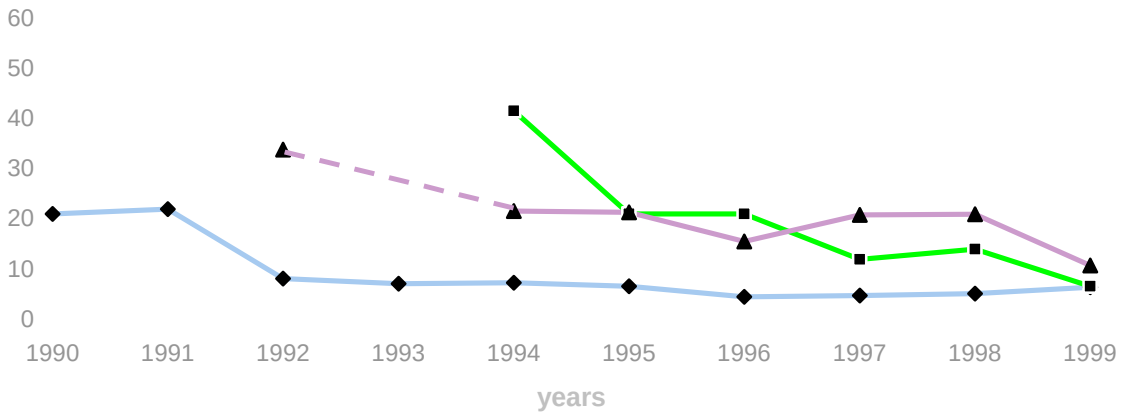
GOVERNMENT/LAENDER WORKING GROUP ON DIOXINS

DIOXINS

Data from Germany

Data on environmental pollution by dioxins 3rd report of the Government/Laender working group on Dioxins (Germany)



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Editors: Prof. Dr. Armin Basler, Federal Ministry for the Environment, Nature Conservation and Nuclear Safety,
Chairman of the Government/Laender working group on DIOXINS

Gerlinde Knetsch, Marianne Rappolder, Federal Environmental Agency,
Management of the database DIOXINS

Dr. Heidi Fiedler, Verein zur Förderung der Umweltforschung - Erziehung und Öffentlichkeitsarbeit e.V. at the University Bayreuth

Preface

All of us who have been fighting for environmental protection for decades remember the title of the book „Seveso is Everywhere" published in the late 70-ies. This book describes the conditions of the omnipresent environmental contamination by dioxins in the 70-ies. Smoking chimneys emitting dioxins have been moulding the picture of the industrial site Germany until the 80-ies; waste incineration plants have been known dioxin sources; in the surrounding of cable carbonizing plants extremely high dioxin concentrations have been measured; chlorine bleaching of paper was regarded to be the standard; polychlorinated biphenyls (PCB) and the wood preservative pentachlorophenol (PCP) showed a high pollution by dioxins. All this dioxin input into environment resulted in contaminated feedingstuffs and foodstuffs and a constant enrichment of the body fat of humans by dioxins.

The discussion going on since the late 80-ies resulted in the appeal to make any effort in order to reduce the contamination of adults by dioxins from at that time 2 picograms per kilogram of body weight and day to 1 picogram. This ambitious target considered to be unrealizable in the foreseeable future demands a drastic minimizing of new inputs into environment. Consequently it was, inter alia, necessary to tighten up the regulations on Ambient air and soil protection, to amend the Ordinance on Sewage Sludge and to take prohibitory measures and restraints as regards regulations on chemicals and environment rehabilitation measures and to monitor their respective implementation by the Laender.

The data collected in the framework of monitoring and the results of measuring programs and research projects were put into the central **data base DIOXINS** established by the Federal Environmental Agency in the early 90-ies. The transmission of data on the contamination of environment by dioxins – as a component of the Administrative Agreement between Government and Laender on Data Exchange in the Environmental Field – serves basically

- to draft an overall picture of the conditions of environmental impact,
- to provide an unrenouncable basis for setting environmental policy targets and priorities
- to provide an expert basis for deriving implementable and scientifically sufficiently secured standard and limiting values and not least
- to check the success of the activity in the field of environmental policy.

