in the final product (pulp, paper) as well as in the pulp and paper sludges. With advanced bleaching technology, the PCDD/PCDF contamination in effluents, products, and sludges was reduced.

In Germany there exist exclusively sulfite mills, which presently do not use molecular chlorine. Dioxin levels detected in German pulp were below 0.1 ng TEQ/kg d.m. (BGA/UBA 1993). The analysis of imported sulfate (Kraft) pulps gave concentrations in the range between 0.2 and 1.3 ng TEQ/kg d.m. Presently the import of Kraft pulps to Germany stands at three million tons, so that the total import of dioxins *via* Kraft pulp is between 0.6 and 3.9 g I-TEQ. Dioxin levels in paper products from fresh fibers generally are less than 1 ng TEQ/kg d.m. In recycling paper, however, average dioxin concentrations are between 5 to 10 ng TEQ/kg.

As a consequence, products containing any of the above mentioned chemicals are contaminated with PCDD/PCF as well. Amongst these, wood treated with pentachlorophenol (PCP) or other chlorinated preservatives as well as PCB-based electric fluids are amongst those with the highest contaminations. Other PCP-treated materials include textiles, leather goods, and cork products.

4.1.2 Thermal Processes

Whereas in the past, the chemical industry and to a lesser extent the pulp and paper industry were considered to be the main source of dioxins and also the cause of today's contaminated sites in many industrialized countries, today’s dioxin input is mainly due to thermal processes. There is still a considerable focus on waste incineration but based on the requirements for dioxin reduction in stack gases set by several national authorities, the importance of this source category has declined during the last years. Examples can be seen especially in the European emission inventories. For example, in Germany the annual input from municipal solid waste incineration (MSWI) *via* exhaust gases of about 400 g TEQ per year in 1988/89 was reduced to less than 4 g TEQ since 1997.

Table 2. PCDD/PCDF trends in emissions from municipal solid waste incineration (MSWI)

	Concentration (ng I-TEQ/m³)	Flux (mg I-TEQ/h)
MSWI of the 1970s	50	5
MSWI around 1990	5	0.5
Modern MSWI	0.1	0.01

The process by which PCDD/PCDF are formed during incineration are not completely understood nor agreed upon. Three possibilities have been proposed to explain the presence of dioxins and furans in incinerator emissions (NATO/CCMS 1988b):

- PCDD/PCDF are already present in the incoming waste - in Germany representative measurements gave about 50 ng I-TEQ/kg waste - and are incompletely destroyed or transformed during combustion. Not relevant for modern MSWIs.

- PCDD/PCDF furans are produced from related chlorinated precursors (= pre-dioxins) such as PCB, chlorinated phenols and chlorinated benzenes.

- PCDD/PCDF are formed *via de novo* synthesis. This is, they are formed from the pyrolysis of chemically unrelated compounds such as polyvinyl chloride (PVC) or other chlorocarbons, and/or the burning of non-chlorinated organic matter such as polystyrene, cellulose, lignin, coal, and particulate carbon in the presence of chlorine-donors.

From the knowledge gained from MSWIs it can be concluded that PCDD/PCDF can be formed in other thermal processes in which chlorine-containing substances are burnt together with carbon and a suitable catalyst (preferably copper) at temperatures above 300 °C in the presence of excess air or oxygen. Preferentially dioxin formation takes place in the zone when combustion gases cool down from about 450 °C to 250 °C (*de novo* synthesis) and not in the combustion chamber. Possible sources of the chlorine input are PVC residues as well as chloroparaffins in waste oils and inorganic chlorine.

An overview on combustion sources known to generate and to emit PCDD/PCDF is shown in Table 3. It should be mentioned that PCDD/PCDF are not only found in the stack gases but also in the solid residues from any combustion process, *e.g.* bottom ashes, slags, and fly ash. Typical concentrations for solid residues from waste incineration are given in Table 4. With advanced technology and better burn-out of the ashes and slags (characterized by a low content on organic carbon), the PCDD/PCDF concentrations declined (Fiedler 1996b).

Table 3: PCDD/PCDF combustion sources

Stationary sources	
Waste incineration:	Municipal solid waste, clinical, hazardous waste, sewage sludge
Steel industry:	Steel mills, sintering plants, hot-strip mills
Recycling plants:	Non-ferrous metals (melting, foundry; Al, Cu, Pb, Zn, Sn),
Energy production:	Fossil fuel power plants, wood combustion, landfill gas
Diffuse sources	
Traffic:	Automobiles
Home heating:	Coal, oil, gas, wood
Accidents:	PCB fires, fires in buildings, forest fires, volcanic eruptions)

Table 4: PCDD/PCDF in residues from municipal solid waste incineration

Matrix	Concentration ng I-TEQ/kg	Remarks
Municipal solid waste incineration		
Fly ash	13,000	Mean, Germany late 1980s
	<1,000	New technology, Germany
Bottom ash	50	Mean Germany, late 1980s
	5-20	New technology, Germany
Wood combustion		
Fly ash	5,800	Mean waste wood, Switzerland
	2.5	Mean natural wood, Switzerland
Bottom ash	820	Mean waste wood, Switzerland
	5.3	Mean natural wood, Switzerland
Home heating systems		
Soot	4-42,048	Range, wood and coal, Germany

4.2 Secondary Sources of PCDD/PCDF = Reservoirs

Dioxin reservoirs are those matrices where PCDD/PCDF are already present, either in the environment or as products. The PCDD/PCDF found in these reservoirs are not newly generated but concentrated from other sources. A characteristic of the reservoir sources is that they have the potential to allow re-entrainment of PCDD/PCDF into the environment. Product reservoirs include PCP-treated wood, PCB-containing transformers and sewage sludge, compost, and liquid manure, which can be used as fertilizers in agriculture and gardens. Reservoirs in the environment are for example landfills and waste dumps, contaminated soils (mainly from former chemical production or handling sites), and contaminated sediments (especially in harbors and rivers with industries discharging directly to the waterways).

A compilation of German sewage sludge and compost data is given in Table 5. A first survey of German sewage sludges where potentially contaminated sludges should be targeted gave a mean concentration of 202 ng TEQ/kg d.m.: in 1990, most sludges were in the range 50-60 ng TEQ/kg d.m. The legal limit concentration for application on agricultural land is 100 ng I-TEQ/kg d.m. Composting of the total organic fraction from municipal waste collection results in a highly contaminated compost, not suitable for application in house gardens or in agriculture (mean concentration: 38 ng TEQ/kg d.m.). Compost from biowaste, kitchen wastes, or green wastes gives better qualities of approximately 14 ng I-TEQ/kg d.m. Such a mean value, however, is close to the guideline concentration of 17 ng I-TEQ/kg d.m.

Table 5: PCDD/PCDF in sewage sludge and compost

Sewage Sludge:	Limit value:	100 ng I-TEQ/kg d.m.
	1986/87:	202 ng I-TEQ/kg d.m.
	1990:	50-60 ng I-TEQ/kg d.m.
Compost:	Guideline value	17 ng I-TEQ/kg d.m.
	All Wastes:	38±22 ng I-TEQ/kg d.m.
	Biowaste	14±9 ng I-TEQ/kg d.m.

So far, hardly any country has done a reservoir inventory for PCDD/PCDF. First attempts were made by the city of Hamburg where stationary reservoirs were calculated (see Table 6, FHH 1995). In other words, there is almost no knowledge about the total amounts of PCDD/PCDF present in sinks such as sediments of harbors, rivers, lakes, and oceans, landfills or contaminated soils from (chemical) production sites. Reference is given within the country reports when data are available.

Table 6: PCDD/PCDF reservoirs in Hamburg; reference year 1992

Compartment	Reservoir (g I-TEQ)	Percentage of Total
Soil (without landfills and contaminated areas)	4,340	68.7 %
Sediments of the river Elbe and the harbor	1,980	31.3 %
Water of the river Elbe and the harbor	1.1	<0.01 %
Air	0.04	<<0.01 %
Vegetation	1	<0.01 %
Total	**6,322**	**100 %**

The PCDD/PCDF concentrations in the soil of Hamburg were found to be similar to those in other German cities. Levels above average were detected in an industrially impacted area in the south-east of Hamburg, on so-called ”Spülflächen“, where excavated sediments from the river Elbe were placed and other areas naturally flooded by the river Elbe. To calculate the dioxin reservoir present in the soil of Hamburg, the total area was split into use categories as shown in Table 7.

Table 7: PCDD/PCDF reservoirs in the soil of Hamburg

Unit	Area (km^2)	Reservoir (g I-TEQ)
Total area of Hamburg (without surface waters)	***695***	***4340***
Agricultural or horticultural use	**200**	**690**
Pasture / Grassland (Inside embankment)	50	60
Arable land (Inside embankment)	101	420
Gardens	49	210
Recreational areas, including forests	**123**	**140**
Forest	43	60
Lawn, parks, cemeteries	58	65
Sport fields / Surface soils	4	4
Children playgrounds	3	3
Areas under natural protection	15	8
Areas impacted by industrial emissions	**69**	**190**
Industrial areas	60	180
Open areas along streets	9	10
Other areas	**239**	**330**
Areas flooded in the past	31	2830
Areas on outside of embankments	11	960
Pasture / Grassland (Outside embankment)	9	760
Arable land (Outside embankment)	2	200
Spülfelder/Areas filled with sediments	20	1660
Bille-Siedlung (residential area)	0.3	210
Lower layers of walkways and sport fields	33	160
Sport fields	2	20
Roads	33	140

The PCDD/PCDF reservoir from Table 7 was calculated by multiplying the size of a given use pattern with the dioxin-relevant depth for agricultural (30 cm), garden (10 cm), horticultural

(20 cm) and other uses of soil (5 cm for litter in forest; 10 cm for forest soils and sports fields) with soil densities representative for the use pattern and typical PCDD/PCDF concentrations. A total dioxin reservoir of 4.3 kg I-TEQ was calculated to be present in the soils of Hamburg (Table 7). The Hamburg inventory contains the „Spülflächen“ but not the landfills and contaminated areas.

Other reservoirs include the former use of PCDD/PCDF-contaminated products such as 2,4,5-T (2,4,5-trichlorophenoxyacetic acid), polychlorinated biphenyls (PCB), and pentachlorophenol/ -phenate (PCP/PCP-Na). Although there are estimates of the total amount of these compounds produced for various purposes, it seems impossible to deduce from these numbers a quantitative impact of PCDD/PCDF to the environment or humans (Fiedler 1995). Within the city of Hamburg, the dioxin reservoir in abandoned waste dumps and former production sites is summarized in Table 8.

Table 8: Hamburg: Reservoirs in landfills and contaminated areas

Location	g I-TEQ
Moorfleeter Brack	1,400
Landfill Georgswerder	270,000
Landfill Müggenburger Straße	100,000
Dump Brümmer	360
Landfill Neuhöfer Straße	No estimate possible on present data basis
Ochsenwerder Landscheideweg	Remediated
Former Production Sites	
Boehringer production site	6,000

It should be noted that similar contaminations were found or can be expected in many other locations. As an example, She and Hagenmaier (1994) found almost 4,000 ng I-TEQ/kg in the sludges from a former chloralkali plant using graphite electrodes. However, so far, no quantification is available.

Although these reservoirs may be highly contaminated with PCDD/PCDF, the chemical-physical properties of these compounds imply that dioxins and furans will stay absorbed to organic carbon of soils or other particles. On the other hand, mobilization can occur in the presence of lipophilic solvents (⇒ leaching into deeper layers of soils and/or groundwater) or in cases of erosion or run-off by rain from topsoil (⇒ translocation into the neighborhood). Experience has shown that PCDD/PCDF transport due to soil erosion and run-off does not play a major role for environmental contamination and human exposure (Fiedler 1995).

5 TECHNICAL ABATEMENT TECHNOLOGIES

As with any other pollutants, reduction of PCDD/PCDF emissions from production facilities into the environment can be achieved in two ways: primary and secondary measures. Primary measures include steps which prevent formation of the pollutants; secondary measures remove the pollutants from entering the environment. Primary measures include:

- Substitution of substances or additives entering the production process
- Modification of the operation of the plant (for example: good combustion conditions such as the three T's = time, temperature, turbulence)
- Changes of the design of the plant.

Normally, these measures are applied first to prevent formation of unwanted byproducts. However, to comply with legal requirements, *e.g.* the limit value of 0.1 ng I-TEQ/m³ for waste incinerators as set by many European countries, additional measures have to be taken to meet the limit value. Such secondary measures were first developed for waste incinerators. Thus, most experiences with abatement technologies are available for municipal solid waste incineration. Technologies developed can be applied individually or in various combinations (It should be noted that compliance with a dioxin limit value is not the only critical number for combustion plants and the technologies developed very often also reduce emissions of other pollutants). Three techniques are capable of reducing dioxin concentrations to the limit value of 0.1 ng I-TEQ/m³ (Fiedler 1998):

- Adsorption of PCDD/PCDF on active carbon or coke
- Dry scrubbers with lime + active carbon followed by fabric filter
- Selective catalytic reduction (SCR) using a TiO_2-DENOX catalyst.

For further details and experiences from European countries, see LUA (1997). Chapters 8.6.1 and 8.12.2 discuss abatement technologies that were applied in Germany and the European Union as well. Such information was compiled to explain trends in PCDD/PCDF emissions within a country or to show potentials for further reduction.

6 LEGISLATIVE AND REGULATORY MEASURES

This chapter briefly addresses legislation and regulation of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF). In the various countries, steps have been undertaken to reduce formation of PCDD/PCDF in industrial and combustion processes or to reduce emissions of these compounds from the various sources. All efforts aim to minimize exposures of the environment and humans to PCDD/PCDF. There are several options for such action and most of them have been used in the past: legally binding instruments, guidelines and recommendations. In addition, industries have committed to changes in processes, to use cleaner input materials or to set maximum concentrations of PCDD/PCDF for their materials.

6.1 Tolerable Daily Intake (WHO)

Scientists today agree that the major pathway of human exposure to PCDD/PCDF accounting for >95 % of the human intake is *via* ingestion of food. Uptake of water and soil (toddlers!), inhalation, and dermal contact are of minor concern.

In May 1998, WHO evaluated the risks which dioxins may cause to health. Previously, in 1990, WHO experts had established a tolerable daily intake (TDI) of 10 pg TCDD/kg bw which subsequently was adopted by many countries. Since then, new epidemiological data have emerged, especially on neurotoxicological development and the endocrine system. Finally, WHO experts agreed on a new TDI of 1 to 4 picogram per kilogram body weight per day based on new scientific evidence. The experts however recognized that subtle effects occur in the general population in developed countries at background levels of 2 to 6 picograms per kilogram body weight and day. Therefore, WHO recommended that every effort should be made to reduce exposure to the lowest possible level. In addition, the range established for 2,3,7,8-Cl_4DD should be applied to a TEQ including the 17 2,3,7,8-substituted PCDD and PCDF as well as the coplanar and *mono-ortho* substituted PCB (WHO 1998).

6.2 Indirect Measures

As polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans have never been produced intentionally on an industrial scale or for any commercial application, the production of PCDD/PCDF cannot be prohibited or phased out by law. Therefore, indirect measures have been taken to reduce new inputs of PCDD/PCDF into the environment. The first laws addressed the ban of chemicals known to be contaminated with PCDD/PCDF (ppb to ppm-range I-TEQ): As a consequence most industrialized countries banned the production and use of polychlorinated biphenyls (PCB) and pentachlorophenol (PCP). Whereas PCB is banned in most industrialized countries, PCP is still being produced and used outside the European Union.

6.3 Direct Measures

6.3.1 Chemicals

In Germany, the First Ordinance on the Prohibition of Certain Chemicals has set stringent limit values for eight PCDD/PCDF in substances, preparations and articles placed on the market. This regulation was amended in 1994 and 1996 (ChemVerbotsV 1996) and the present ordinance sets limit values for all seventeen 2,3,7,8-chlorine substituted congeners as well as for eight of the lower brominated 2,3,7,8-bromine substituted dibenzo-*p*-dioxins and dibenzofurans, PBDD/PBDF). According to the law, substances, preparations and/or articles are not allowed to be placed on the market:

- if the sum of the concentrations of the congeners listed in column 1 under No. 1 of column 1 (Table 9) exceeds a value of 1 µg/kg, or
- if the sum of the concentrations of the congeners listed in column 1 under Nos. 1 and 2 exceeds a value of 5 µg/kg, or
- if the sum of the concentrations of the congeners listed in column 1 under Nos. 1, 2 and 3 (= all congeners substituted in 2,3,7,8-position), exceeds a value of 100 µg/kg.

Table 9: Limit values of PCDD/PCDF as given by the Chemicals Law
Note: the concentrations given in Table 9 are absolute values, not I-TEQ!

	Congeners	Maximum Concentrations
No. 1	2,3,7,8-Cl_4DD, 1,2,3,7,8-Cl_5DD, 2,3,7,8-Cl_4DF, 2,3,4,7,8-Cl_5DF	Sum of congeners under No. 1: 1µg/kg
No. 2	1,2,3,4,7,8-Cl_6DD, 1,2,3,7,8,9-Cl_6DD, 1,2,3,6,7,8-Cl_6DD, 1,2,3,7,8-Cl_5DF, 1,2,3,4,7,8-Cl_6DF, 1,2,3,7,8,9-Cl_6DF, 1,2,3,6,7,8-Cl_6DF, 2,3,4,6,7,8-Cl_6DF	Sum of congeners under Nos. 1 and 2: 5 µg/kg
No 3	1,2,3,4,6,7,8-Cl_7DD, 1,2,3,4,6,7,8,9-Cl_8DD, 1,2,3,4,6,7,8-Cl_7DF, 1,2,3,4,7,8,9-Cl_7DF, 1,2,3,4,6,7,8,9-Cl_8DF	Sum of congeners under Nos. 1, 2 and 3: <100 µg/kg

Similar regulations exist in the United States where in the Toxic Substances Control Act; maximum permissible concentrations for 2,3,7,8-TCDD were set.

6.3.2 Regulations on Incineration and Combustion

As incineration of wastes was considered to be a major source of PCDD/PCDF emissions into the environment, legally binding concentrations for stack emissions have been established in several countries. Whereas first regulations only included a requirement to minimize PCDD/PCDF emissions by applying Best Available Techniques (BAT), countries have moved towards defined limit values. Very often, these laws also include requirements on sampling, analysis and reporting methods. In general, new incinerators have to comply immediately with these limits. For existing incinerators there are transient times between three and six years to comply with regulations.

A legally binding limit value of 0.1 ng I-TEQ/m³ for PCDD/PCDF is asked for hazardous waste incinerators by Directive 94/67/EC. Within two years, the member states of the European Union have to translate the requirements of this directive into national law. It can be assumed that today, all 15 member States have such legislation in place. Countries like Austria, Belgium, Germany, Finland, France, Italy, the Netherlands, and Sweden went beyond this requirement and included municipal waste incinerators, sewage sludge and hospital waste incinerators into their national laws. The remaining countries have the 0.1 ng I-TEQ/m³ limit as a guideline concentration.

In the USA, there is a limit value of 0.2 ng TEQ/m³ for new municipal and of 0.15 ng I-TEQ/m³ for new hazardous waste incinerators.

In its new law, Japan took a tiered approach: new incinerators with a capacity greater than 4 t/h have to comply with a limit concentration of 0.1 ng I-TEQ/m³ (12 % O_2) whereas the limits for MSWI <2 t/h and MSWIs with a capacity between 2-4 t/h are 5 ng I-TEQ/m³ and 1 ng I-TEQ/m³, respectively.

Some countries have issued additional laws:

Austria: a limit value of 0.4 ng I-TEQ/m³ was set for sinter plants built after 1 January 2001 (Austria 1997a). For iron and steel plants, there is a limit value of 0.25 ng I-TEQ/m³ until 31 December 2005. Limit value of 0.1 ng I-TEQ/m³ from 1 January 2006. For electric arc furnaces and induction ovens the limit is at 0.4 ng I-TEQ/m³. For existing plants, the latest date to comply with is five years after publication of the law (Austria 1997b).

Germany: in 1997, a dioxin limit value of 0.1 ng I-TEQ/m³ and a minimum temperature of 850 °C for crematories was set by law (27th BImSchV 1997).

Canada: a guideline concentration of 0.5 ng TEQ/m³ was set for municipal waste, hazardous waste and clinical waste incinerators (CCME 1989 and 1992, respectively)

Japan: electric furnaces used for metal smelters are controlled through the Air Pollution Control Law with 0.5 ng I-TEQ/m³ for newly established furnaces and 1 ng I-TEQ/m³ for existing ones.

6.4 Further Regulations

To stop the entry of dioxins and furans into the environment from use of so-called scavengers (*e.g.* dichloroethane or dibromoethane) as additives in leaded gasoline, a ban of the use opf such scavengers was implemented in 1992 in Germany (19th BImSchV 1992).

It is known that sewage sludge is contaminated with PCDD/PCDF. To reduce input of PCDD/PCDF through application of sewage sludge, Germany established a limit value of 100 ng I-TQ/kg dry matter for sewage sludges used as fertilizer in agriculture, horticulture or forestry ((AbfKlärV 1992). In addition, the law sets a freight limit for 5 tons of dry matter of sewage sludge per hectare once within three years. Application of sewage sludge on pasture is forbidden by law.

Similarly, Austria established maximum concentration of 50 ng I-TEQ/kg in fertilizers, soil additives, culture substrates, and plant additives. In addition, products containing 20-50 ng TEQ/kg have to be labeled with a warning sign “Attention contains dioxins/furans“ (forbidden for use on children’s playgrounds). Culture substrates are not allowed to contain more than 20 ng TEQ/kg (Austria 1994). In addition, the States of Oberösterreich (o.Ö.) and Niederösterreich (N.Ö.) published ordinances with a limit value of 100 ng I-TEQ/kg d.m. for sewage sludge.

6.5 Guidelines for Soil and Milk

Guidelines and recommendations have been established for sensitive uses such as soil and dairy products. The proposed measures and guidelines are recommendations for action but they are not legally binding. Nevertheless, they are a basis for political decisions to protect human health and the environment. In some cases, *e.g.* accidents such as a fire at a plastic store, these recommendations for actions were used for decision making.

6.5.1 Soil

The guideline concentrations established by Germany, the Netherlands, and Sweden are compiled in Table 10.

In Germany, the first report of the Joint [Dioxin] Working Group was published in 1992 and contained the reference values and recommended action for agricultural and horticultural land uses (Table 10, BLAG 1992):

- For preventive reasons and as a long-term objective, the dioxin concentrations of soil used for agricultural purposes should be reduced to below 5 ng TEQ per kg;
- Cultivation of foodstuffs is not restricted in the case where the soil contains 5-40 ng per kg. However, critical land uses, e.g. grazing management, should be avoided if increased dioxin levels were found in foodstuffs grown on such soils;
- Limitations on the cultivation of certain feedstuffs and foodstuffs might be necessary if the dioxin contamination were above 40 ng TEQ per kg soil. However, unlimited cultivation is allowed for plants with minimum dioxin transfer, *e.g*. corn.