The evaluation of the approx. 10,000 samples of soil, air (emission, ambient air, deposition), biota, chemicals (substances, preparations and products), waste (sewage sludge) collected since the establishment of the data base DIOXINS is in the field of vision of the present 3rd report of the Government/Laender working group on DIOXINS. In addition, a few data on the dioxin content of feedingstuffs are available. The 4th report of the Government/Laender working group on DIOXINS presented, at the same time, contains results on the contamination of foodstuffs and humans by dioxins.
The evaluation of the measured values collected in the past 10 years documents that the environmental protection measures initiated have been successful: It was possible to drastically reduce new inputs of dioxins into environment; this had the effect that today also feedingstuffs and foodstuffs are less contaminated than before these measures entered into effect. By means of the data on foodstuff contamination there may be assessed that the target set in the early 90-ies - to reduce the intake of dioxins by adults through food by half, i.e. to about 1 picogram per kilogram of body weight and day – has been reached. The success already achieved may be documented not least by analyses of women's milk for dioxins. The documentation in the 4th report shows that the dioxin content in women's milk declined since the early 90-ies to 50 per cent of the values of the 80-ies.

Even if 25 years after Seveso the immediate averting of hazards is no longer in the fore efforts have to be made in the field of chemical and plant security and monitoring of feedingstuffs and foodstuffs to protect humans and environment against dioxins.

I wish to express my gratitude to all who collected and made available the data and to all who administered the data base and participated in evaluating the data and preparing the reports.

Jürgen Trittin
Federal Minister for the Environment, Nature Conservation and Nuclear Safety

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List of acronyms and abbreviations

AbfKlärV	Ordinance on Sewage Sludge Decree of 1992
ASE	Accelerated Solvent Extraction
Ballschmiter PCB	six PCB congeners frequently used as indicator substances for PCB. These are congeners with the IUPAC numbers: 28, 52, 101, 138, 153 and 180
BBodSchV	Federal Soil Protection and Contaminated Sites Ordinance
BgVV	former Federal Institute for Consumer Health and Veterinary Medicine
BFLR	former Federal Institute for Areal Studies and Regional Planning
BImSchV	Federal Ambient air Control Ordinance
CAC	Cation exchange capacity
dm	dry matter
DRMP	dioxin reference mersearing program
R+D	research and development project
I-TEq	international toxicity equivalents according to NATO/CCMS (North Atlantic Treaty Organisation/Committee on Challenges in Modern Society)
IS	internal standard
LTS	air-dried dry matter, equivalent to 88% dry matter
LABO	Government/Laender working group on Soil protection
-detection limit	without inclusion of the detection limit
PAC	polyaromatic hydrocarbons
PBDD	polybromated dibenzo-*p*-dioxins
PBDF	polybromated dibenzofurans
PCB	polychlorinated biphenyls
PCDD	polychlorinated dibenzo-*p*-dioxins
PCDF	polychlorinated dibenzofurans
PCP	pentachlorophenol
PXDD	polyhalogenated dibenzo-*p*-dioxins
PXDF	polyhalogenated dbenzofurans
RV	co-operative test
TCB	trichlorobenzene
TEq (WHO)	toxicity equivalents according to WHO (1998)
UAG DRMP	working subgroup “Dioxin reference measuring program” of the Government/Laender working group DIOXINS
UBA	Federal Environmental Agency

1. INTRODUCTION

The database DIOXINS of the Federal Environmental Agency is based on a decision taken by the 37th conference of the Federal and Laender Ministers for the Environment in November 1991. Thus, the Government/Laender working group on DIOXINS was commissioned with taking over the central documentation and evaluation of results of research programs as regards the pollution of various compartments by polyhalogenated dibenzo-p-dioxins (PXDD) and polyhalogenated dibenzofurans (PXDF), polychlorinated biphenyls (PCB) and further chlororganic compounds in the Federal Republic of Germany. Thereby, the database DIOXINS, first of all, concentrated on research programs initiated by the Federal Government and the Laender (GRIEM ET AL. 1997).

The data exchange between the Federal Government and the Laender is regulated by a respective administrative agreement (ANONYMUS 1996) referring notably to the compartments foodstuffs, feedingstuffs, soil, sediments, air (emission, ambient air, deposition, indoor air, dust), water, wastewater, biota (wild plants and animals), waste, recoverable materials, residues and substances, preparations and products.

Data collection forms for the various compartments were prepared by the Government/Laender working group on DIOXINS. The data were predominantly collected by the Laender, the results entered into the forms and passed on to the operators of the database.

The database is managed by the Federal Environmental Agency (evaluation of data relating to the contamination of soil, water, air, wastewater and waste, biota, substances, preparations and products by dioxins) in co-operation with the former Federal Institute for Consumer Health and Veterinary Medicine (BgVV) (responsible for human data, consumer goods, foodstuffs and feedingstuffs).