Guideline values were established for measures to be taken on children’s playgrounds and in residential areas:

- Remediation of contaminated soil is required in playgrounds if the soil contains more than 100 ng TEQ per kg. Remediation means sealing, decontamination or exchange of soil;
- In residential areas, such action should be taken if the soil is contaminated with more than 1,000 ng TEQ per kg;
- In industrial areas, the limit value was set to 10,000 ng TEQ per kg.

The recommendations have been translated into governmental decrees in a number of Länder (Federal States in Germany).

In 1988, the Netherlands published a guideline for classification and remediation of soil (Leidraad Bodemsanering 1988). Since its revision in 1994, there is a guideline level for PCDD/PCDF of 1 ng I-TEQ/kg d.m. for agricultural use and of 10 ng I-TEQ/kg d.m. for dairy farming.

In Sweden, there is a guideline concentration of 10 ng I-TEQ/kg d.m. for sensitive uses and of 250 ng I-TEQ/kg d.m. for less sensitive uses.

Table 10: Soil guideline concentrations (Concentrations in ng I-TEQ/kg d.m.)

Germany	
< 5	Target concentration
5-40	Control of products if dioxin transfer
>100	Soil exchange on children playgrounds
>1 000	Soil exchange in residential areas
>10 000	Soil exchange independent of the location
The Netherlands	
1	Agricultural farming
10	Dairy farming
Sweden	
10	Sensitive uses
250	Non-sensitive uses

6.5.2 Milk and Milk Products

Germany, Ireland, The Netherlands, and the United Kingdom established guidelines for PCDD/PCDF concentrations and recommendations in milk. The guideline concentrations are summarized in Table 11.

For Germany, the second report of the Joint Working Group contained guidelines and maximum values for milk and dairy products together with recommendations for action (BLAG 1993).
The limit values as given in Table 11 were derived as follows:

- based on a TDI (total daily intake) value of 10 pg 2,3,7,8-TCDD/kg body weight and day, the maximum dioxin concentration in milk should not exceed 5.0 pg TEQ/g milk fat. Thus, milk and dairy products should not be out on the market if the dioxin contamination exceeds this value. However, from the existing data it is obvious, that a limit value of 5 pg TEQ/g milk fat would cut off only a few cases of extreme contamination, this limit value was considered not to have any consequences at all and would not improve the consumer’s situation;
- to reduce the human impact via consumption of dairy products, a limit value of 3 pg TEQ/g fat was set. If such concentration were exceeded, the dioxin source should be identified and emission reducing measures taken. Moreover, it was recommended that direct supply to the consumer be stopped for milk and dairy products containing more than 3 pg TEQ/g fat; and
- finally, an orientation value of 0.9 pg TEQ/g milk fat, based on the principle of precaution, was set. This concentration was derived from a TDI of 1 pg TEQ/kg body weight and day. The value of 0.9 pg TEQ/g milk fat can only be regarded as target value to be achieved as the data from Germany and other European countries showed that more than 50 % of all dioxin concentrations in milk would exceed this value. To reach this target, it is necessary to further reduce the dioxin release into the environment.

Table 11: Recommendation values and action levels for PCDD/PCDF in milk and milk products. (Concentrations in ng I-TEQ/kg milk fat)

Germany	
< 0.9	Target concentration (minimum of dioxin input)
> 3.0	1. Identification and reduction of sources. If not possible within a short time → stop dairy farming 2. Recommendation not to market milk
>5.0	Milk and milk products are not allowed to be marketed
Ireland	
5.0	Maximum level
The Netherlands	
6.0	Maximum level
United Kingdom	
0.7	Milk and milk products with less than 2 % fat
16.6	Maximum concentration for milk and milk products

7 METHODS TO ESTABLISH DIOXIN INVENTORIES

The data on emissions of dioxins and furans, more precisely polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF), compiled in this report are based on a multitude of sources. This variety is also reflected in the depth of the information available. For some countries (*e.g.* Canada, Denmark, the Netherlands, Belgium, Australia, and Germany) full reports were available given in-depth information on generation of data and aggregation. Unfortunately, some reports are only available in the original language (*e.g.* Belgium, the Netherlands). Sometimes, the emission inventories (or the most recent updates) were taken from references found in the scientific literature (*e.g.* Korea and the United Kingdom). In such cases, less background information is available on how data were generated and how extrapolation to a nationwide basis was performed. The information for Austria and France was found in the Internet on the homepage of the Environmental Agency. Extracts of national emission inventories were provided by the Czech Republic (data together with the Slovak Republic), Hungary, and the Slovak Republic. Very often not more than the “pure” data were available. For two countries, USA and Sweden, the national emission inventories are still in a draft stage. Nevertheless, the USA has distributed several hundred pages of background information (including an electronic database) of the draft report. In addition, data are based on personal communications as some reports are not yet finished, *e.g.* Sweden.

In this report, concentrations of dioxins and furans are given in International Toxicity Equivalents (TEQ) using the International Toxicity Equivalency Factors (I-TEF) as established by the NATO/CCMS Working Group on Dioxins and Related Compounds in 1988 (NATO/CCMS 1988a) (the list of the I-TEFs is Appendix A).

Most authors used the same approach to calculate the annual emission from a given source:

$$\textit{Emission of source} = \textit{Emission factor} \times \textit{“Activity rate”} \qquad (1)$$

Or

$$\textit{Emission of source} = \textit{Concentration in emission} \times \cdot\, \textit{Operational hours} \qquad (2)$$

The PCDD/PCDF emission per year is given in grams TEQ per year. According to equation (1), the annual flux is calculated by multiplying the release of PCDD/PCDF (*e.g.* in µg I-TEQ) per unit of feed material processed or produced (*e.g.* ton or liter) with the amount of feed material processed or produced (ton per year). A second method – equation (2) – calculates the annual emission of a source by multiplying the measured emissions (*e.g.* in ng I-TEQ/m^3) with the operational hours per year.

In order to make a source emission estimate, information is required for

a) Representative dioxin measurements for a given source. In many cases, these estimates are very uncertain as emissions are deduced from a few test results only and extrapolated to a nation-wide basis. In other cases there are no data existing and thus, either numbers from the literature were taken or emissions derived from similar processes.

b) The total amount of feed material processed or produced. Very often, there is a high degree of certainty for such information, *e.g.* all fields of waste disposal, paper and pulp production, fuel consumed, *etc*.

A second approach to calculate annual PCDD/PCDF emissions is to multiply measured concentrations with annual operational hours of a plant and the total volume of exhaust gases emitted.

Presently, there exist no harmonized methods how to generate and how to evaluate PCDD/PCDF inventories. As a consequence, some countries use lower and upper ranges to report the data whereas other countries report mean or median values.

Further differences occur in the way domestic and external data are handled: *e.g.* the United States did not take into account emission data from foreign countries or the non-peer-reviewed literature (*e.g.* from other agencies). Considering the information contained in these data sources would have helped to address uncertainties in the estimates and fill data gaps for release estimates. Other countries, like Australia, almost completely rely on foreign data to establish the national emission inventory.

All PCDDD/PCDF inventories compiled in this report attempt to quantify emissions to the air. A few countries such as the United Kingdom, Canada, Sweden include releases to water and land/landfills or with products. The United Kingdom, the Netherlands and the United States to a certain extent also include releases from reservoirs, *e.g.* PCP in wood.

8 EUROPEAN DIOXIN INVENTORIES

8.1 Austria

In Austria, it is estimated that a total of 28.7 g I-TEQ was emitted from known sources in the year 1994 (UBAVIE 1996). The emissions of the various sectors are summarized in Table 12. The sector of non-industrial combustion plants emits 16.7 g I-TEQ corresponding to 58 % of the total emissions. About 92 % of the dioxin emissions are produced from combustion of fuel-wood. The relative contribution from residential plants amounts to more than 49 % of the PCDD/PCDF emissions of this sector.

Table 12: PCDD/PCDF emissions in Austria. Fluxes in g I-TEQ/a, reference year 1994 (UBAVIE 1996)

Source Category	(g I-TEQ/a)
Combustion in energy and transformation industries	0
Non-industrial combustion plants	16.67
Combustion in manufacturing industry	1.50
Production processes	10.360
Road transport	0
Waste treatment and disposal	0.179
Nature	0.024
Total	**28.733**

8.2 Belgium

A study prepared by De Fré and Wevers (1995) describes the trends of dioxin emissions for the three regions in Belgium in the period 1985-1995. In 1990 it was agreed to reduce the flux of dangerous substances to the sea *via* air, water, and waste by the following means:

- Emission reductions of all persistent, toxic, and bioaccumulating substances from all anthropic sources into the marine environment as agreed in the London Declaration. Before the year 2000, all discharges of these substances are to be decreased below levels that are harmful to man and environment.

- A significant reduction of 50 % or more of the influx *via* rivers and estuaria between 1985 and 1995 and of all atmospheric emissions by 1995 or at the latest by 1999 should be achieved.

- A decrease of discharge *via* all pathways of 70 % or more between 1985 and 1995 for those substances that form a major threat to the marine environment, and at least for dioxins, mercury, cadmium, and lead, provided that the Best Available Technology, or other technologies which generate limited amounts of waste enable such reductions.

The Belgium emission inventory of PCDD/PCDF was compiled in 1995 and summarizes information on dioxin releases to air and waters for 15 sectors (De Fré and Wewers 1995). The emission inventory estimates are based on measurements available in Belgium and on emission factors from international literature. Incineration of domestic and hospital wastes are the main sources of PCDD/PCDF in Belgium, for which also the most reliable data are available. For most other sectors, there is a larger range of uncertainty of the estimates.

The 1995 report summarizes the PCDD/PCDF emission estimates for the years 1985, 1990, and 1995. The overview on these estimates to air is given in Table 13. The last two columns give the Confidence Rating in terms of emission factors and mass of fuel processed in a given plant.

Table 13: Belgium: PCDD/PCDF emissions to air; reference years 1985-1995
All fluxes are given in g I-TEQ/a

	1985	1990	1995	Precision *	
				E-Factor	Volume
Waste Incineration					
Municipal waste	297	385	187	0	0
Medical waste	100	100	95	1	1
Industrial waste	7.65	11.2	20.9	1	1
Sludge incineration	1.45	0.70	0.75	1	1
Industry					
Coke production	1.75	1.59	1.07	2	0
Sintering and agglomeration	62.5	67.5	53.2	2	0
Electrosteel	2.59	4.89	6.42	2-3	0
Non-ferrous	80	109	107	2	1
Cement kilns	16.1	19.7	20.8	2	0
Lime kilns	30.4	34.6	33.4	2	0
Paper and pulp bleaching	0.04	0.05	-	2	1
Chemical industry					
Chloralkali plants	-	-	-		
Production of vinyl chloride	0.447	0.455	0.05	1	0
Energy					
Space heating	228	138	122	2-3	0
Industrial combustion facilities	10.16	8.64	7	2	0
Production of electricity	1.54	2.11	2.31	1	0
Road traffic	6.08	4.76	1.71	1	0
Others					
Crematories	0.08	0.13	0.19	1	0
Fires	3.36	3.22	2.56	2	2
Biogas	0	0	0.012	2	1
Total	**850**	**892**	**662**		

* Precision

- Emission Factor: the precision is given in exponential numbers: Zero is the highest degree of precision. In cases where only one or two measurements were available, the confidence rating was given a “2”.
- Volume: 0 = Exactly known from either data from all facilities or national statistics.
  1 = statistics incomplete and completed with estimations
  2 = No data available, volume was estimated

The PCDD/PCDF emissions to water for the reference period 1985-1995 are summarized in Table 14.

Table 14: Belgium: PCDD/PCDF emissions to water; reference years 1985-1995
All fluxes are given in g I-TEQ/a

	1985	1990	1995	Precision *	
				E-Factor	Volume
Waste Incineration					
Municipal waste	0.0305	0.0471	0.0575	1	1
Medical waste	-	-	-		
Industrial waste	0.0024	0.010	0.021	1	1
Sludge incineration	-	-	-		
Industry					
Coke production	0.37	0.33	0.23	2	0
Sintering and agglomeration	3.75	4.05	3.19	2	0
Electrosteel	-	-	-		
Non-ferrous	-	-	-		
Cement kilns	-	-	-		
Lime kilns	-	-	-		
Paper and pulp bleaching	1.81	2.18	-	1	0
Chemical industry					
Chloralkali plants	0.03	0.03	0.03	1	1
Production of vinyl chloride	0.201	0.205	0.222	1	1
Energy					
Space heating	-	-	-		
Industrial combustion facilities	-	-	-		
Production of electricity	-	-	-		
Road traffic	-	-	-		
Others					
Crematories	-	-	-		
Fires	0.0198	0.0196	0.0151	2	2
Biogas	-	-	-		
Total	**6.21**	**6.87**	**3.77**		
Sewage Water	18.2	17.1	9.74	2	1

The most important PCDD/PCDF sources to air, water, and waste for the three regions in Belgium are compiled in Table 15. The most important source of PCDD/PCDF in Belgium is still municipal solid waste incineration, followed by space heating, and the non-ferrous metal industry. The last column in Table 15 gives the percentage of source reduction for the two reference years 1985 and 1995.

Table 15: Belgium: PCDD/PCDF emissions to air, land, and wastes for the three Regions by sectors (decreasing importance). All fluxes in g I-TEQ/a

	1985	1990	1995	% Change
Municipal Waste Incineration				
Flanders				
To air	162	200	50	-69
To water	0.0234	0.0372	0.0467	+99.5
To waste	192	242	234	+22
Walloon				
To air	32.2	62	11.5	-64
To water	0.0071	0.0099	0.0108	+52
To waste	34	64	108	+218
Brussels				
To air	103	123	125	+21
To water	-	-	-	-
To waste	108	64	130	+20
Belgium				
To air	297	385	187	-37
To water	0.0305	0.0471	0.0575	+88.5
To waste	334	434	472	+41
Space Heating: Emissions to Air				
Flanders	104.2	62.0	53	-49
Walloon	111	68.9	63	-43
Brussels	12	7.25	6.2	-48
Belgium	228	138	122	-46.5
Non-ferrous metal Industry: Emissions to Air				
Flanders	50.38	68.26	67.27	+33.5
Walloon	22.29	30.27	28.89	+29.6
Brussels	7.33	10.15	10.64	+45
Belgium	80	109	107	+33.8
Clinical Waste Incineration: Emissions to Air				
Flanders	67.5	67.5	62.5	-7.4
Walloon	15.7	15.7	15.7	0
Brussels	16.9	16.9	16.9	0
Belgium	100	100	95	0
Steel Industry				
Flanders				
To air	25.7	31.8	30.8	+20
To water	1.51	1.79	1.66	+10
Walloon				
To air	41.1	42	29.9	-27
To water	2.56	2.55	1.74	-32
Brussels				
To air	0.23	0.2	-	-100
To water	0.048	0.042	-	-100
Belgium				
To air	67	74	60.7	-9.4
To water	4.12	4.38	3.40	-17.5

Table 15 (continued)

	1985	1990	1995	% Change
Incineration of Industrial Waste				
Flanders				
To air	0.36	5.76		+4,761
To water	0.0001	0.008		+19,900
To waste	0.007	0.064		+3,271
Walloon				
To air	0.09	0.09	0.09	0
To water	-	-	-	-
To waste	-	-	-	-
Brussels				
To air	7.2	5.34	3.31	-54
To water	0.0023	0.0017	0.001	-56.5
To waste	0.135	0.1	0.062	-54
Belgium				
To air	7.65	11.2	20.9	+173
To water	0.0024	0.010	0.021	+775
To waste	0.142	0.164	0.298	+110
Cement Kilns: Emissions to Air				
Flanders	0.5	0.60	0.60	+20
Walloon	15.6	19.1	20.2	+29
Belgium	16.1	19.7	20.8	+29
Lime Kilns: Emissions to Air				
Walloon	30.4	34.6	33.4	+9.9
Belgium (margins)	30.4 (10.5-49.7)	34.6 (11.9-56.5)	33.4 (11.5-54.5)	+9.9

A summary of all PCDD/PCDF emissions by regions is shown in Table 16. It can be seen that the overall reduction of PCDD/PCDF for air emissions was 22 % from 1985 to 1995. This means that the goal of a 70 % reduction was not achieved. The major reason for such result is that in 1985 only a few dioxin sources were identified, and by 1995, many additional sources were discovered, *e.g.* the sectors of the metal industry and the non-ferrous metal industry. In addition, it takes several years until measures for emission reductions are fully implemented. For example, once all municipal solid waste incinerators in Belgium will comply with legal requirements, the emissions from this source will be reduced to 0.95 g I-TEQ/a, which represents a drop of 99.7 % from the 1985 emissions.

In the report from Belgium it was found that only a limited number of sources were identified that have emissions to water. Generally, the emission factors for these sources were much lower than those to air and waste emissions. Nevertheless, the PCDD/PCDF concentrations in municipal sewage sludges from Belgium indicate dioxin emissions from household and industrial wastewaters probably cause higher emissions into surface waters than the primary sources shown in Table 14.

Table 16: Belgium: Summarizing table - PCDD/PCDF emissions to air, water, and waste for the three Regions; reference years 1985-1995. All fluxes in g I-TEQ/a

	1985	1990	1995	% Reduction
Flanders				
To air	425	448	291	31.5
To water	1.58	1.88	1.78	+12.7
To waste	201	251	243	+21
Walloon				
To air	277	281	208	25
To water	4.58	4.95	1.99	56.5
To waste	38.3	68.3	112	+192
Brussels				
To air	148	164	163	+10
To water	0.05	0.045	0.002	96
To waste	108	128	130	+20
Belgium				
To air	850	892	662	22
To water	6.21	6.87	3.77	39
To waste	347	447	485	+40

8.3 Czech Republic and Slovak Republic

Estimated PCDD/PCDF emissions for the Czech and the Slovak Republics combined (former Czechoslovakia) were calculated in 1993 and 1995 (Holoubek *et al.* 1993, Parma et al. 1995). First semi-quantitative estimates of POPs emissions in the Czech Republic and the Slovak Republic were determined to 30.25 g I-TEQ/a

Table 17: PCDD/PCDF emission estimates combined for the Czech Republic and the Slovak Republic. Concentrations in g I-TEQ/a

Source	Emission Estimate (g I-TEQ/a)
Residential fuel combustion	
Anthracite	0.81
Brown coal	11.35
Grey iron foundries	0.74
Municipal waste incinerators	
Czech Republic	3.6
Slovak Republic	6.00
Industrial waste incinerators	7.68
Incineration of waste oil	Input and output parameters unknown
Sewage sludge incineration	Combusted amount unknown
Incineration of hospital waste	unknown

8.4 Denmark

The 1997 Working Document ”Dioxins, Sources, Levels and Exposures in Denmark“ (Ministry of Environment and Energy 1997) gives an update of an earlier report published in Danish language in 1995. The total PCDD/PCDF emissions to the atmosphere in 1995 from all known sources were estimated at about 50 g I-TEQ. Municipal solid waste incineration is the largest atmospheric dioxin source with an emission of 20 g I-TEQ/a. The other main pollution sources are incineration of hospital clinical wastes/healthcare risk wastes with an air emission of 5 g I-TEQ, and a steel reclamation plant with an annual air emission of 7.5 g I-TEQ. Based on the limited data Danish dioxin emissions seem to be decreasing.

The present inventory does not include Greenland and the Faroe islands. The Ministry of Environment and Energy plans for a broader dioxin study to be performed in the year 2001, which then will also evaluate the situation in these two areas.

The main sources (2 g I-TEQ/a) of soil pollution by dioxins are application of sewage sludge and compost to agricultural land. Most dioxin residues are found in fly ash from coal firing.

The new data is mainly based on very few measurements (spot tests) of a few items, such as wood stoves, biofuels, sewage sludge, and textiles. Based on the available data, it is concluded that dioxin emissions to the Danish environment from known sources are lower than reported earlier. The total Danish dioxin emission to the atmosphere in 1991 from all known sources was estimated to about 100 g N-TEQ. The corresponding figure for 1995 was less than 50 g I-TEQ. For the purpose of a dioxin inventory, the difference in applying Nordic TEFs (N-TEFs) or the International TEFs (I-TEFs) is negligible as the two schemes differ only in a single congener: In the N-TEF scheme 1,2,3,7,8-Cl_5DF is given a TEF of 0.01 whereas in the I-TEF scheme a value of 0.05 is assigned to this specific congener. So far, there is no source identified where this congener plays an important role.

Table 18: Sources of PCDD/PCDF in Denmark; reference year 1995 (Ministry of Environment and Energy 1997)

	Emission to (g I-TEQ/a)				
Activity/ Process	Air	Water	Soil	Waste/Residue	Total
Chemical industry	-	-	-	-	-
Paper and pulp industry	-	-	-	-	-
Asphalt-mixing industries	0.1	-	-	-	0.1
Steel and metal industry					
- steel reclamation	7.5	-	-	34	42
- shredder plants	0.13	-	-	?	?
- cable scrap burning		-	-	-	0.13
Cement industry	0.08-1.5	-	-	-	0.08-1.5
Fossil fuels					
- pit coal	2	-	-	40	42
- other	-	-	-	-	-
Burning biofuels					
- straw	0.07-6.6	-	-	-	0.7-6.6
- woods	0.25	-	-	-	0.25
Wood stoves	1.1	-	-	-	1.1
Heating of houses, gas oil	0.02	-	-	-	0.02
Building, landfill, vehicle fires	?	-	-	-	?
Municipal solid waste	20	-	-	100	120
Incineration of hospital/ healthcare risk waste	5	-	-	-	5
Incineration of hazardous waste					
- hazardous waste+waste oil	0.23	-	-	1.0	1.23
- waste oil (district heating)	0.038	-	-	-	0.038
Emission from crematoria	0.16				0.16
Sewage sludge					
- sludge landfilled	-	-	-	0. 19	0.19
- sludge amended to soil	-	-	1.1	-	1.1
- sludge incinerated	0.072	0.108	-	0.036	0.22
Compost	-	-	1-5	-	1-5
Traffic	0.2	-	-	-	0.2
Textile products	-	0.08	-	-	0.08
Air deposition	-	-	120	-	120

8.5 France

The French Ministry of Regional and National Development and the Environment together with the national Environment Agency (ADEME) has initiated a study to identify and quantify emissions of PCDD/PCDF into air. Preliminary results from a limited number of sources quantified so far are available *via* the internet (Adème 1999). The summary of annual emissions from these sources for the reference year 1997 is displayed in Table 19. The major source identified in France is municipal waste incineration, which contributes to 400 g I-TEQ annually according to sources of Adème whereas CITEPA (Centre interprofessionel technique d'études de la pollution atmosphérique) estimates 510 g I-TEQ annually. These emissions represent 30-40 % of the total inventory.

Table 19: France: PCDD/PCDF emissions to air; reference year 1997 (Adème 1999) Numbers in italics are estimates from Centre interprofessionel technique d'études de la pollution atmosphérique (CITEPA)

Sector	Emission per Year (I-TEQ/a)
Municipal solid waste incinerators	400 *510*
Industrial/hazardous waste incinerators	2
Industrial combustion processes. Combustion of	
• Wood	4
• Coal	9
• Fossil fuels	*10*
Metallurgical processes	
• Iron melting	*220-2200 (400)*
• Iron and steel foundries	1.2-80 *(20)*
• Secondary aluminum	*10*
• Secondary copper	*10*
• Secondary zinc	*7*
Traffic (automobiles)	*1-5*

8.6 Germany

8.6.1 Federal Republic of Germany

In 1997, the Environmental Agency (UBA) published a PCDD/PCDF emission inventory for Germany (Johnke 1997) with past and future trends (Table 20).