The task of the database DIOXINS is to receive and to collect measured data relating to the compounds mentioned which were collected in the Federal Republic and to evaluate this data pool with regard to the pollution level of compartments, temporal and spatial trends, information relating to the transfer from one compartment to the other etc. These evaluations are finally to be considered for proposals relating to the derivation of limits and standard values, the determination of further requirements for data and the fulfilment of national and international obligations to document the condition of the environment.

2. TOTAL INVENTORY IN SEPTEMBER 2000

Table 13 in the Appendix gives a survey of the measuring programs contained in the database DIOXINS in September 2000.

Figure 1 gives a survey of the volume of samples contained in the database DIOXINS.

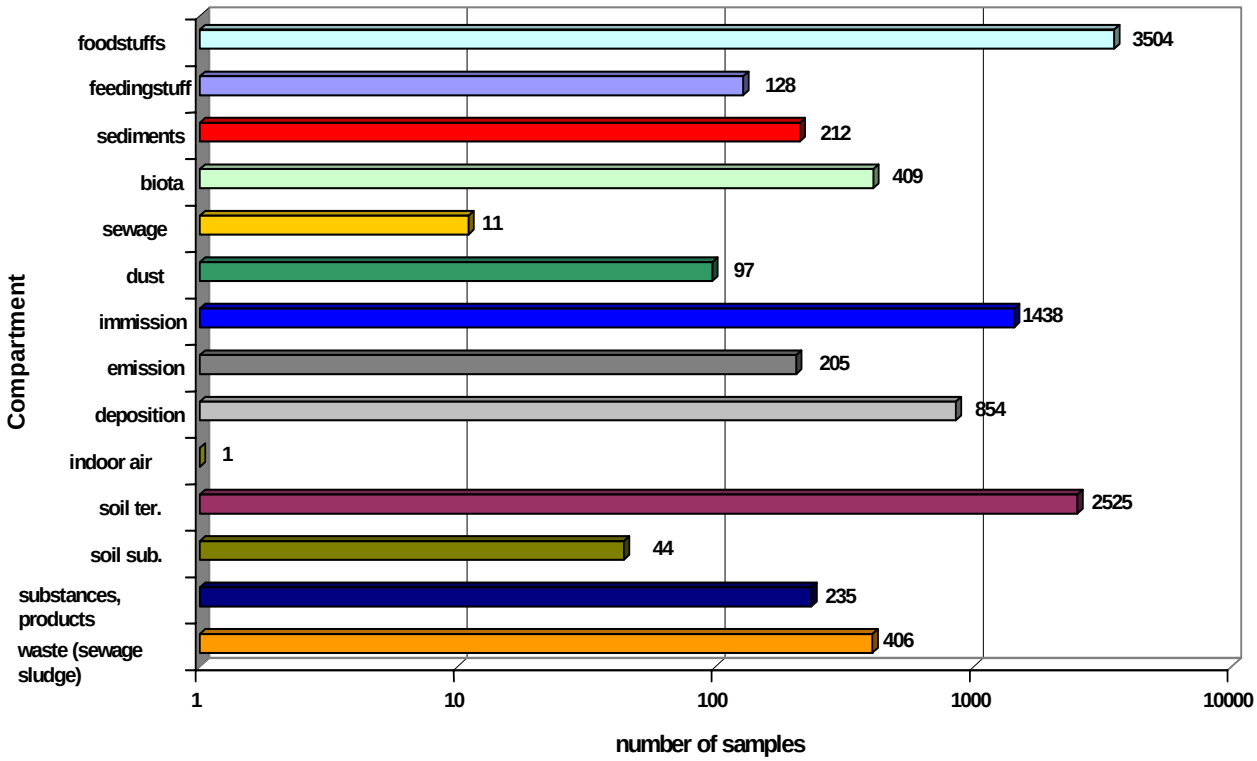

Figure 1: Number of samples contained in the database DIOXINS; up to September 2000 (Federal Environmental Agency, 2000)

The partly insignificant deviations in the number of samples with the number actually included in the evaluation are based on the fact that the Federal Environmental Agency continuously takes up data into the database.

For the environmental field samples for the compartments Soil (terrestrial), Air/ambient air and Air/deposition predominate by far. Most of the available data come from the foodstuffs domain. The compartment Wastes, Recoverable Materials, Residues is notably represented by sewage sludge samples.

3. EVALUATION PROCEDURE

Data from the database DIOXINS are evaluated separately by compartments. In particular, data from the following compartments are evaluated:

- Soil (terrestrial and subhydric)
- Air (emission, ambient air, deposition, dust, indoor air)
- Feedingstuffs
- Biota (wild plants and animals)
- Waste, residues, recoverable materials (sewage sludge, filter dust)
- Substances, preparations, products

Foodstuffs and human data are evaluated in the 4th report of the Government/Laender working group on DIOXINS.