In the 1980s, total emissions in the Federal Republic of Germany ranged from 1 kg I-TEQ/a to 2 kg I-TEQ/a. Since the early 1990s, a continuing decrease in dioxin emissions was observed. Whereas in the beginning of the 1990s, total emissions of PCDD/PCDF were still estimated at more than 1,000 g I-TEQ/a, the estimate for 1994/95 was around 300 g I-TEQ/a. It is expected that dioxin emissions will again decrease considerably to values markedly lower than 70 g I-TEQ/a by the year 2000

A dramatic decrease was observed for waste incinerators since the beginning of the 1990s. Whereas the central estimate for PCDD/PCDF emissions was 400 g I-TEQ for the year 1989/90, retrofitting of existing plants, closure of a few older plants and building of some new MSWIs reduced the dioxin emission by approximately 90 % in the year 1994/95. From 1997, when all existing MSWI have to comply with the emission limit value of 0.1 ng I-TEQ/m^3 (17th BImSchV 1990), it can be assumed that the total dioxin emissions from all municipal solid waste incinerators in Germany are less than 4 g I-TEQ/a. Based on an average emission concentration of 0.015 ng I-TEQ/m^3 and an overall capacity of 15.9 Mio. t/a, total emissions from MSWI will be 1.3 g I-TEQ/a or approximately 2 % of the total inventory (less than 70 g I-TEQ/a) (Johnke 1997, 1998).

As a result of the requirements of the 17th BImSchV, which stipulates an emission limit value of 0.1 ng TEQ/m^3, marked reductions have been obtained in dioxin emissions from waste incinerators. For 1998 - following the retrofitting of all existing plants - a total flux of less than 4 g I-TEQ/a was estimated (based on a stack concentration of 0.05 ng TEQ/m^3 and an overall capacity of 13.6 mio. t/a). It is estimated that waste incineration's share of total dioxin emissions by the year 2000 (based on the value of 0.015 ng TEQ/m³, overall capacity of 15.9 Mio. t/a) will amount to around 2 % (1.3 g/a of a total of <70 g/a).

Table 20: Trends of PCDD/PCDF emissions in Germany (Johnke 1998). Fluxes in g I-TEQ/a

	1989/90	1994/95	Estimate 1999/2000
Waste Incineration	400	32	<<4 (<2)
Metal production and processing, total	750	220	<40
Secondary aluminum	25	18	<1
Smelting plants in foundries	3	2	<2
Sintering plants	575	158	<20
Steel - Oxygen plants	5	4	<3
Steel - Electro	30	5	<2
Others	112	33	<12
Power Plants	5	3	<3
Industrial and Commercial Boilers	20	15	<10
Other Thermal Industrial Processes	1	<1	<1
Crematories	4	2	<1
Traffic	10	4	<1
Domestic Furnaces	20	15	10
Total	**1210**	**291**	**<<70**

A more detailed description of the situation in Germany is given in a report by the IFEU Institute (1998). On behalf of the Federal Environmental Agency (UBA), the Ifeu-Institute calculated total emissions of PCDD/PCDF and polycyclic aromatic hydrocarbons emitted from German sources for the reference year 1994 (Ifeu 1998). Here, the results for PCDD/PCDF will be summarized. The goals of the study were:

- Identification of relevant sources
- Determination of emissions factors for each source
- Total emissions for the reference year 1994
- Identification of state-of-the-art technology for POPs emission reduction
- Quantification of emission reduction potential

For the reference year 1994, the emissions from the ferrous and non-ferrous metal industries were the major contributors to the total dioxin emissions for Germany (see Table 21).

The category named iron, steel industries and foundries includes sinter plants, steel ovens, and steel works and coke plants (as in Germany the total coke production is used by the steel works). Measured PCDD/PCDF emissions below 0.1 ng I-TEQ/m³ are available from one coke plant. However, it was considered that this one plant is not representative of the majority of existing plants in Germany. Thus, for the inventory a 7.5-fold higher emission was assumed. For sinter plants, there are two emission streams to quantify; dedusting of the sinter band and of the hall.

In Germany, approximately 20 % of the total raw steel production is from electro-steel. Scrap materials can be 100 % of the input to these plants.

Table 21: Germany: PCDD/PCDF emissions from iron steel industries and foundries; reference year 1994 (Ifeu 1998)

Source Category	Emission Factor µg I-TEQ/TJ coke µg I-TEQ/t material, resp.	Activity Rate TJ coke/a, t/a, resp.	Flux g I-TEQ/a
Iron and steel and foundries			**181**
Coke plants	8.65	347095	3.00
Sinter plants			168
Dedusting of sinter bands	6.00	25512756	153
Dedusting of indoor environments	00.59	25512756	15.1
Steel plants			0.79
Cowpers	0.021	29923000	0.63
Indoor environments	0.0054	29923000 t/a	0.16
Oxygen steel works			3.72
Primary emissions			
Secondary emissions			
Electro-steel plants	0.48	8940941 t/a	4.33
Iron and steel foundries			1.18
Cold air cupolas	1.11	770474	0.85
Hot air cupolas	0.20	1464954	0.30
Electric arc and induction furnaces	0.032	903651	0.029

In Germany, there are no plants for production of primary aluminum, lead, and zinc production plants (LUA 1997, Ifeu 1998). For the secondary aluminum production, aluminum scrap, chips, and other Al-containing materials are melted in the presence of a salt mixture. Relatively broad ranges of PCDD/PCDF emissions were measured in plants for drying of Al chips (0.10-3.8 ng I-TEQ/m³) as well as in rotary kilns (0.014-11.6 ng I-TEQ/m³). From the differences observed, it was assumed that the chlorine content of the input materials might play a role for the PCDD/PCDF concentrations. The generation of both primary as well as of secondary copper involves several steps. Very low concentrations were measured for stone converters. The PCDD/PCDF emissions from anode ovens used for the generation of secondary copper were two orders of magnitude higher.

Secondary zinc can be recycled from fly ashes of steel works, residues, and sludges. Remelting plants for zinc gave relatively low concentrations around 0.1 ng I-TEQ/m³; hot briquetting plants gave mean concentrations of 10.8 and 38.3 ng I-TEQ/m³, respectively.

Table 22: Germany: PCDD/PCDF emissions from non-ferrous metal and aluminum production; reference year 1994 (Ifeu 1998)

Source Category	Emission Factor µg I-TEQ/t	Activity Rate t/a	Flux g I-TEQ/a
Non-ferrous metal and aluminum production			**49.7**
Secondary aluminum production			22.9
Chips drying	9.76	251701	2.46
Rotary ovens	50.5	402721	20.3
Hearth ovens	1.51	100680	0.16
Prim. and sec. Copper production			26.8
Fluidized melting ovens (prim.)	0.0050	253000	0.0013
Stone converters (prim.)	0.0080	253000	0.0020
Anode ovens (prim. +sec.)	3.57	592000	2.11

Blast furnaces (sec.)	27.1	339000	9.19
Scrap metal converter (sec.)	21.9	339000	7.43
Melting plants and foundries (prim. + sec.)	4.81	1683290	8.10

Table 23: Germany: PCDD/PCDF emissions from secondary zinc production; reference year 1994 (Ifeu 1998)

Source Category	Emission Factor µg I-TEQ/t	Activity Rate t zinc/a	Flux g I-TEQ/a
Secondary zinc production			**41.8**
Rolling mill	175	60000	10.5
Hot briquetting	521	60000	31.3
Zinc remelting	0.29	60000	0.017

The total emissions from the production of iron, steel, and non-ferrous metals produced PCDD/PCDF emissions of almost 280 g I-TEQ in the year 1994 (see Table 24).

Table 24: Germany: Summary of PCDD/PCDF emissions from ferrous and non-ferrous metal industries; reference year 1994

Source Category	Annual Emission g I-TEQ/a
Iron and steel	181
Aluminum production	22.9
Copper production	26.8
Secondary zinc production	41.8
Total	**272.5**

The category of combustion plants includes power plants, industrial combustion plants fired with lignite, brown coal, wood or crude oil. Measured data are available for power plants fueled with various kinds of coal: no data are available for plants fueled with crude oil. For the inventory, the emission factors from the respective power plants were applied. For all plants, the measured stack emissions were far below 0.1 ng I-TEQ/m³. Under industrial combustion plants, plants between 1 MW and 50 MW are summarized. Generally, they produce steam or process heat *via* hot water. There are a few data available for such units fired with lignite and brown coal. PCDD/PCDF emissions from wood combustion plants were studied intensively. Whereas combustion of untreated wood generates low PCDD/PCDF emission independently of the type of combustor used, combustion of waste wood with halogen-organic coatings may have a potential of generating high PCDD/PCDF emissions. Wood treated with wood preservatives have to be combusted in incinerators complying with the 0.1 ng I-TEQ/m³ limit value of the German regulation for any type of waste incineration plants (17th BImSchV 1990).

Small combustion units comprise a very heterogeneous group of units in terms of type of ovens, firing techniques, and fuels. All units have to comply with the 1st Amendment to the German Immission Law (1st BImSchV) and are smaller than <1 MW. The contribution of these sources to the PCDD/PCDF inventory has been under discussion for many years. Ifeu (1998) estimates the total emissions from these sources to 7.13 g I-TEQ for the reference year 1994.

Co-combustion of PVC-containing materials and waste wood in such small units will dramatically increase the annual flux. Launhard (1996) estimates that the additional contribution is in the range of 22 g I-TEQ annually for co-combustion of waste wood (46 PJ) and 42 g I-TEQ/a for co-combustion of PVC-containing materials (7 PJ).

Table 25: Germany: PCDD/PCDF emissions from power plants and other industrial combustion plants; reference year 1994 (Ifeu 1998)

Source Category	Emission Factor mg TEQ/TJ	Activity Rate TJ/a	Flux g I-TEQ/a
Power plants			**5.34**
Lignite	0.004	1249537	5.00
Brown coal	0.0002	1517285	0.30
Crude oil (heavy)	0.0004	70115	0.03
Crude oil (light)	0.0005	25746	0.01
Industrial combustion plants			**6.15**
Lignite	0.005	365516	1.83
Lignite coke	0.005	200174	1.00
Brown coal	0.004	199770	0.80
Brown coal coke	0.012	1977	0.02
Brown coal briquette	0.012	16655	0.20
Wood	0.27	7914	2.14
Crude oil (heavy)	0.0004	208875	0.08
Crude oil (light)	0.0005	166420	0.08

In Germany, municipal solid waste is incinerated without special pre-treatment and burnt on grates. Hazardous waste incinerators are rotary kilns operated between 800 and 1,200 °C: the off-gases pass an afterburner with temperatures between 1,200 and 1,400 °C. Common types of sewage sludge incinerators are fluidized bed or multi-hearth furnaces. Conventional burning of hospital waste was performed in small units and in batches. As most of the old plants could not comply with the requirements of the 17th BImSchV, they were closed in the early 1990s.

With the implementation of the 17th BImSchV in 1990 setting a legal limit concentration of 0.1 ng I-TEQ/m³ for waste incineration in Germany, a downward trend for PCDD/PCDF emissions was observed. Today, all MSWI and HWI comply with this requirement. Whereas in a national survey in 1989, the average stack concentration of German MSWIs was determined to 8 ng I-TEQ/m³, the concentrations were between 0.5 and 5 ng I-TEQ/m³ for the reference year 1994 (Ifeu 1998). Of the 40 MSWI plants operating in 1994, twelve did already comply with the 0.1 ng I-TEQ/m³ limit value. Also, for the inventory, an average concentration of 1 ng I-TEQ/m³ was calculated resulting in an annual emission of 30 g I-TEQ/a. For the inventory, a mean concentration of 0.5 ng I-TEQ/m³ was used for hazardous waste incinerators. In the reference year, 30 plants were in operation. The PCDD/PCDF emissions from sewage sludge incinerators were lower than emissions from MSWI; an average of 0.08 ng I-TEQ/m³ was calculated based on measured data.

Measured PCDD/PCDF emissions from crematories were in the range from 0.05 to 14.4 ng I-TEQ/m³. For the reference year 1994, there were 107 crematories operating approximately 2,000 hours per year. The total emissions from this source were calculated to be 2.38 g I-TEQ/a. Since 1997, there is a legal limit concentration for crematories of 0.1 ng I-TEQ/m³ (27th BImSchV 1997).

Table 26: Germany: PCDD/PCDF emissions from waste incineration plants; reference year 1994 (Ifeu 1998)

Source Category	Emission Concentrations ng I-TEQ/m³	Activity Rate	Flux g I-TEQ/a
Waste incineration			**32**
Municipal solid waste incineration	1		30
Hazardous waste incineration	0.1-0.5		2
Sewage sludge incineration	0.1-1		0.1
Hospital waste incineration	<0.1		<0.1
Crematories	0.05-14.4	2000 h/a	**2.38**

Based on a Directive from the European Union (91/441/EWG), all cars run on gasoline with a permit after 31 December 1992 have to be equipped with a catalytic converter. Although PCDD/PCDF are not addressed in the Directive, it can be assumed that a catalytic converter will also reduce emissions of PCDD/PCDF. The 19th BImSchV, which is in force since 1992 and which forbids the use of halogenated scavengers as additives to gasoline further reduced PCDD/PCDF emissions from gasoline fuelled cars. For the German inventory, emission factors as shown in Table 27 were applied. The highest uncertainty is for the emissions of trucks and busses, as there were much higher emissions published in the literature. However, Ifeu (1998) found these data not appropriate for use in the present inventory. The total annual emissions from automobile transport were less than 5 g I-TEQ/a.

Table 27: Germany: PCDD/PCDF emissions from road transport; reference year 1994 (Ifeu 1998)

Source Category	Emission Factors ng I-TEQ/L	Activity Rate t/a	Flux g I-TEQ/a
Road transport			**4.804**
Passenger cars, catalyst	0.07	18361383	1.714
Passenger cars, no catalyst	0.095	9780766	1.226
Passenger cars, Diesel	0.04	5775074	0.275
Busses, Diesel	0.076	1120772	0.101
Trucks <3.5 t, Diesel	0.076	2386434	0.216
Trucks, small, gasoline	0.094	1511932	0.190
Trucks, Diesel	0.076	11957804	1.082

Production of mineral materials such as cement, lime, glass, and brick occurs at very high temperatures. Measured PCDD/PCDF emissions were far below 0.1 ng I-TEQ/m³. In addition, the estimate for production of ethylene dichloride and the vinyl chloride monomer is included. As can be seen, the contribution to the national inventory from these processes is less than 100 mg I-TEQ/a. Finally, other high temperature processes where PCDD/PCDF emission measurements were performed are listed in Table 28 as well. Overall, the contribution of these sources to the total PCDD/PCDF emission inventory to air is very small (<<1 g I-TEQ/a).

Single measurement data exist for smokehouses (0.02 ng I-TEQ/m³), pyrolysis plants (0.055-2.6 ng I-TEQ/m³), sewage drying plants (0.05-0.11 ng I-TEQ/m³) and for manufacture of lacquer wires (0.0035 ng I-TEQ/m³). As there are no activity rates available for these industrial sectors and the emissions were very low, it was found that the contribution of these processes to the total inventory is negligible.

Table 28: Germany: PCDD/PCDF emissions from production of mineral materials and other production processes; reference year 1994 (Ifeu 1998)

Source Category	Emission Factors ng I-TEQ/t	Activity Rate Mio t/a	Flux g I-TEQ/a
Cement production	44.8	4.14	0.19
Lime production	78.1	8.51	0.66
Glass production	32.2	4.44	0.14
Brick production	86.6	16.7	1.45
Drying of wood chips	739		0.09
Drying of green fodder	100		0.04
Asphalt mixing plant	2.4		
Car shredders	246		0.37
	Emission Concentration ng I-TEQ/m³		Flux mg I-TEQ/a
Production of vinyl chloride			**76.5**
Oxychlorination	0.044		2.6
HCl-recycling from EDC manufacture	0.77		18.5
Combustion of VC plant	0.4		55.4

Further potential PCDD/PCDF sources with emissions can be briefly summarized as follows:

- Pulp and paper production is of minor importance in Germany. The total emissions were estimated to be less than 1 g I-TEQ/a with the major pathway via wastewater.

- Areas, especially sport fields, covered with a copper slag named “Kieselrot” are a special case in Germany. Re-entrainment of PCDD/PCDF from these highly contaminated areas via erosion and evaporation might occur. Lahl *et al.* (1995) estimated about 100 g I-TEQ annual for this source. However, there is no strong scientific basis for such estimate.

- High uncertainty is associated with the emissions from accidental fires, especially when PVC is involved. Lahl and co-authors estimated 1-10 g I-TEQ annually from this source, assuming 500,000 accidental fires occurring annually.

- PCDD/PCDF were found in the sludges from dry-cleaning units. As the residues from these units are treated as special waste, there is no release into the environment.

In conclusion, the inventory for the reference year 1994 gave PCDD/PCDF emissions of 333 g I-TEQ. The largest contributor was the iron and steel plants and foundries that accounted for 54 % of the total inventory. Within this source category, the sinter plants were the major contributor. The second largest source with 15 % were the non-ferrous metal industries with production plants for aluminum and copper. The third major sector was the secondary zinc production with 12.5 %. In 1994, municipal solid waste incineration was a minor contributor with annual emissions of 30 g I-TEQ/a accounting to 9.6 % of the total inventory.

Potentials for Reductions

Although PCDD/PCDF minimization measures were already implemented in Germany in 1994, potentials for further reducing PCD/PCDF emissions was identified. Within these, the greatest reduction potential was identified for sintering plants. Tests at sinter plants have shown that baghouse filters as used in many other industries cannot be applied at sinter plants. However, addition of adsorbent materials has proven to efficiently reduce the PCDD/PCDF emissions from the sinter bands. With such technology a limit concentration of 0.1 ng I-TEQ/m³ can be met. Once established, the annual emissions will be reduced by 98 % to 9.3 g I-TEQ/a (Ifeu 1998).

Presently, there are no experiences with minimization methods tested at zinc plants. However, results obtained from other industries indicate that the same measures could be applied in this sector as well, leading to an annual emission of only 0.2 g I-TEQ.

The addition of an adsorbens after the combustion unit will reduce the PCDD/PCDF emissions in many industrial sectors, *e.g.* cupolas, iron foundries, secondary aluminum production

For secondary copper plants, injection of oxygen resulted in PCDD/PCDF emissions <0.1 ng I-TEQ/m³.

For home heating units (small combustion plants for solid fuels), care should be taken that only untreated wood is used.

Overall, it can be concluded that the German metal sector has undertaken many efforts to reduce the PCDD/PCDF from its plants. However, presently, the effectiveness of these efforts cannot be quantified.

From the year 1994 it can be concluded that there is no further need for actions to reduce PCDD/PCDF emissions for oxygen steel plants and electro steel plants. There is hardly any reduction potential for power plants operated with fossil fuels. The annual emissions of about 5 g I-TEQ are due to the large volumes emitted.

For waste incineration primary measures and secondary technologies such addition of coke or active carbon, non-catalytic reduction, and catalytic converters were successfully applied to meet the legal limit concentration. Since 1 December 1996, all waste incinerators in Germany comply with this value. Results from more than 150 test data from cement kilns have shown that co-combustion of other materials including waste oil and used tires do not increase the PCDD/PCDF emissions. All measured concentrations were below 0.1 ng I-TEQ/m³.

As there is no longer leaded fuel available in Germany, it is guaranteed that the emissions from the use of leaded gasoline have been eliminated.

8.6.2 Hamburg

For the year 1992, annual fluxes and emission inventories of PCDD/PCDF were estimated for combustion sources for the city of Hamburg, Germany (Lau *et al.* 1996). Measures taken during the last years in Hamburg have already markedly reduced emissions of PCDD/PCDF into the environment. To further minimize dioxin exposure, national and international measures have to be taken.

With a population of 1.65 million inhabitants Hamburg is the second largest city in Germany; in addition, Hamburg is a federal state, too. Its area is 755 km². Hamburg has a highly urban industrial infrastructure with 2,200 inhabitants/km² (for comparison: German average = 228 inhabitants/km²). Hamburg has a long dioxin history including a former 2,4,5-T production site and the landfill “Georgswerder“ and others are contaminated with PCDD/PCDF. In addition, sediments from the harbor and the river Elbe are contaminated with dioxins and furans. Hamburg has several combustion sources (two municipal waste incinerators, one hazardous waste incinerator, steel works, and non-ferrous metal industry).

Since 1983, more than 800 congener-specific dioxin analyses were performed in Hamburg to determine levels of PCDD/PCDF in water, soil, and air. The number and quality of dioxin results are unique for any defined region in Germany. Almost complete statistical data were obtained for the year 1992; whenever possible, newer data were incorporated into the dioxin balance for Hamburg.

For Hamburg, PCDD/PCDF emissions to the air are predominantly from industrial and commercial activities (production/manufacture and finishing processes), domestic activities (home heating), waste incineration, and traffic (automobiles). According to the evaluation, in 1992 a total of 6.5 g I-TEQ were emitted from identified and quantified sources in Hamburg. Table 29 gives an overview on the combustion sources and the annual amount of PCDD/PCDF released from these sources to the atmosphere.

In the year 1992, incineration of municipal solid and hazardous waste was the major sector (industry) to emit PCDD/PCDF into the air: an annual flux of 4.5 g I-TEQ was attributed to this category (see Table 29). Retrofitting of the combustion facilities, shut-down of one old incinerator and installation of a new incinerator resulted in a reduction of more than 95 % for the year 1995 (total emissions from waste incineration and less than 0.2 g I-TEQ/a). Emissions from thermal industrial processes, especially from a copper plant and to a minor extent from iron and steel production, generation of aluminum, and smokehouses were also of importance. The contribution from generation of energy - fossil fuel power plants and combustion of sewer gas - was low when compared to the total amount of PCDD/PCDF emitted. Relatively large uncertainties exist for the estimate of PCDD/PCDF emissions from home heating. The best estimate for 1992 was between 70 and 340 mg I-TEQ/a.

PCDD/PCDF emissions from automobiles were due to the presence of halogenated scavengers in leaded fuel. The decrease in the use of leaded gasoline and increasing numbers of cars equipped with catalysts can be expected to result in a further reduction of dioxin emissions from traffic in Hamburg, too. Based on the evaluation by Lau *et al.* (1996) there is still a potential to further reduce dioxin emissions within the industrial sources. These reductions can only be realized by cost-extensive technological changes, *e.g.* smaller stack volumes.

A high degree of uncertainty still exists for the estimate of the contribution from private heating. Although there exists an abundance of PCDD/PCDF emission data for incineration of wood and fewer for coal and oil, the number of installations and the frequency of using the heating systems remain unknown. Uncertainty is also with the type of fuel burnt and whether the combustion of fossil fuels in private ovens is just for heating during winter or also for generation of hot water and other energy. Due to such lack of information, the authors came to the conclusion that additional dioxin measurements even of a large number of installations will not give further insight to more precisely calculate the annual load of PCDD/PCDF released from this source category.

Table 29: PCDD/PCDF emissions to the air from combustion sources in Hamburg

Source	Flux (mg I-TEQ/a)
Stationary Sources	
Industry:	
Copper industry	1,200
Aluminum industry	8
Iron and steel manufacture	166
Petrochemical industry	0.009
Smoke houses (smoked fish and meat)	8
Others:	
Crematories	4-21
Generation of Power:	
Power plants (fossil fuels)	69
Industrial wood combustion	150
Combustion of sewer gas	14
Waste disposal in Hamburg	
Municipal waste incineration	3,913
Hazardous waste incineration	560
Subtotal Stationary Sources	6,092-6,109
Diffuse Sources	
Traffic	
Automobiles	250
Households:	
Home heating	70-340
Accidental fires in homes	8
Subtotal Diffuse Sources	328-598
Total of All Known Sources	**6,420-6,707**

8.7 Hungary

In 1997, the Hungarian Institute of Environmental Protection (KGI 1997) published a report on emissions of persistent organic pollutants in preparation of international agreements. In this report, an inventory for PCDD/PCDF from sources in Hungary was included. Based on emission factors as established in the report by TNO (UBA 1997), PCDD/PCDF emissions were determined for various reference years. A comparison for the reference years 1985, 1990, 1993, 1995, and 1996 is shown in Table 30. In 1985, the highest dioxin emission was estimated to be 215 g I-TEQ/a (in 1980 - not shown here - the total emission was calculated to be 185 g I-TEQ). It can be seen that for all estimates, the dioxin emissions from municipal solid waste incineration – one single plant - did not change at all.

Table 30: PCDD/PCDF emission inventory for Hungary (KGI 1997) (g I-TEQ/a)

	1985	1990	1993	1995	1996
Households	93.98	66.17	34.79	27.19	25.54
Services	11.02	4.83	4.91	4.15	3.48
Power plants	15.13	12.21	12.98	13.07	13.69
District heating	2.59	1.63	2.98	1.48	0.021
Agriculture	4.78	2.76	2.71	3.29	3.01
Industry	8.51	5.27	3.61	3.40	3.46
Road transport	0.98	0.88	0.52	0.27	0.20
Incineration	46.50	46.50	46.50	46.50	46.50
Sintering process	15.38	12.50	7.40	8.20	4.00
Secondary copper processes	n.a.	1.08	0.62	0.70	0.76
Electric arc furnaces	15.79	12.53	3.43	3.96	2.87
Total	**214.65**	**167.36**	**119.57**	**112.21**	**103.53**

8.8 Slovak Republic

In 1994, the Slovak Republic undertook a first study to establish an emission inventory of POPs. The estimate is based on available information on emission sources, source parameters and emission factors obtained from either own measurements or from published literature sources. The reference year is 1993 (Kocan 1994).

The total emissions of PCDD/PCDF into the atmosphere were estimated to be 42 g I-TEQ/a for the reference year 1993. In 1993, incineration of waste was responsible for 61 % of the total emissions, the various sectors of the metal industry accounted for 20 % and energy production for 17 %. An overview is shown in Table 31. The Slovak Republic is aware of potential further sources, *e.g.* emissions from the chemical industry (including oil exploitation, transport and storage, petrochemical industry, production of organohalogen compounds and paints), but could not be quantified at this time. Emissions from landfills and waste dumps, re-entrainment of PCDD/PCDF from contaminated soils and from water surfaces, were identified but could not be quantified. In addition, the numbers as shown in Table 31 should be regarded as lower estimates; actual values are expected to be higher as for some sources there are high uncertainties in emission rates.

The uncertainties in the emission estimates result from the uncertainty in the emission factor, the activity rate (the amount of fuel processed) and the efficiency of the pollution control device (flue gas cleaning system).

Table 31: Slovak Republic: PCDD/PCDF emissions into air; reference year 1993

Source	Flux g I-TEQ/a	% of Total Inventory	Confidence Rating
Waste incineration	**26.0**	**61.2**	
Municipal waste	11		
Hospital waste	6		ND
Industrial waste	9		ND
Metallurgical industry	**8.6**	**20.2**	
Iron, steel, cast iron, ferroalloys	7.4		
Light non-ferrous metals (Al)	0.4		
Other non-ferrous metals (Cu)	0.77		
Fuel/Energy sector	**7.3**	**17.2**	
Combustion facilities >5 MW	4.8		
Combustion facilities 0.2-5 MW	0.4		
Combustion facilities <0.2 MW	1.6		
Coke production	0.43		
Energy gases production and combustion	0.024		
Mineral products industry	**0.05**	**0.1**	ND
Cement	0.023		ND
Lime	0.014		ND
Asphalt mixing installations	0.0006		ND
Glass, stoneware, mineral fibers	0.003		
Refractory materials	0.003		
Brick manufacture	0.0006		
Other high temperature processes	**0.54**	**1.3**	ND
Traffic	0.35		
Cremation	0.025		
Smoking	0.0009		
Fires	0.16		ND
Total	**42.49**	**100.0**	

ND = Values are less reliable as no measurements (no data) are available and estimates were derived from hardly comparable activities

8.9 Sweden

Within the Swedish Dioxin Study, emission estimates were made for the years 1990 and 1993 (Table 32) (de Wit 1995). Whereas no change was postulated for the amounts of PCDD/PCDF emitted to water and found in products and wastes, a decrease was calculated for the total emissions to air: the air emissions of 31.8-115 g N-TEQ from 1990 were assumed to decrease to 21.6-88 g N-TEQ in the year 1993. The categories with the largest reductions were primary lime production, municipal waste incineration, and wood burning stoves. However, small increases due to larger amounts of material or fuel processed were found, too (natural and biogas burning as well as use of Diesel and unleaded gasoline; cremation). Decreases are indicated Table 32 by bold numbers and increases by an asterisk.

Sweden is aware of additional sources, such as textile production, dry cleaners, and forest fires; however, no estimate was made. Sweden and the USA (results of the 1994 draft Dioxin Reassessment, not shown here) are the only countries that try to include dioxin emission streams to air, water, and sinks such as products and wastes.

Table 32: Sweden: Preliminary estimates of PCDD/PCDF emissions from various sources in Sweden during 1990 and 1993 given in g Nordic-TEQ (N-TEQ)/a (de Wit 1995).

Source	To Air 1990	To Air 1993	To Water	In Wastes	In Products
Industry					
Bleached kraft pulp	1	1	1.5-5		0.3-6.6
Recycling paper pulp					2.8
Chloralkali plants			0.28-0.6	0.25	
Mercury distillation	0.000005	0.000005	0.003	0.004	
Sewage sludge			0.11	2.9	1.5
Steel mills	1.6-4	1.6-4		8.2	
Scrap steel mills	0.4-15	0.4-15		20	
Prim. non-ferrous smelters	0.13-0.27	0.13-0.27		0.38	
Sec. non-ferrous smelters	4.3	4.3		2.1	
Foundries	0.007-0.49	0.007-0.49		0.82-15	
Cement kiln - gases	0.07-1.3	0.07-1.3			
Cement kilns - dust	0.35	0.28			
Lime burning, prim. prod.	9.8-18	2-2.5			
Lime burning - industrial use	0.5-6.3	0.5-6.3			
Ceramic salt glazing	0.01	0.01			
Incineration					
Glassfiber waste incineration	0.00004	0.00006 *			
Municipal waste incineration	4.5	3.0		0.6-2.4	
Hazardous waste incineration	0.01	0.007			
Hospital waste incineration	0.001	0.001			
Cremation	0.34-0.68	0.37-0.73*			
Uncontrolled landfill fires	2.8-30	2.8-30			
Illegal metal reclamation	?	?		0.02-0.04	
Heating/Energy					
Oil	0.1-2.6	0.1-2.5			
Coal	0.75	0.61			
Natural gas	0.03	0.04 *			
Wood burning stoves	3-13.8	2.1-9.8			
Other biofuel	1.2-6.4	1.4-7.7 *			
Traffic					
Ferries (gas oil, heavy oil)	0.7	0.63			
Planes (aviation fuel)	0.03-0.93	0.02-0.86			
Vehicles - Diesel	0.067-0.14	0.73-0.15*			
Gasoline - leaded	0.026-2.9	0.009-0.97			
Gasoline - unleaded	0.08-0.16	0.14-0.27*			
Total	**31.8-115**	**21.6-88**	**1.6-5.1**	**35-52**	**4.6-10.9**

8.10 Switzerland

The Swiss Agency for Environment, Forest, and Nature estimates that before 1955, annual emissions of PCDD/PCDF were less than 40 g I-TEQ. Between 1955 and 1980 dioxin emissions increased about 12-fold and peaked in 1980 with approximately 480 g I-TEQ/a. Since 1980 a steady decline was observed resulting in an estimated emission of approximately 180 g I-TEQ/a for the year 1995. Due to legal requirements, it is assumed that by the year 2000 only 70 g I-TEQ will be emitted annually from identified sources (BUWAL 1997).

Whereas in the past, waste incineration was the major contributor to the overall dioxin emissions (see Table 33), illegal combustion of wastes in private households will be the dominating source in the future. Although forbidden by law since 1986, such behavior is common practice still today. In the year 2000, dioxin emissions from illegal waste combustion will be about 3-fold higher than legal waste incineration from all municipal solid waste incinerators.

Table 33: Switzerland: PCDD/PCDF emissions into air (BUWAL 1997)

Source	1990	1995	2000
Households	*27.4*	*27.4*	*29.8*
Waste incineration	26.8	26.9	29.4
Firing systems	0.5	0.5	0.4
Waste Incineration	*112.8*	*112.8*	*16.2*
Municipal solid waste incineration	96.2	96.2	9.9
Hazardous waste incineration	7.3	7.3	4.9
Sewage sludge incineration	0	0	0
Burning on landfills	9.1	9.1	1.2
Cable burning	0	0	0
Shredders	0.2	0.2	0.2
Construction	*22.9*	*15.4*	*4.0*
Combustion of debris	19.9	12.7	1.6
Use of wood preservatives	3.0	2.7	2.4
Metal Industry	*20.7*	*11.2*	*11.6*
Steel works	12.2	8.0	8.4
Non-ferrous metal foundries	6.4	1.8	1.9
Foundries (iron)	1.3	1.2	1.3
Galvanization	0.1	0.1	0.1
Aluminum remelting	0.7	0	
Miscellaneous Industries	*16.7*	*13.1*	*9.7*
Combustion in small industries and commerce	1.0	1.1	1.1
Hospital waste incinerators	13.8	10.4	6.9
Cement kilns	0.8	0.7	0.7
Smoke plants for meat	0.4	0.3	0.3
Crematoria	0.4	0.4	0.5
Chip-board production	0.2	0.2	0.2
Others	0.1	0.1	0.1
Traffic	*1.9*	*0.9*	*0.3*
Vehicles without catalyst	1.8	0.7	0.2
Agriculture and Forestry	*0.1*	*0.1*	*0.1*
Total	**242**	**180**	**72**

8.11 The Netherlands

Annual PCDD/PCDF emissions were estimated for the Netherlands by de Koning *et al.* (1994). As shown in Table 34, in the year 1989, the total flux to the air was much higher (960 g I-TEQ) than in 1991. The decrease is due to the fact that some municipal waste incinerators with high emissions (up to 92 ng I-TEQ/m^3) were either shut-down or retrofitted.

In 1991, municipal waste incineration was still the largest contributor with 79 %. The residual 21 % were split on 16 process categories. There is high uncertainty with the emission estimates from cable processing, wood combustion, and the former use of wood preservatives.

Interestingly, the estimate for the largest source in 1993, namely use of pentachlorophenol, is based on no data. No data were available for accidental fires; thus, this category was not quantified.

Table 34: Estimated annual PCDD/PCDF emissions into the air for the Netherlands (de Koning *et al.* 1994)

Source	Emission for 1991	Emission for 2000 [1)]
Municipal waste incineration	382	2-4
Hazardous waste incineration	16	1.7
Landfill, biogas, and sludge incineration	0.3	1.5
Cable and electromotor burning	1.5	1.5
Hospital waste incineration	2.1	0
Asphalt mixing plants [2)]	0.3	0.3
Combustion of oil	1.0	1.0
Combustion of coal	3.7	3.7
Combustion of wood [3)]	12	9
Crematories	0.2	0.2
Traffic	7.0	0.2-5
Sintering plants	26	3
NE-Non-ferrous metal industry	4.0	4.0
Chemical production processes	0.5	0.5
Use of wood preservatives	25	20
Other high temperature processes [4)]	2.7	2.7
Accidental fires	?	?
Total (without fires)	**484**	**max. 58**

1) The emission estimate for the year 2000, with the exemption of MWI, does not account for growth or decrease in the total mass processed
2) Emission as a result of heat generation is mentioned under the energy generators concerned
3) Highly uncertain; further research recommended
4) Thermal soil treatment, drying of fly ash, cement and glass/mineral wool production, *etc*.

8.12 United Kingdom

8.12.1 Emissions to Air

An inventory of PCDD/PCDF releases into the atmosphere of the United Kingdom was performed in 1995 (HMIP 1995, Eduljee and Dyke 1996). The numbers given in the second column of Table 35 for the 1993 estimate by Eduljee and Dyke were derived from emission factors (µg I-TEQ/ton of material produced or treated) and the amount of material treated or produced by each of the processes. In the case of crematoria and traffic, the emission factors were expressed in terms of the quantity of PCDD/PCDF emitted per cremation and per kilometer traveled, respectively. Measured data were used for municipal waste incineration, clinical and chemical waste incineration, coal combustion, and sinter plants which covered the main emissions. For the other cases, emission factors were obtained from the literature. A confidence rating was included. As industrial practices in the UK are currently undergoing significant changes, the 1996 estimates as shown in Table 35 (2nd column) could not be taken as definitive:

rather they should be regarded as indicative of the situation in the UK at that time using the best data then available.

As can be seen from Table 35 and the reference year 1993, industrial processes account for approximately 90 % of the total inventory which ranges from 560 to 1,100 g I-TEQ per year. Municipal waste incineration (MWI) was identified as the major source of PCDD/PCDF emissions to the atmosphere. However, as these emissions were subject to stringent control from 1995/96, the contribution of this source will reduce significantly. The non-industrial sources contributed emissions of 24-145 g I-TEQ/a, approximately 4-15 % of the inventory. By December 1996, all MSWI in the UK must either be upgraded to meet the limit of 1 ng TEQ/m³ [‡] as given in the Environmental Protection Act or cease operation. Thus, due to legal enforcement, the emissions of old plants are expected to be reduced by a factor of 10 or 20 by 1996 compared to the 1993 estimate. In addition, emissions to the atmosphere from combustion sources such as thermal waste disposal and thermal metallurgical processes, will undergo significant changes over the next few years as a result of the implementation of Integrated Pollution Control.

Since the first UK national emission estimates (Eduljee and Dyke 1996), a large amount of data from monitoring programs initiated by the UK Environment Agency was collected. New information was obtained for sectors such as MSW incinerators, sinter plants, cement kilns, chemical waste incinerators, and metal processing. The reference period is from 1995 to early 1997. Very often the monitoring was driven by the need to demonstrate compliance with emission standards. Although the majority of sources were sampled only once, except for MSWI, the new data did allow for a more precise estimate of total PCDD/PCDF emissions from UK sources. These new estimates are shown in the 3rd and the 4th columns of Table 35 for the lower and the upper estimates, respectively (Alcock *et al.* 1998).

Recent monitoring programs only included normal operation of plants. All analyses were carried out with high-resolution gas chromatography and high-resolution mass spectrometry (HRGC/HRMS). Nevertheless, limitations occurred for isokinetic flue gas sampling which could not be achieved at all plants.

As for many other countries, municipal solid waste incinerators represented the sector with most measured data available. Up to 1998, 14 sites have been analyzed for PCDD/PCDF. MSWI also represented the sector with the broadest range of results with stack concentrations from 0.07 to 74.4 ng I-TEQ/m³. A correlation between stack concentrations and age of the incinerator, type of flue gas cleaning system was not observed. Despite the broad range of concentrations found, the patterns of 2,3,7,8-substituted congeners of individual MSWI were quite similar: Typically, the emissions were dominated by Cl_8DD and 1,2,3,4,6,7,8-Cl_7DD followed by 1,2,3,4,6,7,8-Cl_7DF and Cl_8DF. Emissions from cement kilns, sewage sludge incinerators, and sinter plants (0.43-1.08 ng I-TEQ/m³) showed the least variation in measured emissions (normalized to I-TEQ) within a given industry sector. The I-TEQs for the ten chemical waste incinerators ranged from <<0.1 to 1.27 ng I-TEQ/m³. The pattern from the sinter plants was distinct from the pattern of the MSWI: In the sinter plant emissions PCDF were dominating with 2,3,4,7,8-Cl_5DF as the most abundant congener followed by 1,2,3,4,6,7,8-Cl_7DF, 1,2,3,7,8,9-Cl_6DF, and 1,2,3,4,7,8-Cl_6DF.

For the establishment of the inventory, measured emission data for each industry were compiled and the PCDD/PCDF released per year calculated from known throughputs of input material and gas volumes generated. For some sectors such as accidental fires, data from the literature were applied. As can be seen from Table 35 right columns, municipal solid waste incineration was still a significant source in the 1997 estimate contributing between 30 % and 56 % to the total inventory. However, this represents a significant decline from the 80 % estimated for 1993 by Eduljee and Dyke (1996). Emissions from sinter plants contribute between 5 % and 11 % to the total

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[‡] The legislation includes a target of 0.1 ng I-TEQ/m³

inventory. Compared to the 1994 inventory, combustion of coal increased in significance the 1997 share being about 18 %. The total PCDD/PCDF emissions from known UK sources are in the range between 200 and 660 g I-TEQ/a and thus much lower than the 1994 estimate which gave 560-1,100 g I-TEQ/a.

Table 35: United Kingdom: Estimated emissions of PCDD/PCDF to the atmosphere for the reference years 1993 and 1997 (g I-TEQ/a) (HMIP 1995, Eduljee and Dyke 1996, Alcock *et al.* 1998)

Process	1993 Estimate *	1997 Range/Low	1997 Range/High
Coke production	2	2	2
Coal combustion (power generation)	5-67	5	67
Waste oil combustion	0.8-2.4	0.8	2.4
Wood combustion	1.4-1.9	1.4	1.9
Straw combustion	3.4-10	3.4	10
Tire combustion	1.7	1.7	1.7
Landfill gas combustion	1.6-5.5	1.6	5.5
Sinter plants	29-54	25	30
Iron and steel	3-41	3	41
Non-ferrous metals (1 site)	5-35	29.8	29.8
Cement manufacture (5 sites)	0.2-11	0.29	10.4
Lime manufacture	0.04-2.2	0.04	2.2
Glass manufacture	0.005-0.01	0.005	0.01
Ceramic production	0.02-0.06	0.02	0.06
Halogenated chemicals	0.02	0.02	0.02
Pesticides production	0.1-0.3	0.1	0.3
MSW incineration	460-580	122	199
Chemical waste (10 sites)	1.5-8.7	0.02	8.7
Clinical waste (5 sites)	18-88	0.99	18.3
Sewage sludge (5 sites)	0.7-6	0.001	0.37
Carbon regeneration	0.006	0.006	0.006
Asphalt mixing	1.6	1.6	1.6
PCP in timber processes	0.8	0.8	0.8
Animal carcass incineration **		0.001	0.18
Crematoria	1-35	1	35
Domestic wood combustion (clean)	2-18	2	18
Domestic wood combustion (treated)		1	5
Domestic coal combustion	20-34	20	34
Traffic	1-45	1	45
Natural fires (wood)	0.4-12	0.4	12
Fires (accidental)	10	10	81
Total	**559-1,099**	**219**	**663**

* from Eduljee and Dyke 1996;
** assuming 7,1160 carcasses processed mid-June 1997 (associated with over 30 months scheme)

8.12.2 Emissions to Water and Land

The UK Environment Agency required information on the scale of PCDD/PCDF releases to land and water. In 1997, the report was published by the Environment Agency (1997) based on data compiled between February and August 1996. In the study, industrial and non-industrial sources were assessed. The releases to land included materials sent to landfills; similarly the releases to water covered releases to sewer. Where possible, UK data were used; where there were no domestic data available, data from other countries were applied. Nevertheless, an urgent need for generation of more data was identified. The results were given as ranges of estimates. Due to the lack of data, releases to water were quantified according to the potential of a process to release PCDD/PCDF as H (high = greater than 1.0 g I-TEQ/a), M (medium = 0.1-1 g I-TEQ/a) or L (low = < 0.1 g I-TEQ/a).

Despite the scarcity of data, it was possible to make estimates to land from a large number of industrial and non-industrial processes. Even for processes where it was possible to estimate releases, great uncertainties were recognized. The uncertainty was either with a) the amounts of material arising and b) the high variability of PCDD/PCDF concentrations in the same kind of materials from one sample to another sample or from process to process.

Total releases to land were estimated to be between 1,500 and 12,000 g I-TEQ per year (Table 36). This number is significantly higher than the estimates of annual releases to air. The majority of releases to land were contained in residues and materials for disposal in landfills. Under good management practices, such contamination is thought not to be mobile and bioavailable. Nevertheless, field monitoring may be recommended for surveillance. The major releases to were thought to come from open use of chemicals, the incineration of municipal waste in old plants, accidental fires, chemical production processes, and the non-ferrous metal industry. Newer data on concentrations in chemicals especially are needed to better estimate the total releases.

As can be seen from Table 36 it was not possible to quantify the release of PCDD/PCDF to water. Only a few sources could be identified. In general, the numbers estimated were low but may be the result that many water treatment systems are capable to remove PCDD/PCDF from effluents.