The evaluation hereinafter follows the above-mentioned sequence of the compartments. Here, an overview of data and measurement and research projects involved. A survey of the procedure adopted in evaluating the respective number of samples included are placed in front of each compartment. This allows, on the one hand, - in conformity with the targets of the database DIOXINS – to get a quick survey of the data sets individual compartments and, on the other hand, to estimate data still required/lacking (e.g. from other regions, other matrices etc.). In addition, each compartment forms a separate unit containing all information from the database DIOXINS required for this compartment which thus may be considered and used isolated from other chapters of the report.

In conformity with the targets of the database DIOXINS primarily evaluations concerning temporal and spatial trends and the differentiation between samples from “background areas” and those from “polluted areas” were in the fore.

For the statistical evaluation of data sets we used the program package SPSS. As we were not able to assume from normally distributed data for all compartments the median, 10 percentile, 90 percentile and the minimum and maximum of the respective data sets were calculated to obtain a descriptive statistics of the data sets. The results were represented in a modification of the Box-Whisker plots (Fig. 2), with the median and 90 percentile being entered as values into the figure – as far as this was graphically possible.

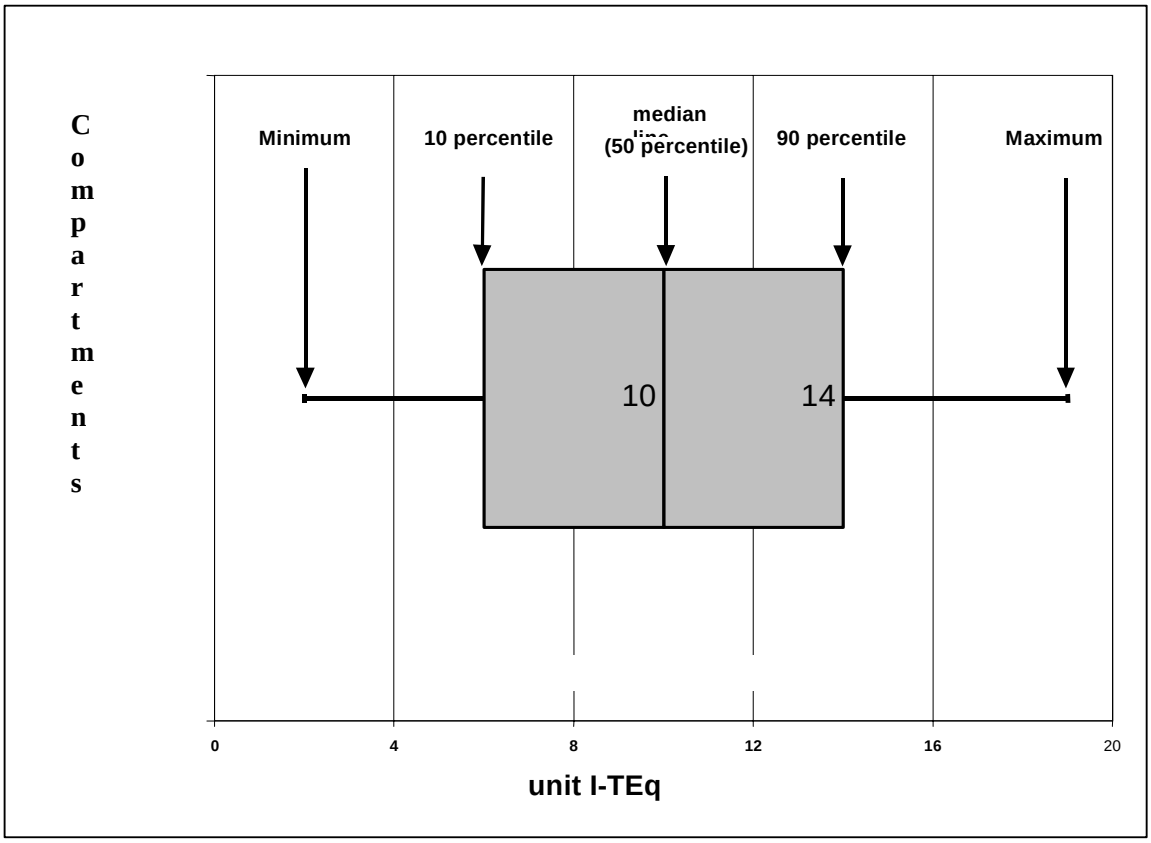

Figure 2: Modified Box-Whisker plot

Furthermore, the question about significant deviations between two data sets – with regard to not normally distributed data sets was answered with the aid of non-parametric tests. In this connection, the question whether the random samples to be compared are to be considered independent or dependent of each other was important for choosing the test method. The U-test according to Mann and Whitney was applied to two random samples considered independently and the H-test according to Kruskal and Wallis was applied to several few independent random samples.