Table 36: United Kingdom: PCDD/PCDF releases to land and water; reference year 1996 (Environment Agency 1997). All concentrations in g I-TEQ/a

Process	Release to Land	Potential Release to Water	Comments
Carbonization	0.023-0.85	L (0.0044)	
Coal combustion			
- Power stations	1.6-81	L	Half ash is sold, may not be released
- All industrial	0.015-4.3	L	
- Domestic: Ash	0.090-0.31	Negligible	
- Domestic: Soot	0.16-30	Negligible	Includes soot from oil and wood use
Oil combustion			
- All industrial	Not quantified but low	Negligible	Only soot discharge
- Domestic	See coal soot	Negligible	
Waste oil combustion	Not quantified	Negligible	No data
Wood combustion			
- All industrial	0.40-4.4	Negligible	
- Domestic	0.27-16	Negligible	See domestic coal soot
Straw combustion	0.20-10	Negligible	
Tire combustion	1.9-2.8	L	
Crematoria	Not quantified	Negligible	
Refuse-derived fuel combust.	3.7-6.4	L/M	
Poultry litter combustion	0.26	Negligible	
Petroleum processes	Not quantified	L	
Sinter plants	0.020-0.060	Negligible	
Electric arc furnaces	59	Negligible	Average PCDD/PCDF data used
Primary aluminum	0.082	L	
Secondary aluminum	29-320	L/M	
Secondary magnesium	0.38-3.2	L/M	Based on estimates for Al processes
Copper	24	L/M	
Secondary lead	95-220	L/M	
Cement	0.00040-12	L	
Lime	0.000060-1.8	L	
Other mineral processes	Not quantified	M (<0.45)	Only maximal data for asphalt mixing
Chlorine production	6.0	L	
PVC/EDC production	25-80	M (0.070-0.040)	Discharge to land goes to containment and into lagoon
Per-, trichloroethylene production	350-630	M (0.070-0.040)	Discharge to land goes to containment
Pesticides production	8.9-2000	H (0.089-2.0)	Very limited data
Chlorophenol production	Not quantified	H	Waste usually incinerated, releases to water reducing
Other chemical processes	-		Insufficient data for quantification; potentially insignificant

Table 36 (continued)

Process	Release to Land	Potential Release to Water	Comments
MSW incineration			
• Old plants	510-2400	M	Old plants closed or upgraded by Dec. 1996
• New plants	14-38	L	
Chemical waste incineration	0.0058-2.0	H (0.018-1.1)	Water discharge depends on treatment applied
Clinical waste incineration		M	Releases to land are expected to increase
• Under national authority	5.3-14		
• Under local authority	6.4-22		
• Other	0.050-0.90		
Sewage sludge incineration	0.98	L (0.0020)	
Paper and pulp processes	2.8-11	M	Little UK data
Textile treatment	Not quantified	M (0.032-0.93)	Based on PCP usage only
Manufacture of dyes	Not quantified	M	Insufficient information
Timber treatment	0.011-0.32	L (0.0028-0.083)	Based on PCP usage
Accidental fires	7.5-2400	H (0.075-24)	
Sewage sludge disposal	14-56	H (0.41-1.6)	
Waste oil disposal	Not quantified	H (0.28-1.2)	No modern concentration data
Bonfires, accidental fires	0.075-42	Negligible	
Vehicles	-	-	Covered in other sections
Disposal of MSW to landfill	150	M (0.23-0.59)	Mean contamination data; leachate may contain PCDD/PCDF
Compost from MSW	1.7	L	
Dredging	29	Not applicable	Average data
Open use of chemicals			
• PCP uses	100-3000	H	No Data on other chlorophenols
• Other pesticides	4.8-250	H	Little contemporary data
Surface runoff	Not quantified	M	Not quantified, liquid discharge may be significant
Dry cleaning	0.68-9.8	L (0.00090)	
PCB	0.31-0.38	M	No TEQ data
Total	**1500-12000**		

8.13 European Emission Inventory

Presently, the European Union (EU) comprises 15 Member States, namely Austria, Belgium, Denmark, France, Finland, Germany, Greece, Ireland, Luxembourg, The Netherlands, Portugal, Spain, Sweden, and the United Kingdom. In February 1993, within the "5th European Program of Policy and Action", the Council of Ministers laid down its political goal to reduce the release of PCDD/PCDF into the environment and into the human food-chain as a precautionary measure. The final goal to be achieved is to reduce the emissions of PCDD/PCDF by 90 % until the year 2005 as compared to the reference year 1985.

To identify dioxin emitting sources and to quantify total releases of PCDD/PCDF within the Member States, the General Directorate (DG) XI commissioned a research program to the State Environmental Agency North Rhine-Westphalia (LUA). The program started in 1994 and has two stages (LUA 1997):

(1) Stage I: Collection of data on dioxin releases for the reference period 1993-1995. Besides emissions to air, releases of PCDD/PCDF to water, land, and residual matter should be covered as well. Based on the information available, the most important data gaps should be identified; and

(2) Stage II, the gaps should be closed both for countries where only little information is available so far and for processes that need better characterization.

In 1997, LUA published the results of Stage I, a two-year research program on information available with regard to emissions of PCDD/PCDF. The report covers the 15 member states of the European Union and additionally includes Norway and Switzerland. The results of the 1997 evaluation based on national documentation are summarized in Table 37.

It should be noted, however, that national emission inventories performed shortly afterwards gave different estimates (*e.g.* emissions from MSWI in Germany, see section 8.5).

One of the major outcomes of the LUA study was that although the knowledge on PCDD/PCDF emissions in European countries has improved considerable, the national inventories are far away from being complete. No information was available from the member States Greece, Ireland, Luxembourg, and Portugal. Data gaps existed in particular for municipal solid waste incineration in Italy, Spain, and the large number of small facilities in France. Information on non-atmospheric emissions was reported only for very few countries and a small number of source types. In general, no emission factors were reported.

Within Stage II of the project, some of the above mentioned data gaps will be closed by the ongoing French National Dioxin Program and by a measurement campaign carried out in Spain. Special attention will be paid to sintering plants operated in Belgium, Spain, France, Italy and Portugal. A better database has to be established for clinical waste incineration, especially for the number and type of the facilities and waste statistics. Measurements are scheduled to be performed within the Stage II framework in Spain, Denmark, and Poland. It is hoped to obtain a more reliable range of emission factors for this process. Further sub-projects will deal with non-ferrous metal production in the United Kingdom. Finally, PCDD/PCDF emissions from wood combustion and Diesel-fueled vehicles will be evaluated.

The information in the LUA report is organized according to SNAP (Selected Nomenclature for Atmospheric Pollutants) as established by the CORINAIR (European Air Emission Data Base) inventory system.

8.13.1 Emissions to Air

Analyses of basic documents obtained from the 17 European countries gave total PCDD/PCDF emissions into the air from known sources of approximately 3,300 g I-TEQ/a (see Table 37). However, a re-evaluation of the data using emission factors for a given process were applied taking into account parameters such as abatement technologies. The results of the re-evaluation procedure yielded PCDD/PCDF emissions to the air from the most relevant sources of 5,800 g I-TEQ/a which are thought to represent 90 % of the total emissions. Thus, it was estimated that the annual air emissions of PCDD/PCDF from all known sources in the 17 countries are 6,500 g

I-TEQ/a. A comparison of the two estimates for air emissions on a country basis is shown in Table 38. It can be seen that large differences could be found. This was an expected finding as for some countries almost no or only limited informations was available. In addition, the sources of information varied from comprehensive reports based on large numbers of emissions measurements to short estimates based on literature data. A further complication is given by the fact that within the last years considerable technical improvements were implemented in many areas, so that the emission estimates undergo rapid changes. Nevertheless, the re-estimate still has to be considered as considerably uncertain due to the lack of many basic data or low quality of data

Based on the present European emission inventory as compiled by LUA (LUA 1997)

62 % of the PCDD/PCDF emissions are due to:
* Incinerators for municipal solid waste
* Iron ore sinter plants
* Incineration for clinical waste, and
* Facilities of non-ferrous metal industry.

38 % of the PCDD/PCDF emissions are dominated by non-industrial sources, such as:
* Domestic heating, *esp.* wood combustion
* Accidental fires, and
* Traffic (mainly if leaded gasoline is used).

Legend to Table 37

Code Category	Emission Source Groups Included
01	
02	Residential combustion in boilers, stoves, fireplaces (Process materials: wood, coal/lignite)
03	Combustion in industry/boilers, gas turbines, stationary engines, sinter plants, secondary zinc, copper, aluminum production, cement, lime, metal reclamation from cables
04	Electric furnace steel plants, non-ferrous metal foundries, sintering of special materials and drossing facilities; coke ovens, also chlorine production (EDC, PVC), pulp and paper
06	Preservation of wood, textile finishing
07	
08	
09	Incineration of domestic or municipal wastes, incineration of hazardous wastes, incineration of hospital wastes, cremation, sludge spreading
12	Fires of buildings, vehicles, and landfill fires (not originally included in CORINAIR subgroups)

A closer look at the data, *e.g.* for Germany, gives a total flux of 160.7 g I-TEQ/a for waste disposal treatment. Of these 157.2 g I-TEQ/a were attributed to municipal solid waste incineration for the reference period 1993-1995; for the year 1997, when all MSWI have to comply with the limit value of 0.1 ng I-TEQ/m^3 the LUA prediction is at 5.6 g I-TEQ/a (LUA 1997).

Table 37: National data: PCDD/PCDF Air Emissions in the European Community (g I-TEQ/a)

Source Sector Code#	Energy Prod. 01	Non-ind. Comb. 02	Manufact. Industry 03	Production Processes 04	Solvent Use 06	Road Transport 07	Mobile Sources 08	Waste Treat-ment/Disposal 09	Agriculture, Forestry 10	Nature 11	Fires 12	All
A	0.1	15.1	11.8					0.2		0.02		27
B	2.3	122.2	221.8	8.0	25.0	1.7		343.9	0.1		1.9	727
CH	0.8	0.7	2.7	10.6	2.7	0.8	0.1	154.0			9.1	182
D	3.6	14.5	215.0	121.5		3.8		160.7			81.0	600
DK	2.0	1.1	1.5	7.5		0.2		30.3				43
E	3.9	43.9	67.5	18.0				0.6				134
F			457.5					163.4				621
FIN	0.5	1.1	0.5	15.7		3.6		3.2				25
L			26.8	2.4				0.1				29
N			37.3	4.8				2.8				45
NL	3.0	11.3	33.7	3.3	25.1	2.1	0.6	8.8	0.3			89
S	0.6	5.1	14.3	3.6		0.6	0.8	3.5			7.9	36
UK	10.7	32.0	82.7	12.8	0.3	6.1		568.4		2.2		715
Sum	27.5	247.0	1173.1	208.2	53.1	19.0	1.5	1440.0	0.4	2.2	99.9	3273

Country codes are: A = Austria B = Belgium CH = Switzerland D = Germany DK = Denmark E = Spain F = France
FIN = Finland L = Luxembourg N = Norway NL = The Netherlands S = Sweden
UK = United Kingdom

Table 38: PCDD/PCDF air emissions in the European Community – Comparison of results from national reports and LUA re-estimate; reference years 1993-1995 (g I-TEQ/a) (Quaß *et al.* 1998)

Country	National Reports	LUA Re-Estimate
Austria	29	121
Belgium	727	484
Denmark	43	50
Finland	25	69
France	621	1119
Germany	600	840
Greece	No data	122
Ireland	No data	33
Italy	No data	1050
Luxembourg	29	50
The Netherlands	89	117
Norway	45	41
Portugal	No data	127
Spain	134	327
Sweden	36	89
Switzerland	182	183
United Kingdom	715	928
Total	**3273**	**5750**

An evaluation of the re-estimated emissions revealed that the European dioxin inventory is dominated by a few source categories. In the reference period 1993-1995, the largest contributor to the inventory was municipal waste incineration followed by sinter plants (see Table 39). Strong decreasing trends were identified for two source categories: Many countries are in the process to phase out leaded gasoline what will reduce dioxin emissions from this sector. The strongest decline in the present inventory can be expected for municipal waste incineration: Whereas for the reference period 1993-1995, the re-estimate was calculated to be 1,467 I-TEQ/a, only 20 g I-TEQ/a will be emitted if all MSWIs in Europe will comply with a limit value of 0.1 ng I-TEQ/m³ as proposed by the recent draft of the European Commission (EU 1998).

Table 39: PCDD/PCDF air emissions in the European Community – Most important sources for reference years 1993-1995, LUA re-estimate (g I-TEQ/a) (Quaß *et al.* 1998)
EF = Emission factor, AR = Activity rate

Source Type	PCDD/PCDF g I-TEQ/a)	Remarks	Uncertainty EF / AR
Municipal waste incineration	1467 + 174	Decreasing trend Illegal burning of domestic waste	Low / Low
Sinter plants	1010 +115	 Sinter plants for recycling materials	Medium / Low
Residential wood combustion	945	Use of contaminated wood uncertain	Medium / High
Clinical waste incineration	816	Few plant data and statistics	High / High
Wood preservation	381	From PCP-treated materials	v. High / v. High
Fires	380	Based on one reference only	v. High / v. High
Non-ferrous metals	136	Cu, Al., Zn	Medium / Low
Road transport	111	Mainly leaded fuel; decreasing trend	Low / Low

8.13.2 PCDD/PCDF Releases to Water and Residues

Apparently, much less is known about PCDD/F emissions through waste water and process residues/wastes. Some of the reviewed national documents contain emission estimates which amount to a total of 3,500 g I-TEQ per year for residues and about 20 g I-TEQ per year for waste water. Both values must be considered as being uncertain, since no data are available to date from a huge number of emission sources. Emissions *via* wastewaters seem to be negligible compared with atmospheric emissions and emissions *via* residual materials. Thus the total emissions into the atmosphere and through residues disposed off in landfills or other reservoirs are assessed to be of the same order of magnitude. Due to the lack of pertinent emission factors and activity rates a recalculation of the emissions *via* wastewater or residues for all countries was impossible.

8.13.3 Conclusions

As indicated by the large emission margins calculated to be about 1,300-20,000 g I-TEQ/a for air emissions and 500-9,000 g I-TEQ/a for emissions *via* residual materials the overall uncertainty of the inventoried emission estimates is quite high. Moreover, the quality of the information available from the respective national institutions differs widely particularly with respect to the number of sources considered and the extent of measurements on which estimates are based. Most data are available concerning air emissions; the data situation is much worse with respect to other pathways of PCDD/PCDF into the environment. In particular, measurements of PCDD/PCDF concentrations in residual materials are often not available; information about the amount and the fate of production wastes is scarce. Regarding the most relevant air emission sources data gaps still exist and thus, causing considerable uncertainties of the emission estimates:

Municipal waste incineration: The main data gaps exist for plants located in Spain and Italy where no measured data were available. In addition, there are no emission data from the large number of small incineration facilities (capacity <3 t/h) operated in France. Nevertheless, the emission estimate is assumed to have quite a high degree of certainty.

Iron ore sinter plants: PCDD/PCDF air emissions from about 10-15 sinter plants have been measured during the last years with almost similar results. However in a few cases, unexpectedly high flue gas concentrations (20-fold higher) were detected. Consequently, there is an urgent need to obtain more and reliable data from all remaining plants. It has been shown that the whole European emission inventory is highly sensitive to such outliers.

Clinical waste incineration: The major difficulty with the dioxin emission estimate is with the fact that there are no consistent statistical data available on the amount of clinical waste being incinerated. Apparently, only total generation data are available without specification. Moreover, no information was available about the number and technology of the facilities presently in operation. It is known that in some countries (*e.g.* Germany, the Netherlands) small in-house facilities were shut down in the early 1990s. The few existing emission measurements gave a wide range of emission factors. Consequently, the uncertainty of the emission estimate is very high for this sector. Additional work to improve the database was strongly recommended.

Non ferrous metal industry: Among the large number of production facilities in this sector, the plants to recover metals from secondary materials need special attention, especially plants in Belgium, France, Spain, Italy, and the United Kingdom. Present emission factors were derived from data gathered in Switzerland, Germany, and The Netherlands and may not be applicable for other countries. Since the dioxin emissions may vary considerably with the technology applied (*e.g.* like in the case of hot and cold cupola furnaces), the use of pure statistical production data without relation to the actual technology will result in additional errors into the emission estimate. Moreover, no information was found on the number of cable recovering facilities. As this emission source may have a significant local impact, better knowledge on the locations of such plants and measured data are needed. In countries like Germany, thermal cable recycling was stopped about ten years ago.

Residential Wood Combustion: This sector is still highly uncertain, especially concerning the amount and the composition of wood being fueled into the stoves. The extent of co-combustion of household wastes is almost unknown and should be assessed by empirical studies since this practice may influence considerably the PCDD/PCDF emissions from stoves and fireplaces.

Fires: It is well known that PCDD/PCDF may be formed during accidental fires of buildings, equipment, and vehicles; however, a prediction of the emission concentrations is very uncertain. The emission factors derived so far from a few experiments must be regarded as highly uncertain. Results from real fires have shown that very often, laboratory and pilot scale experiments overestimated the dioxin concentrations. Fire statistics generally do not contain data about the mass and composition of burnt materials; these data can only roughly be estimated. An improvement of the data situation is regarded to be almost impossible and would require large efforts; hence no recommendation was given by LUA to further improve the database.

Road traffic: According to the available data, PCDD/PCDF emissions from road traffic predominantly are caused by combustion of leaded fuels. Therefore they play a role only in countries with this fuel still in use. On the basis of emission factors derived from various investigations, the annual emission from this source category could be estimated with an acceptable precision. However, in the scientific literature, possible PCDD/PCDF emissions from heavy-duty Diesel engines are being discussed repeatedly; thus additional work seems to be necessary to get more reliable data.

8.14 European Atmospheric Emission Inventory

The TNO Institute of Environmental Sciences, Energy Research and Process Innovation, Apeldoorn (The Netherlands) published “The European Atmospheric Emission Inventory of Heavy Metals and Persistent Organic Pollutants for 1990“ on behalf of the German Environmental Agency (UBA) (UBA 1997). The report presents the results of an emission inventory project for heavy metals and persistent organic pollutants for 1990 on the basis of submissions of emission data from the Parties to OSPARCOM, HELCOM and the Convention on LRTAP. The inventory includes all countries in Europe, except the three Caucasian countries and Turkey (total of 38 countries), and is based on official country submissions and supplementary default emission estimates by TNO. The report’s substance list includes amongst others polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF). For the countries, sources or compounds missing in official submissions, default emission estimates have been prepared and applied to complete the inventory. The inventory practically covers the whole of Europe and the inventory results are available on a 50x50 km² EMEP grid for modeling purposes. The report is intended to be a preparatory step towards the preparation of the European CORINAIR95 emission inventory. Similarly to the LUA Dioxin Emission Inventory Report (LUA 1997), the sources are categorized according to the SNAP categories.

This report presents the results of the inventory in the form of tables, maps and figures for all compounds investigated. Here, only the results for PCDD/PCDF are summarized. The results are a mixture of official country submitted data and default emission estimates by TNO as not all countries have been in the position to offer complete information on source categories and substances. The default emission database was prepared for the base year 1990.

In general terms, the PCDD/PCDF emission per source category has been estimated for each country by multiplying the activity rate of the source (*e.g.* ton cement produced) with an emission factor (*e.g*. mg I-TEQ emitted per ton cement produced). A large number of handbooks and official statistics have been used to determine activity rate data for the project-specific SNAP sectors applied in this inventory.

The default emission factors have been applied uniformly over Europe for most categories due to lack of detailed and reliable data rather than deliberate choice. Differences in default emission factors between regions and countries in the validation database are based on differences both in techniques and in abatement.

In order to determine the total emissions for the 38 European countries, Europe was split in three groups: North-western (including Italy), Southern (excluding Italy), and Central and Eastern countries. Table 40 shows which countries belong to which group.

Table 40: Classification of countries into three groups to assign emission factors

Region	Countries
North-western Europe	Austria, Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Italy, Luxembourg, the Netherlands, Norway, Sweden, Switzerland and the United Kingdom
Southern Europe	Cyprus, Greece, Portugal and Spain
Central and Eastern Europe	Albania, Belarus, Bosnia and Herzegovina, Bulgaria, Croatia, the Czech Republic, Estonia, Hungary, Latvia, Lithuania, Former Yugoslav Republic of Macedonia, Moldova, Poland, Romania, the Russian Federation, the Slovak Republic, Slovenia, Ukraine and Yugoslavia (Serbia and Montenegro)

Default emission factors were determined for PCDD/PCDF depending on the sector and the region. As can be seen from Table 41, except for municipal solid waste incineration, no difference was made between regions. In other words: the same emission factors applied to an industrial sector independent of the country. As a consequence it was assumed that there is no difference in technology throughout Europe for the reference year 1990. For municipal solid waste incineration, only the Western European countries were given a lower emission factor of 40 mg I-TEQ/kton of waste incinerated whereas for all other regions an emission factor of 150 mg I-TEQ/kton was assigned. An emission factor of 40 mg I-TEQ/kton corresponds to a stack concentration of 8 ng I-TEQ/m³ and a factor of 150 mg I-TEQ/kton to a concentration of 40 ng I-TEQ/m³. The limit concentration of 0.1 ng I-TEQ/m³ established by many countries for municipal solid waste incineration would result in an emission factor of 0.5 mg I-TEQ/kton.

For the year 1990, there are hardly any measurements available from the ferrous and non-ferrous metal industries as well as for crematoria, *etc.* and thus, the TNO estimates cannot be verified by actual measurements. Nevertheless, the relation between industrial sectors does not seem to be supported on present knowledge. As an example, it is hard to understand that the most advanced municipal solid waste incinerators on an average emit eight times more dioxins than sinter plants (based on all existing plants). In addition, it is well known that the copper industry is much more susceptible to generating PCDD/PCDF than the lead sector. Until present, PCDD/PCDF have not been detected in the emissions of kerosene combustion.

The total PCDD/PCDF emissions are summarized in Table 42. The total PCDD/PCDF emissions were calculated to 11,300 g I-TEQ for the reference year 1990. The major emitters were stationary combustion processes with 4,300 g I-TEQ/a. The largest single source was waste incineration with 2,660 g I-TEQ/a. The contribution to the total inventory by country is given in Table 43.

Aggregated emissions are summarized in Table 44. For the OSPARCOM (15) and EU member states (15), the sectors with the highest PCDD/PCDF emissions were municipal waste incineration, combustion of hard coal and other fuels and combustion processes in sinter plants. For the HELCOM (9) and the Central and Eastern European countries (19) combustion of hard coal was identified as the major contributor to the dioxin emission inventory followed by sinter plants and the copper industry.

The TNO European Emissions Inventory was established for a broad range of compounds, heavy metals and organic substances, and makes a lot of generalizations. It determined emission factors and does not account for differences in technology. Especially for PCDD/PCDF, where so many measurements are available and almost all potential source categories have been analyzed for their air emissions, the methodology applied here is too coarse and the data too outdated for the real situation today. The document should be taken for screening purposes for a large number of chemicals and for countries where no data are available. To establish a national dioxin inventory, experiences from other countries and from their inventories should be applied.

Table 41: Region-specific default emission factors for PCDD/PCDF
Emission factors are in mg I-TEQ/kton or µg I-TEQ/t
NW = Northwestern Europe; S = Southern Europe
C+E = Central and Eastern Europe

<table>
<tr><th>Branch</th><th>Description</th><th>NW</th><th>S</th><th>C+E</th></tr>
<tr><td>Transformation brown coal</td><td>All types of brown coal</td><td colspan="3">0.10</td></tr>
<tr><td>Transformation fuel oils</td><td>Light fuel oils</td><td colspan="3">1.0</td></tr>
<tr><td>Transformation fuel oils</td><td>Heavy fuel oil, crude oil</td><td colspan="3">1.0</td></tr>
<tr><td>Transformation hard coal</td><td>All hard coal types</td><td colspan="3">0.10</td></tr>
<tr><td>Transformation other fuels</td><td>Other solid fuels</td><td colspan="3">0.10</td></tr>
<tr><td>Transformation other fuels</td><td>Peat</td><td colspan="3">0.10</td></tr>
<tr><td>Small comb. sources brown coal</td><td>Brown coal briquettes</td><td colspan="3">10</td></tr>
<tr><td>Small comb. sources brown coal</td><td>Brown coal, lignite coke</td><td colspan="3">10</td></tr>
<tr><td>Small comb. sources fuel oils</td><td>Fuel oils light</td><td colspan="3">1.0</td></tr>
<tr><td>Small comb. sources fuel oils</td><td>Heavy fuel oil, crude oil</td><td colspan="3">1.0</td></tr>
<tr><td>Small comb. sources hard coal</td><td>All hard coal types</td><td colspan="3">10</td></tr>
<tr><td>Small comb. sources other fuels</td><td>Other solid fuels</td><td colspan="3">10</td></tr>
<tr><td>Small comb. sources other fuels</td><td>Peat</td><td colspan="3">10</td></tr>
<tr><td>Small comb. sources other fuels</td><td>Wood</td><td colspan="3">5.0</td></tr>
<tr><td>Industry brown coal</td><td>All types of brown coal</td><td colspan="3">1.0</td></tr>
<tr><td>Industry fuel oils</td><td>Fuel oils light</td><td colspan="3">1.0</td></tr>
<tr><td>Industry fuel oils</td><td>Heavy fuel oil, crude oil</td><td colspan="3">1.0</td></tr>
<tr><td>Industry hard coal</td><td>All hard coal types</td><td colspan="3">1.0</td></tr>
<tr><td>Industry other fuels</td><td>Other solid fuels</td><td colspan="3">1.0</td></tr>
<tr><td>Industry other fuels</td><td>Peat</td><td colspan="3">1.0</td></tr>
<tr><td>Industry other fuels</td><td>Wood</td><td colspan="3">1.0</td></tr>
<tr><td>Electric arc furnace</td><td></td><td colspan="3">2.0</td></tr>
<tr><td>Sinter plants</td><td></td><td colspan="3">5.0</td></tr>
<tr><td>Cu industry</td><td>Secondary production</td><td colspan="3">20</td></tr>
<tr><td>Pb industry</td><td>Secondary production</td><td colspan="3">20</td></tr>
<tr><td>Other transport combustion</td><td>Diesel</td><td colspan="3">1.0</td></tr>
<tr><td>Other transport combustion</td><td>Heavy fuel oil, patent fuel</td><td colspan="3">1.0</td></tr>
<tr><td>Other transport combustion</td><td>Kerosene</td><td colspan="3">1.0</td></tr>
<tr><td>Other transport combustion</td><td>Motor gasoline</td><td colspan="3">1.0</td></tr>
<tr><td>Waste incineration</td><td>Municipal waste</td><td>40</td><td colspan="2">150</td></tr>
</table>

Table 42: PCDD/PCDF emissions to air in 1990m for 38 European countries (UBA 1997). Fluxes in g I-TEQ/a

Total	**11300**		
Stationary combustion	**4300**	**Production processes**	**3630**
Publ. power, cogen., district heating	594	Iron & steel	1960
Public power etc. brown coal	48.2	Coke production	5.43
Public power etc. hard coal	48.1	Pig iron	16.8
Public power etc. fuel oils	136	Open hearth furnace	7.93
Public power etc. other fuels	361	Basic oxygen furnace	19.4
Comm. Institutional, residential comb.	2450	Electric arc furnace	287
Commercial etc. brown coal	381	Sinter plants	1650
Commercial etc. hard coal	1440	Foundries	4.00
Commercial etc. fuel oils	156	Non-ferrous metal industry	1610
Commercial etc. other fuels	471	Al industry	37.8
Industrial combustion	1270	Cu industry	1500
Industrial combustion brown coal	82.1	Other non-ferrous metal industry	14.4
Industrial combustion hard coal	633	Pb industry	18.0
Industrial combustion fuel oils	332	Zn industry	30.0
Industrial combustion other fuels	219	Organic chemical industry,	
Solvent use	**3.40**	here: Halogenated HC production	0.24
Wood preservation	0.40	Paper pulp (Kraft process)	0.10
Road transport	**73.1**	Paper and pulp industry	4.50
Other mobile sources & machinery	**63.0**	Road paving with asphalt	1.00
Waste treatment & disposal	**2710**	Cement industry	19.1
Waste incineration	2660	Glass industry	0.40
Landfill	19.9	**Nature**	**210**
Cremation	23.1		

Table 43: 38 European countries: PCDD/PCDF emissions in 1990 by country (UBA 1997). Fluxes in g I-TEQ/a

Country	Flux/a	Country	Flux/a
Albania	12.1	Ireland	43.9
Austria	84.8	Iceland	0.553
Belgium	616	Italy	583
Bulgaria	154	Lithuania	23.0
Bosnia and Herzegovina	7.13	Luxembourg	27.6
Belarus	106	Latvia	13.5
Switzerland	242	Moldova	22.7
Cyprus	1.02	Former Yugoslav Republic of Macedonia	4.90
Czech Republic	224	The Netherlands	505
Germany	1196	Norway	38.7
Denmark	70.6	Poland	359
Spain	134	Portugal	17.4
Estonia	17.7	Romania	1500
Finland	53.3	Russian Federation	1412
France	1636	Slovak Republic	43.0
United Kingdom	881	Slovenia	5.99
Greece	25.4	Sweden	83.5
Croatia	12.9	Ukraine	877
Hungary	167	Yugoslavia (Serbia and Montenegro)	112
		Total Europe	**11314**

Table 44: Aggregated PCDD/PCDF emissions; reference year 1990. Fluxes in g I-TEQ/a

	Flux/a
OSPARCOM (15) Belgium, Denmark, Finland, France, Germany, Iceland, Ireland, Luxembourg, The Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, United Kingdom	5840
HELCOM (9) Denmark, Estonia, Finland, Germany, Latvia, Lithuania, Poland, Russian Federation, Sweden	3230
EU (15) Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Italy, Luxembourg, The Netherlands, Portugal, Spain, Sweden, United Kingdom	6250
Central and Eastern Europe (19) Albania, Belarus, Bosnia and Herzegovina, Bulgaria, Croatia, Czech republic, Estonia, Former Yugoslav Rep. of Macedonia, Hungary, Latvia, Lithuania, Moldova, Poland, Romania, Russian Federation, Slovak Republic, Slovenia, Ukraine, Yugoslavia (Serbia and Montenegro)	5070

9 DIOXIN INVENTORIES FROM THE AMERICAN CONTINENTS

9.1 Canada

As part of the mandate given to the Federal/Provincial Task Force on Dioxins and Furans to develop an inventory of sources of releases of PCDD/PCDF in Canada, a report was published in January 1999 (Environment Canada 1999). The purpose of the inventory report is to assist the Federal/Provincial Advisory Committee for the Canadian Environmental Protection Act (CEPA-FPAC) in identifying priority sectors for the development of prevention or reduction measures regarding the releases of dioxins and furans (and hexachlorobenzene, HCB). The final goal is the virtual elimination of these toxic substances. The inventory report covers man-made sources known to release PCDD/PCDF in a continuous (or batch) process. The results are summarized in Table 45 to * Projections

Table 49. The report covers releases of PCDD/PCDF to air, water, land and with products.

Between 1990 and 1997 an 18 % reduction in atmospheric releases was observed. For the year 1999, it is projected that an additional 25 % reduction will be achieved as a result of announced up-grades or closures of industrial facilities for a total reduction of 43 % compared to base year 1990. A number of priority sectors were identified and reduction measures initiated. The releases of individual sources quantified so far are summarized in Table 45.

Municipal solid waste incineration is currently the largest emitting sector in Canada. There are currently nine operating large municipal waste incinerators in Canada (66.9 g TEQ/year - 23.1 % of total). Of these, stack testing for PCDD/PCDF has been performed on eight facilities and the remaining facility emissions was estimated. Of the total 66.9 g I-TEQ/a emissions, 61.8 g I-TEQ/a is emitted from one facility in Lévis, Québec. This facility is undergoing renovations to be completed in the fall of 1998, which should reduce the dioxin/furan emissions to 0.08 g TEQ/a. In 1999, the total PCDD/PCDF release from this sector is expected to drop to 5.1 g I-TEQ/a. Measured concentrations ranged 0.0057 to 390 ng I-TEQ/m^3. Small municipal waste incinerators emitted 9.4 % in 1997. All small incinerators in British Columbia will be shut down and replaced by other facilities by the end of 1998. The third category of municipal waste incinerators in Canada is low temperature Teepee burners (45 existing) located in Newfoundland. The estimated releases accounted to 74.5 g I-TEQ in the year 1997. It should be noted that none of these facilities has been tested; the estimate is based on emissions factors from the literature.

Residential wood combustion is another important sector, which is estimated to discharge 12% of the total PCDD/PCDF inventory. For both sectors, the Task Force recommends that options should be investigated which might lead to reduced concentrations of PCDD/PCDF from existing facilities and that feasible targets and timelines be established applicable to new municipal incinerators and new residential wood combustion equipment.

There are two sintering plants in Ontario: one of these was tested for PCDD/PCDF emissions (5.6 ng I-TEQ/m^3). The actual emissions from the iron and steel sector need to be confirmed. There are 13 steel plants (electric arc furnaces) in Canada: none of these were tested for PCDD/PCDF stack emissions so far. For the inventory, European emission factors were used. It was recommended that Canadian operations should be requested to perform representative source testing for PCDD/PCDF.

A survey of all hospital incinerators in 1995 revealed that there were 219 incinerators burning bio-medical waste in Canada. Testing for PCDD/PCDF has been completed at six typical Ontario incinerators and the results of these tests extrapolated to all facilities in Canada based on the quantity of material burned. The estimated PCDD/PCDF emissions from hospital incinerators in Canada were 8.3 g TEQ/a in 1995. Since 1995, a number of hospital incinerators had been shut down, thus reducing the total number of hospital incinerators to 160 and the total releases to 2.5 g I-TEQ/a. All hospital incinerators in B.C. are expected to shut down by December 1998. This would further reduce the release from this sector.

A unique source exists in British Columbia where that the wood used in the pulp and paper industry is stored in salty water before combustion. The emissions from this sector were estimated to be 10.5 g I-TEQ in the year 1997.
Releases to water were reduced by 99 % when comparing 1990 data with 1997 data. This is the result of the adoption and implementation of pulp and paper regulations. Releases to effluents from the pulp and paper sector have been reduced to below the “measurable concentration” level as per the regulations, which is in keeping with the objective of virtual elimination. For this sector, for release to water no additional work is recommended. Emissions to water are shown in Numbers for 1999 are projections. Numbers in bold are priority sectors

Table 46. Before the adoption of the Pulp and Paper Regulations in 1992, the PCDD/PCDF released from this sector in liquid effluents amounted to approximately 450 g TEQ/a. The last status report for this sector indicates that the total release from the sector is now 5 g TEQ/a. The PCDD/PCDF releases for the Pulp and Paper sector are based on an inventory of the 1995 discharges. The high contribution from British Columbia mills is due to the character of the effluents from the coastal mills. These show the presence of higher concentrations of certain homologues. The congener pattern of these are linked to the operations of boilers that burn salty hog, where waste ash is disposed in the effluent treatment plant. Also there are indications of a pattern associated with pulping of chips from wood treated with pentachlorophenol. These activities were not associated with either Inland British Columbia mills or mills elsewhere. The unique circumstances of the coastal mills explain why the British Columbia discharges are greater than the production from these mills would account for.

Direct PCDD/PCDF releases to soil are expected as a result of the use of pesticides and of sewage sludge.· The Pest Management Regulatory Agency (PMRA) is currently quantifying the releases resulting from the use of pesticides.· Sewage sludge has been identified as a source of PCDD/PCDF, however, the release to soil has not been yet quantified. Studies carried out indicate that this might be a concern. An additional source is in-service treated wood (utility poles and railroad ties). Results are compiled in Table 47.
Reported amounts of PCDD/PCDF in products that may lead to releases to soil are from the ash resulting from the combustion of salt laden wood in British Columbia and the disposal of treated wood. The total amounts are not expected to have changed between 1990 and 1999 (see * Projections

Table 48). The pulp and paper industry in British Columbia has measured the quantities and concentrations of PCDD/PCDF in the ash produced by these boilers. The concentration of PCDD/PCDF in the ash varied between 0.009 and 3.7 ng TEQ/g ash. The ash is disposed of in two cases in ash ponds and in eight cases in a landfill. PCDD/PCDF are formed due to the fact that the wood used is stored in the salty water before combustion. This problem is unique to British Columbia, it is not found at mills on the east coast.

Table 45: Canada: PCDD/PCDF releases to the atmosphere for three different years. Fluxes in g I-TEQ/a

	1990	1997	1999 *	Initiative/Comments
Municipal Waste Incineration	204	**152**	**82.2**	CCME GL
Wood Combustion (residential)	35.7	35.7	35.7	
Iron Manufacturing: Sintering Plants	42.9	**42.9**	**23.5**	Shut-down of Algoma, ON (98/6)
Pulp & Paper: Boilers Burning Salt Laden Wood	10.5	10.5	10.5	WG in BC, developing reduction options
Steel Man.: Electric Arc Furnaces	9.1	10.2	10.2	
Fuel Combustion Diesel (Traffic)	8.7	8.7	8.7	
Oil Combustion (residential)	7.0	7.0	7.0	
Electric Power Generation	3.4	4.6	4.6	
Wood waste combustion (saw mills & P&P mills)	4.4	4.4	4.4	
Cement Kilns	2.6	2.8	2.8	CCME GL
Hospital Incinerators	8.3	2.5	2.5	CCME GL
Chemical Production (air releases)	2.2	2.0	**0.3**	
In-service Utility Poles	1.9	1.9	1.9	SOP Wood Preservation
Wood Preserving Plants	1.8	1.8	1.8	SOP Wood Preservation
Hazardous Waste Incinerators	2.1	1.3	**0.8**	CCME GL, 1 plant in Alberta permanently closed in 1998
Pulp & Paper: Kraft Liquour Boilers	0.7	0.7	0.7	
Federal Incinerators	1.3	0.6	0.6	
Steel Foundries EAF	0.4	0.5	0.5	
Sewage Sludge Incinerators	0.3	0.3	0.3	
Base Metals Smelting	0.1	0.1	0.1	
Secondary Lead Smelters	0.1	0.1	0.1	
Biomedical Waste Incineration	4.9	0.0	0.0	
Petroleum refineries (to follow)				
Total	**353**	**290**	**199**	

Numbers for 1999 are projections. Numbers in bold are priority sectors

Table 46: Canada: PCDD/PCDF in effluents

	Annual Release (g TEQ/a)			
	1990	1997	1999	Limit
Pulp and paper effluents	450.0	4.7	4.7	for 2,3,7,8-TCDD <15 pg/L for 2,3,7,8-TCDF <50 pg/L
Chemical production	3.7	0.0	0.0	
Sewage sludge (to follow)				
Total	**454**	**5**	**5**	

Table 47: Canada: PCDD/PCDF in soil

Sector	Annual Release (g TEQ/a)		
	1990	1997	1999*
Utility poles - in service	9	9	9
Railroad ties – in service	164	164	164
Total	**173**	**173**	**173**

* Projections

Table 48: Canada: PCDD/PCDF releases in products (g I-TEQ/a)

Sector	Annual Release (g TEQ/a)		
	1990	1997	1999*
Pulp d paper: Boilers burning salt laden wood	137	137	137
Out-of-service (landfilled) treated wood	89	89	89
Total	**226**	**226**	**226**

* Projections

Table 49: Canada: Summary of PCDD/PCDF releases to all media (g TEQ/a)
* Projections

	1990	1997	1999 *
Air releases	353	290	199
Effluents	454	5	5
To soil (not complete)	173	173	173
Total	**980**	**468**	**377**

* Projections

9.2 United States of America

In 1992, the U.S. Environmental Protection Agency's Office of Research and Development (ORD) began an effort to reassess the exposure and health effects associated with polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/PCDF). Originally, the exposure portion of the Dioxin Reassessment did not include an emission inventory component as there was a concern that the database for such an inventory was inadequate. However, reviewers of these early drafts strongly urged EPA to attempt an emissions inventory using the available data. Responding to this suggestion, an inventory was developed and first published in September 1994 as part of the overall draft Reassessment.

The draft Reassessment underwent reviews by both the public and EPA's Science Advisory Board (SAB). In response to all comments and the availability of additional data, a number of changes have been made to the inventory since the 1994 draft resulting in significant revisions to both the inventory structure and actual emission estimates. The information presented in this section 9.2 is taken from the April 1998 draft report, ORD’s most recent but not finalized inventory (US-EPA 1998).

In contrast to the 1992 version of the USA Emission inventory, the present inventory recognizes the time-dependence of any emission inventory and thus reports emission estimates for the two reference years 1987 and 1995. 1987 was selected primarily because, prior to this time, little empirical data existed for making source specific emission estimates. In addition, the year 1987 also corresponds roughly with the time that significant advances occurred in emissions measurement techniques and in the development of high resolution mass spectrometry and gas chromatography necessary for analytical laboratories to achieve low level detection of PCDD/PCDF in environmental samples. As a consequence, a number of facilities began upgrades specifically intended to reduce PCDD/PCDF emissions. 1995 was selected as the latest time period where almost complete information was available consistent with the timetable for producing the rest of the Reassessment document.

A key element of the inventory is the method of extrapolation from tested facilities to national estimates. Because only a few U.S. facilities in most source categories have been tested for PCDD/PCDF emissions, an extrapolation was needed to estimate national emissions for most source categories. Many of the national emission estimates were, therefore, developed using a "top down" approach. The first step in this approach is to derive from the available emission monitoring data an emission factor (or series of emission factors) deemed to be representative of the source category (or segments of a source category that differ in configuration, fuel type, air pollution control equipment, *etc*.). The emission factor was then multiplied by a national value for the activity level basis of the emission factor (*e.g.*, total kg of material processed in the United States annually). Although no categories had estimates developed from a true "bottom up" approach (*i.e.*, estimates developed using site-specific emissions and activity data for all individual sources in a category and then summed to obtain a national total), existing facility-specific emissions testing and activity level data for some source categories (*e.g.*, municipal solid waste incinerators) supported a semi-"bottom up" approach. In this approach, facility-specific annual emissions were calculated for those facilities with adequate data. For the untested facilities in the class, a subcategory (or class) emission factor was developed by averaging the emission factors for the tested facilities in the class. This average emission factor was then multiplied by the measure of activity for the non-tested facilities in the class. Emissions were summed for the tested facilities and non-tested facilities. It should be noted that EPA used identical emission factors for a given subcategory for the two reference years 1987 and 1995 although it can be assumed that PCDD/PCDF emissions decreased due to improved designand operational practices (*e.g.* for MSWI, there exist 17 subcategories based on furnace type and Air Pollution Control Devices).

The inventory work in the USA is still on progress. One of the ongoing activities are the investigations of the vinyl and PVC manufacturing industries to better characterize the emissions of PCDD/PCDF from this sector. There are additional samples being analyzed. This initiative is accompanied by a third party review panel to improve the study.

The April 1998 External Review Draft “The Inventory of Sources of Dioxin in the United States“ (US-EPA 1998) released by the U.S. Environmental Protection Agency (US-EPA) lists five major groups of PCDD/PCDF sources:

- Combustion sources
- Metals smelting and refining sources and processing sources
- Chemical manufacture
- Biological and photochemical processes
- Reservoir sources.

For sources in each of the above classes - with the exception of reservoir sources - emission estimates have been made for air, land, water, and products. For all estimates, lower, central, and upper estimates were given. The emission inventory was set up for the two reference years 1987 and 1995 giving an upper and a lower estimate as well as the central estimate. Here we only report the central number of the two reference years. The last column of Table 50 gives the change of the emissions in percent when comparing the two reference years. All data were labeled with a confidence rating (CR) as defined below as well as the number of tested facilities out of total facilities were reported in the 1998 Emission Inventory. A summary of the most important findings was published at DIOXIN'98, the 18th International Dioxin Symposium, held in Stockholm, August 1998 (Cleverly *et al.* 1998).

Confidence Rating	Activity Level Estimate	Emission Factor Estimate
High	Derived from comprehensive survey	Derived from comprehensive survey
Medium	Based on estimates of average plant activity level and number of plants or limited survey	Derived from testing at a limited but reasonable number of few facilities believed to be representative of source category
Low	Based on expert judgement or unpublished estimates	Derived from testing at only a few, possibly non-representative facilities or from similar source categories or foreign surveys where differences in industry practices may be likely

EPA is aware that there are several thousand individual dioxin sources existing. Of these only 10 % were characterized for their PCDD/PCDF releases. A total of 390 sources were analyzed for PCDD/PCDF. For some of the source categories quite good coverage was achieved: for example, all 104 kraft pulp mills were analyzed for PCDD/PCDF. A much smaller number facilities were tested in other sectors; *e.g.* only one of the 24 secondary copper smelters was analyzed for stack emissions. So far, none of the ferrous metal processing plants is analyzed within the frame of the US inventory.

In addition, EPA makes clear that there are sources that have not been properly characterized so far and which are not included in the present draft inventory. Sources such as landfill fires, backyard trash burning, *etc.* can be very important (Lemieux *et al.* 1997). Of the chemical sources with releases in the product, the largest amount is associated with pentachlorophenol. However, the EPA inventory does not mention the PCDF present in PCB at all (Table 51).

The present findings were summarized by US-EPA as follows:

- The environmental releases of PCDD/PCDF in the United States occur from a wide variety of sources, but are dominated by releases to the air from combustion sources.

- Insufficient measurement data are available to comprehensively estimate source releases of PCDD/PCDF to water and land.

- Central estimates of dioxin releases to all environmental media were approximately 12 kg TEQ in 1987 and 3 kg TEQ in the year 1995. As a reflection of uncertainties in the estimate for individual sources, PCDD/PCDF releases ranged from 5 kg to about 30 kg TEQ in 1987 and from 1.2 kg TEQ to about 8 kg in 1995, respectively.

- Comparison of the central estimates reveals that there was approximately a 75 % reduction of dioxin releases in the United States between 1997 and 1995. This reduction was primarily caused by reductions in air emissions from municipal and medical waste incinerators.

Table 50: USA: PCDD/PCDF inventory to the air; reference year 1995. All fluxes in g TEQ/a

Emission Source	Central 1987	Central 1995	Changes
Waste Incineration			
Municipal waste incineration	7,900	1,100	-86 %
Hazardous waste incineration	5	5.7	+20 %
Boilers/industrial furnaces	1	0.38	-60 %
Medical waste/pathological incineration	2,470	477	-80 %
Crematoria		0.24	
Sewage sludge incineration	6	6.0	0 %
Tire combustion		NEG	
Pulp and paper mill sludge incineration		e	
Biogas combustion		**	
Power/Energy Generation			
Vehicle fuel combustion leaded	32	NEG	-99 %
unleaded	4	6.3	+50 %
Diesel	26	33.5	+30 %
Wood combustion residential	90	62.5	-30 %
industrial	27	29.1	+10 %
Coal combustion residential		**	
industrial/utility	63	72.8	+16 %
Oil combustion residential		**	
industrial/utility	16	9.3	-44 %
Other High Temperature Sources			
Cement kilns (haz. waste burning)	117	153	+30 %
Cement kilns (non-haz. waste burning)	14	17.8	+30 %
Asphalt mixing plants		**	
Petroleum refining catalyst regeneration		*	
Cigarette combustion	1	0.81	-20 %
Carbon reactivation furnaces		NEG	
Kraft recovery boilers		2.3	
Minimally Controlled/Uncontrolled Combust.			
Combustion of landfill gas in flares		**	
Landfill fires		**	
Accidental fires (structural)		*	
Accidental fires (vehicles)		**	
Forest, brush, and straw fires	170	208	+22 %
Backyard trash burning		**	
Uncontrolled combustion of PCB		*	
Metallurgical Processes			
Ferrous metal smelting/refining			
– Sintering plants		**	
– Coke production		**	
– Electric arc furnaces		**	
– Ferrous foundries		**	
Non-ferrous metal smelting/refining			
– Secondary aluminum smelting	10	17.0	
– Secondary copper smelting	304	541	+78 %
– Secondary lead smelting	1	1.63	+100 %
Scrap electric wire recovery		*	
Drum and barrel reclamation		NEG	
Chemical Manufacture/Processing Sources		*	

Total	**12,000**	**2,745**	**-75 %**

Legend to Table 50:

e: Included within total for wood combustion - industrial
NA Not applicable
* Some evidence exists suggesting that this category is a source of PCDD/PCDF emissions. However, insufficient data are available for making a quantitative or qualitative emission estimate.
** Evidence exists suggesting that this category is a source of PCDD/PCDF emissions. Preliminary estimates of emissions for reference year 1995 have been made, but the confidence in the emission factor estimates or activity level estimates are so low that the estimates are too uncertain to include in the inventory

Table 51: USA: PCDD/PCDF emissions to water, land and products; reference year 1995 - Central estimates (US-EPA 1998)

Emission Source	Water	Land*	Product
Waste Incineration			
Power/Energy Generation			
Minimally Controlled or Uncontrolled Combustion			
Chemical Manufacture/Processing Sources			
Bleached chemical wood pulp and paper mills	19.5	1.4	24.1
Mono- to tetrachlorophenols	NEG	NEG	NEG
Pentachlorophenol	NEG	NEG	25,000
Chlorobenzene	NEG	NEG	NEG
Chlorobiphenyls (leaks/spills)	NEG	NEG	NA
Etyhlene dichloride/vinyl chloride	*	NEG	*
Dioxazine dyes and pigments	NEG	NEG	0.36
2,4-Dichlorophenoxy acetic acid	NEG	NEG	18.4
Non-incinerated municipal	NA	207	7.0
Tall oil based liquid soaps	NEG	NEG	*
Biological Formation	*	*	NA
Photochemical Formation	*	*	NA
Total	**20**	**208**	**25,050**

e: Included within total for wood combustion - industrial
NA Not applicable
* Some evidence exists suggesting that this category is a source of PCDD/PCDF emissions. However, insufficient data are available for making a quantitative or qualitative emission estimate.
** Evidence exists suggesting that this category is a source of PCDD/PCDF emissions. Preliminary estimates of emissions for reference year 1995 have been made, but the confidence in the emission factor estimates or activity level estimates are so low that the estimates are too uncertain to include in the inventory

9.3 Others

For Central and South America, neither PCDD/PCDF emission data nor inventories are available.

10 DIOXIN INVENTORIES IN ASIA AND THE PACIFIC

10.1 Australia

On behalf of Environment Australia's Environment Protection Group (EPG), Pacific Air & Environment investigated the sources of PCDD/PCDF emissions in Australia. The report published in August 1998, aims at identifying, quantifying, and recommending technologies and strategies for the reduction of risks to human health and the environment arising from the environmental release of the twelve key POPs as identified by the United Nations Environment Programme (UNEP) Governing Council (EPG 1998).

The estimates made for Australia are not based on own emission or deposition measurements. The basic methodology used in the study has been to review international PCDD/PCDF inventory studies, and then a list of industries that release PCDD/PCDF to the atmosphere was collated. The emission factors for each of the industries were compiled and compared, and a final emission factor range was then assumed for Australia. Where possible, Australian source test data was also reviewed, however reliance on international data was significant due to the general lack of domestic source test data.

As a result of this procedure, the emission estimates made in this study are subject to uncertainty. Activity data relating to each industry source was collected from a number of sources. Sometimes only limited data was available to make best estimates. However, for some sources no activity data was found and so emissions from these industries could not be determined. Using the emission factors and appropriate activity data emissions were quantified for each industry.

The emissions as quantified using the methodology above are presented in Table 52. These estimates are subject to considerable uncertainties. Based on the upper bound of each range, biomass combustion from prescribed burning and wild bushfires are potentially the most significant sources of PCDD/PCDF compounds in Australia. These two sources contribute approximately 75 % to the total PCDD/PCDF emissions. The second major group of sources includes (in decreasing order of contribution):

- Cement production;
- Residential wood combustion;
- Coal combustion (utility and industrial);
- Sinter production;
- Industrial wood combustion; and
- Lime production.

If the fossil fuel and biomass combustion sources are grouped together, and the upper emission bounds are used, they contribute about 50 % of the emissions from these six sources. Cement and lime production account for 40 %, with sinter production accounting for the remaining 10 %. All six sources contribute about 20 % to the total emission inventory (based on upper bound estimates). Therefore, these sources combined with prescribed burning and bushfire sources account for approximately 95 % of total emissions. The most significant non-industrial source is residential wood burning, ranked second place. Other sources such as motor vehicles however are not considered significant, contributing less than 1 % to total emissions. Emissions from residential oil combustion are even less significant.

Table 52: Australia: PCDD/PCDF emission estimates (Fluxes in g TEQ/a)

Source	Flux
Fires – prescribed burning	65-1300
Bushfires	7-400
Cement	0.12-153
Residential Wood Combustion	15-98
Coal Combustion	4.5-73
Sinter Production	9-68
Industrial Wood Combustion	10-65
Lime	0.035-44
Oil Combustion (industrial and utility)	21
Non-Ferrous Metal Production	1-19
Medical Waste Incineration	0.9-19
Motor Vehicles	0.35-17
Asphalt Mixing	14.5
Iron and Steel Production	0.7-9.4
Landfill Gas	0.8-2.5
Residential Oil Combustion	0.075-0.2
Ceramic	0.02-0.05
Coke Production	0.03
Glass	0.0014-0.0035
Municipal Waste Incineration	0
Sewage Sludge Incineration	ND
Hazardous Waste Incineration	ND
Crematoria	ND
Activated Carbon Regeneration	ND
Halogen Chemicals and VCM	ND
Pesticide manufacture	ND
PCP Wood Treating	ND
Total	**150-2300**

Due to the methodology and for other reasons, it is clear that this study is subject to various shortcomings. The principal reasons are below:

- The most significant limitation is the lack of own source test data, resulting in a heavy reliance on international data. Emission factors as a tool for estimating emissions are inherently prone to uncertainties as they are typically based on limited testing of a source category. When applying these international emission data to Australian sources this uncertainty is increased. Most emission factors were taken from the USA, UK, and Dutch inventories,

- In addition, some international emission factors were originally derived using assumed conversion factors to supplement data gaps. This adds further to the uncertainties in the original emission factors.

- The emission factors for a large number of sources span several orders or magnitude. This is indicative of the potentially large variations that are observed within a particular source category. With such large ranges it becomes difficult to identify significant PCDD/PCDF contributors, particularly if the upper bound indicates that the source may be significant, while the lower bound indicates a minor contribution.

- Some source categories may have such variable process technologies, operational conditions *etc.* that it may be difficult to reliably predict emissions from these sources using limited data. Better characterization of industry will enable the identification of these industry types.

- Emission factors for many industries are based on test data taken during very short sample periods. The emission results are likely to be reflective of relatively good combustion and operational practice and therefore may not be indicative of likely emissions during process upsets and/or abnormal operation.

- The authors did not find emission factors for glass and ceramic production and residential sources and thus, added these sources as highly uncertain for their estimate. Nevertheless, there are existing data (UBA 1995).

- The Australian inventory used emissions from residential ‘wood burning stoves’ to bushfires and the prescribed burning of agricultural waste and grasslands. Due to this lack of data, and considering the highly variable conditions under which these fires can burn emissions estimated from these sources are considered highly uncertain.

- Another limitation relates to the lack of activity data for a number of sources. This lack of data prevents the compilation of a comprehensive list of sources with emission estimates. Therefore, a complete picture of the Australian situation cannot be attained. Some of the sources not included are potentially significant contributors (such as waste incineration) and so their inclusion is important.

- An additional source that was identified as a potential PCDD/PCDF emitter is accidental fires. However, due to the lack of data it was not possible to derive emission estimates. This is a source that requires further consideration.

Considering these limitations, it is stressed that the emission estimates as determined by this study are indicative only of the likely PCDD/PCDF releases by various sources in Australia. The estimates have been developed based on the best information currently available. These estimates must be further refined as additional data is collected.

The most important recommendation made by the authors is the need for Australian data. Thus, it is important that PCDD/PCDF testing will be performed over a broad range of industries. The principal focus should initially be on those industries identified in this study as being ‘potentially’ significant. It is important that pertinent information such as feed or production rates, process technologies, operating conditions and pollution control equipment utilized be recorded with any test data. Information such as operating temperatures of flue gas control systems should also be included. Additionally, it is important that all source testing is performed in line with the appropriate PCDD/PCDF source testing protocols.

As for any other country, there is a lack of activity data for a number of PCDD/PCDF sources. It is considered desirable to have better estimates for bushfires, however, this may be a difficult task. Testing of residential wood combustion equipment using various wood types is recommended to allow the development of more definitive emission factors

10.2 Japan

Hiraoka and Okajima (1994) published a PCDD/PCDF emission inventory for Japan, 1990 being the reference year. The results of the national estimate is shown in Table 53; as can be seen, only rough estimates are given.

Table 53: Japan: PCDD/PCDF annual emissions from known sources (Hiraoka and Okajima 1994)

Source	Emission (g TEQ/a)
MSW incineration	3,100-7,400
Incineration of organic chloride waste and waste oil	460
Clinical waste incineration	80-240
Sewage sludge incineration	5
paper mill sludge incineration	2
Paper mill black liquor incineration	3
Wood and waste wood incineration	0.2
Metal works	250
Cigarette smoke	16
Paper and paperboard	40
Motor vehicles	0.07
Lubrication oil	20
Total	**3,981-8,351**

In Japan, 50.3 Mio. tons of municipal solid waste was generated in 1993. Of this, 74.3 % were incinerated in 1,854 incineration facilities. At this time, MSWI were considered to be the largest source of PCDD/PCDF emissions in Japan.

An overview on the total PCDD/PCDF emitted from Japanese MSWI is shown in Table 54. The annual flux from MSWI for 1990 is based on an average PCDD/PCDF emission of more than 80 ng I-TEQ/m^3. For the next twenty years an overall reduction of PCDD/PCDF emissions from municipal solid waste incinerators by 99.6 % is assumed due to implementation of a New Guideline. The revised Air Pollution Control Law was promulgated in December 1997. The law established the following maximum concentrations:

For newly established incinerators with a capacity of 4 t/h	0.1 ng-TEQ/m^3
For new incinerators with a capacity of 2-4 t/h	1 ng-TEQ/m^3;
For new incinerators with a capacity of 2-0.2 t/h	5 ng-TEQ/m^3.

In addition, measures to reduce PCDD/PCDF concentrations in incinerator residues, like fly ash and bottom ash, are addressed in the guideline as well.

Table 54: Time trend of PCDD/PCDF emissions from MSWI in Japan and prediction for the future

Year (Period)	g I-TEQ/a	Reduction	Reference
1990	3,100-7,400		Hiraoka and Okajima (1994)
1996	4,300	35 %	Ministry of Health and Welfare (1996)
2001	590	86 %	Prediction, Hiraoka *et al.* 1997
2006	100	98 %	Prediction, Hiraoka *et al.* 1997
2016	20	99.6 %	Prediction, Hiraoka *et al.* 1997

In November 1998, the Japanese Ministry of International Trade and Industry (MITI) published a new dioxin emission inventory for Japan (Table 55). The annual emissions for the reference year 1998 were 5,300 g I-TEQ. These results are not much different from the estimate made by Hiraoka in 1990. The new estimate is based on measured emissions and investigations voluntarily conducted by the industrial sectors following the advice of MITI. According to the inventory, municipal solid waste (MSW) incineration with 4,300 g I-TEQ/a is still the largest source contributing to approximately 80 % of the total emission load. The sum of the other 27 sources investigated was around 1,000 g I-TEQ/a. In January 1997, there were still approximately 100 MSWI that had emissions greater than 80 ng I-TEQ/m³. The emissions of these facilities alone contributed to an annual load of 1,338 g I-TEQ. Immediate actions taken by the Ministry of Health should reduce these emissions to 94 g I-TEQ/a by September 1998. Hazardous waste incinerators were estimated to emit 460 g I-TEQ annually.

In addition to waste management processes, various industrial processes were investigated as well. As can be seen from Table 55, among these processes, steel manufacturing (electric furnace) processes generated 190 g I-TEQ/a which amount to 3.5 % of the total. Sintering processes generated 119 g I-TEQ/a, which is 2.2 % of the annual air input. Zinc recovery processes were estimated to generate 34 g I-TEQ/a (0.6 %), and aluminum alloy manufacture produces 16 g I-TEQ/a (0.3 %). In Japan, electric furnaces for steel manufacturing are facilities subject to special regulations within the Air Pollution Control Law. At present, for the sintering process in steel manufacture, zinc recovery and aluminum alloy manufacturing there are only industry guidelines to promote the control of PCDD/PCDF emissions on a voluntary basis. Special attention was paid to the zinc recovering processes, which utilize PCDD/PCDF-contaminated fly ash from the steel manufacturing (electric furnace) as raw material. Although most PCDD/PCDF contained in the fly ash are destroyed in the refining process, some contamination will not be destroyed and some new PCDD/PCDF may be newly generated in the process. MITI estimated an annual release of 34 g I-TEQ for the most recent year (Sakai 1999)

Table 55: Japan: PCDD/PCDF emissions to air – New estimates (MITI 1998)

Source	Air Emission g I-TEQ/a	Percent of Total
MSW incineration	4,300	80 %
Hazardous waste incineration	460	9 %
Medical waste incineration	80-240	1.6-4.6 %
Sewage sludge incineration	5	≈0 %
Steel manufacturing (electric furnace)	190	3.5 %
Steel manufacturing (sintering)	119	2.2 %
Zinc recovery	34	0.6 %
Aluminum alloy manufacturing:	16	0.3 %
Total emission into air (form quantified sources)	**5,300**	**100 %**

10.3 New Zealand

A dioxin inventory will be available soon.

10.4 South Korea

So far, only emissions from municipal solid waste incinerators were evaluated. In Korea, eleven municipal solid waste incinerators are operated since 1986 to dispose of approximately 3 % of the total waste. It is planned to increase this share up to 20 % by the year 2001. As there is a serious concern on dioxin emissions from present MSWIs, a first survey on dioxin emissions and environmental levels was conducted (Chang *et al.* 1998).

In this first stage, flue gas samples were collected from all eleven MSWI in Korea according to a modified US-EPA Method 23. The stack emissions ranged 0.07-22.33 ng I-TEQ/m^3. The total annual emissions from this source category were estimated to be 10.81 g I-TEQ for the reference year 1997. Concentrations in fly ash were found to range from 0.37 to 21.0 ng I-TEQ/g resulting in an annual total of 127 g I-TEQ (Chang *et al.* 1998).

11 GLOBAL EMISSION INVENTORY

Brzuzy and Hites (1995) studied PCDD/PCDF concentrations in soils to determine depositional fluxes on a global basis. They collected soil samples from undisturbed locations as well as sediments. For the determination of global fluxes, 107 soil samples globally were collected from several areas of the world covering the northern and the Southern hemispheres (Brzuzy and Hites 1996). To determine fluxes, the authors established a correlation between the deposition of PCDD/PCDF and various climatic zones of the world. The results of the flux determinations are shown in Table 56.

Table 56: Climate zones and average total fluxes of PCDD/PCDF to each zone (Brzuzy and Hites 1996)

Climate Zone	Area (10^{13} m²)	Average Flux (ng $m^{-2} \cdot a^{-1}$)	90 % Confidence Level
Subtropical	0.4	610	90
Temperate	2.4	280	52
Polar/Boreal	2.2	41	6
Tropical	5.5	25	7
Arid	3.7	18	8
Total Land Area	**14.2**		

The highest average fluxes were determined for the subtropical and the temperate zones. All homologue profiles were found to be the same for all climatic zones (except for the boreal zone where a slightly more even distribution was found). Multiplication of the total fluxes with the area of a given zone was used to calculate the total deposition rate. The total global deposition was estimated to 12,500±1,300 kg PCDD/PCDF per year (Note: Totals for PCDD/PCDF not TEQ!).

Up to now, there is no data on depositional fluxes to oceans. Unpublished data and non-detectable PCDD/PCDF concentrations in Antarctica, indicate that the fluxes to remote ocean areas are approaching zero. The authors add another 610±1,500 kg PCDD/PCDF/a from the flow to the oceans. Thus, the total annual flux of PCDD/PCDF from deposition to land and water gives an annual total of 13,100±2,000 kg. The authors used a factor of 60 to convert PCDD/PCDF totals (=sum of homologues, Cl_4-Cl_8) into TEQ. In TEQ, the annual depositional flux would be 220±30 kg TEQ/a.

The same authors did a rough estimate on known dioxin emissions into air. Based on the assumptions as shown in Table 57, they calculated annual global emission to 3,000±600 kg PCDD/PCDF (total), equivalent to approximately 50±10 kg I-TEQ.

Table 57: Emission factors, global production rates, average emissions, and 90 % confidence level for major PCDD/PCDF sources; reference year 1990 (Brzuzy and Hites 1996)

	Emission Factor µg/kg	Production 10^9 kg	Total Emission kg/a	90 % Confidence
Waste incineration	13	87	1,130	450
Biomass combustion	0.04	8700	350	140
Ferrous metal production	0.5	700	350	140
Cement kilns (haz. Waste)	2.6	260	680	280
Cement kilns (non-haz. W.)	0.2	1600	320	130
Sec. Copper smelting	39	2	78	31
Medical waste incineration	22 [a]	4	84	35
Unleaded fuel combustion	320 [b]	3800 [b]	1	0.4
Leaded fuel combustion	2800 [b]	3800 [b]	11	5
Total (kg PCDD/PCDF/a)			3000	600
Total (g I-TEQ/a)			**50,000**	**10,000**

[a] Emission factor and production data apply for USA only
[b] emission factor units = pg/km; production units = km/a

12 CONCLUSIONS FROM NATIONAL EMISSION INVENTORIES

Presently, there are PCDD/PCDF inventories available for 15 countries. All countries estimated emissions to the air. A few attempts were made to quantify releases to water and to soil, *e.g.* Belgium, Canada, Denmark, Sweden, and the United States. The best estimates of the most recent evaluations (time period: 1993-1997) are summarized in Table 58. Based on these data, the central estimate of total annual PCDD/PCDF emissions is approximately 10,500 g I-TEQ. The lower estimate is around 8,300 g I-TEQ/a and the upper estimate approximately 36,000 g I-TEQ/a. A few countries that reported high PCDD/PCDF emissions drive this global number. In particular, Japan published large emissions from waste incinerators. It should be kept in mid that the Japanese inventory was established in 1994 when there was not much awareness about PCDD/PCDF emissions in this country. Once aware of the importance of municipal waste incinerators as potential dioxin sources, the Japanese agencies started intensive measurement programs and analyzed more than 1,900 MSWIs for PCDD/PCDF stack emissions. The most recent estimate gave even higher numbers of more than 5,000 g I-TEQ emitted in the year 1997. With the new guideline in place for municipal waste incinerators, it can be assumed that there will be a strong downward trend for this source category in the near future. Nevertheless, based on many measurements, the updated Japanese dioxin inventory estimates a total emission of 5,300 g I-TEQ for the year 1998. This new number will add another 1,300 g I-TEQ to the "global" inventory.

A graphical sketch of the PCDD/PCDF emissions to air is given in Figure 3.

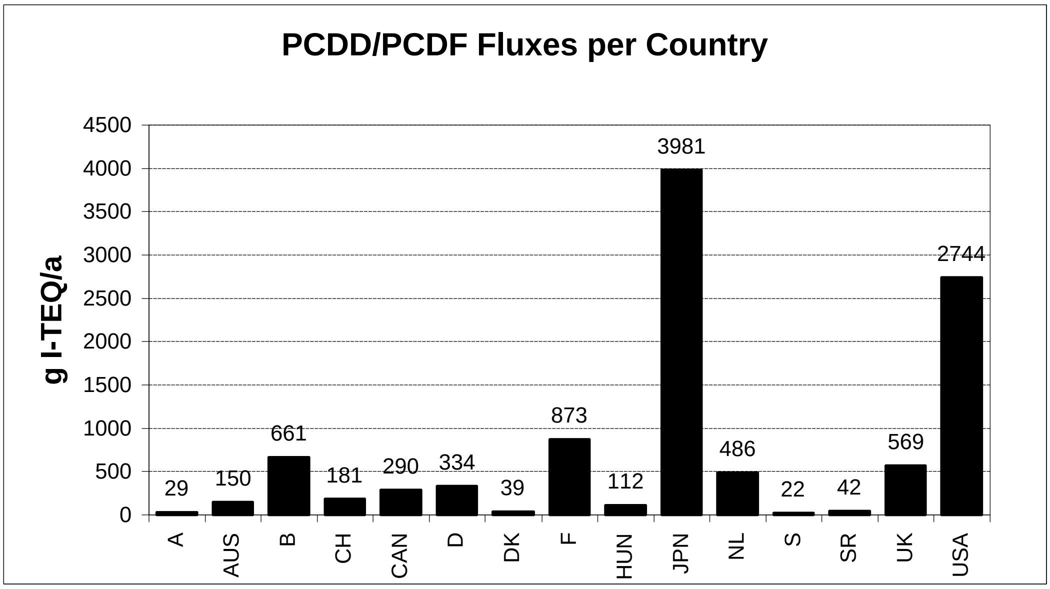

Figure 3: PCDD/PCDF annual fluxes into air (g I-TEQ/a); reference year 1995.
Total emissions from known sources = 10,500 g I-TEQ/a

The numbers generated in this survey can be compared with the EU Inventory and the TNO inventory which both cover broader geographical areas. The EU report covers 17 countries and estimates approximately 5,800 g I-TEQ/a to be emitted from known sources. However, the authors assume a large margin of uncertainty and cannot exclude an upper estimate of approximately 20,000 g I-TEQ being released into the air from the 17 European countries.

The TNO study is less specific to PCDD/PCDF sources and estimates an annual release of 1,300 g I-TEQ for the reference year 1990. Finally, based on relatively few assumptions and no detailed evaluation of emission inventories, Brzuzy and Hites estimate a global PCDD/PCDF release of 50,000 g I-TEQ/a.

Table 58: Global PCDD/PCDF emission inventory to air from known sources; based on existing national inventories; reference year 1995
* Central estimates or minimum concentrations

		Annual Emissions * g I-TEQ/a	Additional Data and Remarks	
Austria	A	29		
Australia	AUS	150	2300	Upper estimate
Belgium	B	661		
Switzerland	CH	181		
Canada	CAN	290		
Denmark	DK	39	50.05	Upper estimate
France	F	873	674-2737	Range: Min-Max
Germany	D	334		
Hungary	HUN	112		
Japan	JPN	3,981	8436	Upper estimate
The Netherlands	NL	486	58	Prediction for 2000
Sweden	S	22	88	Upper estimate
Slovak Republic	SR	42		
United Kingdom	UK	569	1,099	Upper estimate
United States of America	USA	2,744	1200-7900	Range: Min-Max
Annual Flux		**10,514**	**28,615**	**Maximum**
Korea		10.8	MSWI only	
EU Inventory		5,750	1,300-20,	17 Countries, lower and upper estimates
TNO Inventory		11,300		38 Countries, reference year 1990
Brzuzy and Hites		50,000		Global

The split of the major categories of PCDD/PCDF source sectors per country to the overall emission of 10,500 g I-TEQ/a is given in Table 59. The percent share of the major groups per country is shown in Table 60. It can be seen that the waste incineration is still the major emitter with almost 70 %. The majority of PCDD/PCDF emissions from this sector are due to the municipal (and to a lesser extent to the hazardous) waste incinerators located in Japan. This source sector alone is responsible for almost 34 % of the total PCDD/PCDF inventory to air from identified sources so far. In addition, Table 60 shows that in countries with stringent regulation of PCDD/PCDF emissions from municipal and hazardous waste incineration, *e.g.* the member states of the European Union that started legislation in the early 1990, incineration of waste is a minor contributor. A clearer picture of this finding can be seen in Figure 2 (page 4). It should be mentioned as well that in Australia there is no municipal waste incinerator at all and in other countries, landfilling is still the preferred option for waste disposal.

Table 59: PCDD/PCDF emissions per sector and country. Reference year approximately 1995. Fluxes in g I-TEQ/a

	A	AUS	B	CH	CAN	D	DK	F	HUN	JPN	NL	S	SR	UK	USA	Global Flux
Iron and Steel	10.36	10.0	59.6	9.2	53.6	181	7.63	420	12.2	250	26	2.01	7.83	34		1,083
Non-ferrous Metals		1.0	107	1.9	0.2	91.6		27	0.7		4	4.43	1.17	5.0	560	804
Power Plants			2.31		4.6	5.3	2		13.1		16.7		6.82	6.4		57
Ind. Combust. Plants	1.5	35.5	7	1.6	15.6	6.2		23			2.7				111	204
Small Comb. Units	16.7	15.1	122	27.4	42.7	7.1	3.17		27.2			4.25		25.4	62.5	354
Waste Incineration	0.179	1.70	303	127	156	32.1	25.3	402	46.5	3645	402	3.01	26	483	1589	7,241
Road Transport	0	0.35	1.7	0.9	8.7	4.8	0.2	1	0.27	0.07	7	0.88	0.35	1.0	39.8	67
Mineral Production		0.18	54.2	0.7	2.8	2.4	0.08					2.86	0.04	0.3	171	234
Others	0.024	86.5	4.66	12.7	5.7	2.9	0.26		12.32	86.2	27.5	4.82	0.19	14.6	211	470
Sum	28.8	150	661	181	290	334	38.7	873	112	3981	486	22.3	42.4	569	2744	10,514

Table 60: PCDD/PCDF emissions per sector and country in percent of the total annual emission (10,514 g I-TEQ/a). Reference year approximately 1995

	A	AUS	B	CH	CAN	D	DK	F	HUN	JPN	NL	S	SR	UK	USA	Global Flux
Iron and Steel	0.1%	0.1%	0.6%	0.1%	0.5%	1.7%	0.1%	4.0%	0.1%	2.4%	0.2%	0.0%	0.1%	0.3%	0.0%	10%
Non-ferrous Metals	0.0%	0.0%	1.0%	0.0%	0.0%	0.9%	0.0%	0.3%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	5.3%	8%
Power Plants	0.0%	0.0%	0.0%	0.0%	0.0%	0.1%	0.0%	0.0%	0.1%	0.0%	0.2%	0.0%	0.1%	0.1%	0.0%	1%
Ind. Combust. Plants	0.0%	0.3%	0.1%	0.0%	0.1%	0.1%	0.0%	0.2%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	1.1%	2%
Small Comb. Units	0.2%	0.1%	1.2%	0.3%	0.4%	0.1%	0.0%	0.0%	0.3%	0.0%	0.0%	0.0%	0.0%	0.2%	0.6%	3%
Waste Incineration	0.0%	0.0%	2.9%	1.2%	1.5%	0.3%	0.2%	3.8%	0.4%	34.7%	3.8%	0.0%	0.2%	4.6%	15.1%	69%
Road Transport	0.0%	0.0%	0.0%	0.0%	0.1%	0.0%	0.0%	0.0%	0.0%	0.0%	0.1%	0.0%	0.0%	0.0%	0.4%	1%
Mineral Production	0.0%	0.0%	0.5%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	1.6%	2%
Others	0.0%	0.8%	0.0%	0.1%	0.1%	0.0%	0.0%	0.0%	0.1%	0.8%	0.3%	0.0%	0.0%	0.1%	2.0%	4%
Sum	0.3%	1.4%	6.3%	1.7%	2.8%	3.2%	0.4%	8.3%	1.1%	37.9%	4.6%	0.2%	0.4%	5.4%	26.1%	100%

Country codes are as follows:

A Austria	AUS Australia	B Belgium	CH Switzerland	CAN Canada	D Germany
DK Denmark	F France	HUN Hungary	JPN Japan	NL Netherlands	S Sweden
SR Slovak Republic	UK United Kingdom	USA United States of America			

The present situation of PCDD/PCDF inventories in terms of geographic coverage and methods used can be summarized as follows:

- The present number of national PCDD/PCDF emission inventories is very small (15 based on national data).

- Most data is available for countries from Western Europe and Northern America. However, the inventory from the United States is still in a draft stage. From Asia, there is only one inventory for Japan covering a few sectors and the estimate for MSWIs for South Korea. From the Southern Hemisphere, so far, only Australia has estimated annual emissions based on emission factors from the literature. From Africa, Central and Southern America, there are no data at all.

- Some countries have based their inventories on emission factors generated outside the own country. For some sources, there are no emission factors determined, *e.g.* open garbage burning, landfill fires, *etc.*

- Most inventories cover emissions to air only; there are few estimates on emissions to water and land or with products. If existing, the estimates have a high degree of uncertainty.

From the existing inventories the following conclusions can be drawn:

- Amongst the source sectors, the best coverage exists for municipal solid waste incineration for both stack emission measurements and activity rates. This sector also undergoes the most dramatic changes in technology and thus, emission factors and PCDD/PCDF emissions change rapidly. As a consequence, strong downward trends are recognized in countries with modern technology or stringent legislation.
- The sector of hazardous waste incineration is relatively homogeneous and does not present a major source in any country. However, it should be taken into account that the such evaluation is based on data from industrialized countries and such results not necessarily apply for less developed countries.
- There is only limited information available from the iron and steel-producing sector. Some European countries have identified this sector as the most major contributor to national dioxin inventories. The United States and Canada are aware of these sources, but so far, no measurements have been performed. Here, generation of reliable data is urgently required.

- From the few examples on dioxin and furan emissions to water, land and with products it can be concluded that emissions to water only cover wastewaters from the pulp and paper industry. Contamination in products largely is limited to pentachlorophenol (PCP) and the PCDF in polychlorinated biphenyls (PCB) are being ignored.
- For some countries, the inventory should be updated to improve estimates of the present situation, especially where more stringent regulation has been established since the current inventory.

- Presently there exists no harmonized methods for generating and evaluating data for national PCDD/PCDF inventories. In addition, the coverage of sources varies from country to country. Some countries such as the United Kingdom and the United States to a certain extent include releases of PCDD/PCDF to landfills and land. Some countries give ranges of lower and upper estimates whereas other countries use mean/median values to calculate the annual dioxin emissions for a given source. Harmonization of data acquisition and evaluation is an obvious need and will help to better compare national dioxin inventories.

- Finally, harmonization of protocols for sampling stack emissions, water, soil, *etc.* and for analyzing these samples is highly recommended.

The present report should only be seen as a snapshot on PCDD/PCDF emissions and estimates of total releases of these compounds into the environment. Only major sectors of PCDD/PCDF releases into the air were identified. Further, PCDD/PCDF sources may exist which have not yet been identified nor quantified, especially in geographic areas with no existing data.

Presently, the coverage is not sufficient to estimate accurately global emissions of PCDD/PCDF. Nevertheless, there are several efforts underway to identify dioxin sources better in parts of the world where so far, there is no information available. In addition, existing inventories will be updated, as it is obvious that measures were taken by many countries to reduce emissions of PCDD/PCDF into the environment. For some industrialized countries in Europe and North America, strong downward trends were observed during the last years. Implementation of dioxin abatement technologies in industrial sectors and advanced combustion technology will help to reinforce such trends.

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APPENDIX A

Table A 1: International Toxicity Equivalency Factors (I-TEFs) for PCDD/PCDF (Kutz *et al.* 1988)

Congener	I-TEF
2,3,7,8-Cl_4DD	1
1,2,3,7,8-Cl_5DD	0.5
1,2,3,4,7,8-Cl_6DD	0.1
1,2,3,7,8,9-Cl_6DD	0.1
1,2,3,6,7,8-Cl_6DD	0.1
1,2,3,4,6,7,8-Cl_7DD	0.01
Cl_8DD	0.001
2,3,7,8-Cl_4DF	0.1
1,2,3,7,8-Cl_5DF	0.05
2,3,4,7,8-Cl_5DF	0.5
1,2,3,4,7,8-Cl_6DF	0.1
1,2,3,7,8,9-Cl_6DF	0.1
1,2,3,6,7,8-Cl_6DF	0.1
2,3,4,6,7,8-Cl_6DF	0.1
1,2,3,4,6,7,8-Cl_7DF	0.01
1,2,3,4,7,8,9-Cl_7DF	0.01
Cl_8DF	0.001

For all non-2,3,7,8-substituted congeners, no TEF has been assigned.

Table A 2: WHO Toxicity Equivalency Factors (WHO-TEFs) for PCDD/PCDF (WHO 1998)

Congener	Humans/Mammals	Fish	Birds
2,3,7,8-Cl_4DD	1	1	1
1,2,3,7,8-Cl_5DD	1	1	1
1,2,3,4,7,8-Cl_6DD	0.1	0.5	0.05
1,2,3,7,8,9-Cl_6DD	0.1	0.01	0.01
1,2,3,6,7,8-Cl_6DD	0.1	0.01	0.1
1,2,3,4,6,7,8-Cl_7DD	0.01	0.001	<0.001
Cl_8DD	0.0001	-	-
2,3,7,8-Cl_4DF	0.1	0.05	1
1,2,3,7,8-Cl_5DF	0.05	0.05	0.1
2,3,4,7,8-Cl_5DF	0.5	0.5	1
1,2,3,4,7,8-Cl_6DF	0.1	0.1	0.1
1,2,3,7,8,9-Cl_6DF	0.1	0.1	0.1
1,2,3,6,7,8-Cl_6DF	0.1	0.1	0.1
2,3,4,6,7,8-Cl_6DF	0.1	0.1	0.1
1,2,3,4,6,7,8-Cl_7DF	0.01	0.01	0.01
1,2,3,4,7,8,9-Cl_7DF	0.01	0.01	0.01
Cl_8DF	0.0001	0.0001	0.0001

Table A 3: Toxicity Equivalency Factors (TEFs) for *non-ortho* and *mono-ortho* substituted PCB (WHO 1997)

PCB Congener	Substitution	IUPAC No.	WHO-TEF Humans	WHO-TEF Fish	WHO-TEF Birds
Non-ortho substituted:	3,3',4,4'-	77	0.0001	0.0005	0.1
	3,4,4',5-	81	0.0001	0.0001	0.05
	3,3',4,4',5-	126	0.1	0.005	0.1
	3,3',4,4',5,5'-	169	0.01	0.00005	0.001
Mono-ortho substituted:	2,3,3',4,4'-	105	0.0001	<0.000005	0.0001
	2,3,4,4',5-	114	0.0005	<0.000005	0.0001
	2,3',4,4',5-	118	0.0001	<0.000005	0.00001
	2',3,4,4',5-	123	0.0001	<0.000005	0.00001
	2,3,3',4,4',5-	156	0.0005	<0.000005	0.0001
	2,3,3',4,4',5'-	157	0.0005	<0.000005	0.0001
	2,3',4,4',5,5'-	167	0.00001	<0.000005	0.00001
	2,3,3',4,4',5,-	189	0.0001	<0.000005	0.00001
Di-ortho substituted:	2,2',3,3',4,4',5	170	0	0	0
	2,2',3,4,4',5,5'	180	0	0	0

APPENDIX B: DIOXIN AND FURAN PROFILE INFORMATION – UNEP CHEMICAL'S SURVEY

B.1 General

In 1997, UNEP Chemicals sent questionnaires to the POPs focal points requesting information on POPs uses and releases. To March 1999, responses were obtained from 68 countries; of these, 43 countries addressed dioxins and furans. Table B 1 lists in alphabetical order all countries that responded with the information if a dioxin inventory was performed or not. Although most countries are aware that there might exist sources of PCDD/PCDF, these classes of compounds have not been quantified so far. The major reason for not having identified dioxin and furan sources is lack of financial or analytical capacities.

New information giving quantitative data not yet published in the literature so far were obtained for Croatia, Finland, and Norway. The data are shown in Table B 2, Table B 3 and Table B 5. Countries like Austria, the Czech Republic, Denmark, The Netherlands, the United Kingdom, and Switzerland reported the same data as were published in the literature before and thus, these results can be found in the main part of the report under the given country's section. As the PCDD/PCDF releases reported by Germany and Sweden differ slightly from the data found in the literature and thus, are listed in this Annex as well (Table B 4 and Table B 6).

Table B 1: Dioxin and furan sources information received at UNEP Chemicals

	Dioxin Inventory	
Country	Yes	No
Armenia		X
Austria	X [1]	
Belgium	X [2]	
Benin		X
Bulgaria		X
Burkina Faso		X
Cameroon		X
Chile		X
China		X
Congo		X
Costa Rica		X
Croatia	X	
Czech Republic	?	
Denmark	X [1]	
Dominica		X
Fiji		X
Finland	X	
Gambia		X
Germany	X [1]	
Guinée		X
Ireland	X	
Republic of Korea		X
Kuwait		X

Country	Dioxin Inventory Yes	No
Lebanon	X	
Lithuania		X
Luxembourg	X [2]	
Macedonia		X
Madagascar		X
The Netherlands	X [1]	
New Zealand	X [3]	
Niger	X	
Nicaragua		X
Norway	X	
Romania		X
Rwanda		X
Singapore		X
Slovak Republic	X [1]	
Slovenia		X
Sweden	X	
Switzerland	X [1]	
Togo		X
United Kingdom	X [1]	
Vietnam	X	

[1] Same data or data sources that were used for the main part of this report
[2] Refer to attached information which was not included
[3] No quantitative data so far

B.2 Countries with Qualitative Data

B.2.1 Armenia

Studies on identification of sources started in July 1997.

B.2.2 Burkina Faso

Burkina Faso has not performed any studies to identify sources of PCDD/PCDF. However, the country is aware of some sectors that may be potential sources as large amounts are processed, *e.g.* Coal combustion, forest fires, open garbage burning, chemical products (*esp*. PCB, pesticides), traffic and transportation (using leaded gasoline and mixtures of fuels), hazardous waste incineration, and wood combustion.

B.2.3 Cameroon

Cameroon has not undertaken studies to identify sources of PCDD/PCDF but is aware of activities with the potential to generate and release PCDD/PCDF. Among the industrial processes are: PVC and paint manufacture, textile industry which uses hexachlorobenzene and pentachlorophenol. HCB is also in use in the sector of the aluminum industry that also utilizes PCB. PCP-treated wood and the timber industry are considered as potential sources. Municipal wastes, often together with cables, are burnt in open landfills as well as in public incinerators.

B.2.4 China

China refers to a publication where emissions from a waste incinerator burning PCB wastes were reported: The publication gives very high concentrations of 47,210 ng I-TEQ/kg fly ash.

Reference: Jiang Ke, L. Li, J. Jin. D. Zhang, and C. Shao (1995): Specific Congeners Analysis of Dioxins from the Emissions of a Testing Incinerator. Organohalogen Compd. **23**, 451-454

B.2.5 Costa Rica

The country is aware of potential sources such as: asphalt mixing, incineration, chemical industry, forest fires, fuel combustion, hospital waste incineration, foundries, PCB in capacitors and transformers, sintering, traffic emissions, transportation, wood combustion. There is a need for information.

B.2.6 Guinée

Potential sources may exist, *e.g.* aluminum production, calcination of bauxite, traffic, and medical waste incineration.

B.2.7 Ireland

Ireland has a number of small industries likely to emit PCDD/PCDF. Studies are required to analyze these emissions. Some sources of PCDD/PCDF identified in other countries do not exist in Ireland (*e.g.* cable burning, landfill gas incineration, municipal, hazardous and medical waste incineration). First surveys on environmental matrices such as soil and cow milk have indicated that dioxin levels are generally very low or close to detection limits.

B.2.8 Lebanon

Lebanon has initiated a study to quantify sources of PCDD/PCDF in the country by taking samples from the six counties. Sources targeted are chemicals and derivatives, plastics and rubber, hospital waste incinerators, cement kilns, metal sector, power plants and traffic emissions. Finally, one compost sample will be analyzed as well.

B.2.9 Madagascar

The country is aware of potential sources.

B.2.10 Niger

Assumes PCDD/PCDF contaminations in PCB and thus, may be present in the country. Research has been started.

B.2.11 Vietnam

The country assumes that from the Vietnam War (1961-1970) there is ca. 170 kg of dioxins in Vietnam. Assuming that all the dioxin is from the herbicide 2,4,5-T, which is exclusively contaminated with 2,3,7,8-Cl_4DD, the total mass corresponds to 170 kg TEQ (the forms received do not give the base unit).

B.3 Countries with Quantitative Data

B.3.1 Croatia

Croatia gave first estimates on emissions of PCD/PCDF into the air in the report "Emission Inventory of Air Pollutants in Republic of Croatia for 1996" published in February 1998. The methodology used is the same as described in European CORINAIR project. Croatia estimates annual emissions of approximately 96 g TEQ per year from six major sectors. Of these, almost all emissions are due to the combustion of wood (Table B 2).

Table B 2: Croatia: Estimated PCDD/PCDF releases into air. Fluxes in g I-TEQ/a

	g TEQ/a
Coal combustion	0.036
Fuel combustion	0.966
Steel industry	1.7
Traffic emissions	0.21
Medical waste incineration	0.0016
Wood combustion	92.568
Total	**95.5**

B.3.1 Finland

The emissions of PCDD/PCDF from sources in Finland are between 100 and 200 g TEQ per year. Major sources are waste incineration, sintering processes, steel industry, and wood combustion.

Table B 3: Finland: Estimated annual PCDD/PCDF releases into air. Fluxes in g TEQ/a

	g TEQ/a
Asphalt mixing installations	0.5-1
By-products of chlorinated substances manufacture	<1
Cable incineration	<1
Chemical industry	<1
Coal combustion	2
Contaminated pesticides	<1
Forest/grass fire	<1
Fuel combustion	20-30
High temperature processes	NA
Wastes incineration	30-80
Industrial processes	NA
Ironworks	NA
Landfill gas incineration	NA
Non-Fe metal industry oil combustion	5-15
Other contaminated chemicals e.g. PCBs)	NA
Pesticides	Very low
Sintering processes	15-20
Sludge incineration	1
Steel industry	5-10
Traffic emissions	1-10
Transportation	0.5-2
Hazardous waste incineration	0.3
Municipal waste incineration	1-5
Medical waste incineration	2-5
Wood combustion	10-20
Total	**98.3-198**

B.3.3 Germany

The Environmental Agency refers to two reports:

- Release Inventory and Abatement Options of POP in Germany. Umweltbundesamt, Final Report, 3/98

- Identification of Relevant Industrial Sources of Dioxins and Furans in Europe. Landesumweltamt Nordrhein-Wesstfalen, 1997

The data reported are the same as in the main part of this report. In addition, the forms refer to some abatement technologies applied in Germany as well as to the legal situation (*e.g.* phase-out of PCB and PCP, phase-out of halogenated scavengers in gasoline) or actions taken to reduce emissions of PCDD/PCDF (*e.g.* no thermal cable reclamation in Germany).

PCDD/PCDF emissions from the production of vinyl chloride are estimated to be about 76.5 mg TEQ/a. Otherwise, the emissions from the chemical industry are negligible due to the fact that the production of critical products is prohibited in Germany.

Table B 4: Germany: PCDD/PCDF releases as summarized by UBA (Federal Environment Agency)

	g TEQ/a
Asphalt mixing installations	0.03
Coal combustion	14.2
Fuel combustion	1.59
Ironworks (primary iron production)	0.79
Landfill gas incineration	0.3
Non-Fe metal industry oil combustion (without fuel used)	91.6
Pesticides	0.87
Sintering processes	168
Sludge incineration	<0.1
Steel industry	4.9
Traffic emissions	3.1
Transportation	1.6
Hazardous waste incineration	2
Municipal waste incineration	30
Medical waste incineration	0.1
Wood combustion	2.7
Others	2.3
Total	**334**

B.3.4 Norway

Norway has quantified a few sources of PCDD/PCDF. The total emissions from identified sources are 9 g TEQ per year. The two major sources are municipal solid waste incineration and production of aluminum and magnesium. A minor source is the production of the vinyl chloride monomer (chemical industry). Norway does not have sewage sludge incinerators.

Table B 5: Norway: PCDD/PCDF emissions to air

	g TEQ/a
Chemical industry (VCM)	0.05
Industrial processes (Hydro aluminum and hydro magnesium)	4.4
Sludge incineration	
Hazardous waste incineration	0.3
Municipal waste incineration	4.4
Total	**9.15**

B.3.5 Sweden

Sweden has banned several products containing PCDD/PCDF such as PCB, PCP, and 2,4,5-T. Emissions from waste incinerators are controlled. The Swedish inventory considers releases to air, water and with wastes or products. The reference year is 1993.

Table B 6: Sweden: PCDD/PCDF releases to air, water and wastes or products; reference year 1993. Fluxes in g TEQ/a

	To Air g TEQ/a	To Water g TEQ/a	With Wastes (W)/ Products (P) g TEQ/a
By-products of chlorinated substances manufacture		0.5-0.8	
Coal combustion	0.6		
Ironworks	0.5		1-15 (W)
Sintering processes	3		
Steel industry	2-19		28 (W)
Traffic emissions	0.2-1		
Transportation	1-2		
Hazardous waste incineration	0.007		
Municipal waste incineration	3		1-2 (W)
Medical waste incineration	0.001		
Wood combustion	4-18		
Bleached kraft pulp	1	2-5	0.3-7 (P)
Recycled pulp			3 (P)
Sewage sludge		0.1	4 (W)
Primary non-ferrous smelters	0.1-0.3		0.4 (W)
Secondary non-ferrous smelters	4		2 (W)
Cement kilns	3-5		
Cremation	0.4-2		
Lime burning	3-5		
Uncontrolled landfill fires	3-30		
Oil heating	0.1-3		
Natural gas heating	0.04		
Total	**23.3-93**	**2.6-5.8**	**11.7-24**