Concentrations based on I-TEq were fixed as evaluation criterion for all compartments. Here, detection limits for non quantifiable congeners were not included (detection limit = 0) when

calculating the Teq (lower bound approach). The problems associated with including the detection limit, in particular in older investigations, will be dealt with in discussing the respective compartments.

4 COMPARTMENT SOIL

4.1 Introduction

According to the Federal Soil Protection Act the compartment Soil performs „natural functions as a basis for life and a habitat for people, animals, plants and soil organisms.“ Soil is a „part of natural systems“ performing essential functions as „a medium for decomposition, balance and restoration as a result of its filtering, buffering and substance-converting properties, and especially groundwater protection“. Furthermore, its „useful functions as land for settlement and recreation“ and „land for agricultural and forest use“ are, inter alia, in the fore (Federal Soil Protection Act, 1998). With regard to additional detrimental impacts on humans the compartment Soil plays a central part via the pathways soil-man, soil-useful plant and soil-groundwater.

Against this background and with regard to the targets of the database DIOXINS notably two problems have to be dealt with in the framework of evaluation: First of all, data relating to potential background contents shall be compiled i.e. relating to contents met in a location without special impacts. In addition, contents as they might occur in various special situations of impact shall be considered.

Information on contents without special impacts and contents in special situations of impact may be directly connected with information on action levels in view of harmful changes of soil and contaminated sites as well as remediation targets associated with them.

4.2 Data inventory in the compartment Soil, conventions and evaluation procedure

Fig. 3 gives a survey of parameters which may be put into the database DIOXINS to describe the samples in greater detail. Thereby parameters relevant to the evaluations (temporal, spatial, situation of impact) required are pointed out. Yet, the evaluation of these parameters presupposes a sufficient number of data; if this was given it also is shown subsequently.

Compartment Soil:

1. Reason for the investigation
   - environmental monitoring (y/n)
   - permanent monitoring (y/n)
   - authorization procedure (y/n)
   - investigation of contaminated sites/soil
   - research project (y/n))
   - investigation prescribed by law (y/n)
   - ***incident investigation (y/n)***
   - measuring program/name
   - miscellaneous

2. Target of investigation
   - detection of background contamination rural (y/n)
   - detection of background contamination urban (y/n)
   - detection of background contamination agglomeration (y/n)
   - special impact situation/cause
   - consideration of transfer/consideration of pathways
   - connection with other samples
   - connection with other measuring programs

3. Location data:
   - *community indicators/BLFR type* ✓
   - easting/northing values
   - *height amsl*
   - ***sampling on the territory = classified as contaminated site (y/n)***
   - sampling in nature preserve areas (y/n)
   - ***sampling on permanent training area (y/n)***
   - sampling on land expected to be used for building (y/n)
   - sampling in recultivation areas (y/n)
   - (in the case of subhydric soils: name of river, km of river)
   - ***dioxin-relevant industrial area (type/distance))***
   - ***dioxin-relevant old plant (type/distance)***
   - ***traffic routes (type/distance)***
   - ***site suspected of being contaminated (type/distance)***
   - ***placing of sewage sludge (y/n)***

4. Sampling procedure:
   - date ✓
   - subcompartment total soil (y/n)
   - subcompartment soil solution (y/n)
   - subcompartment sediment (y/n)
   - subcompartment soil matrix (y/n)
   - subcompartment soil air (y/n)
   - subcompartment suspended matter (y/n)
   - **land use** ✓
   - **org. layer (y/n)**
   - **depth from to** ✓
   - **horizon** ✓
   - type of soil
   - colour
   - **org. substance (%)**
   - rooting
   - humidity
   - PH value
   - **soil density (g/cm³)**
   - type of soil
   - random sample (y/n)
   - mixed sample (y/n)
   - suspected sample (y/n) + cause
   - mixed sample (y/n) + score
   - sampling quantity
   - sampling area
   - area random samples
   - way of sampling (grab (y/n), bore hole etc.)
   - transport of samples (vessel/conditions/duration)

5. Laboratory data:
   - storage duration
   - storage temperature
   - regulation for the preparation of samples
   - regulation for analysing
   - number IS (PCDD/PCDF, PCB)
   - quantity IS
   - confidence interval
   - participation in interlaboratory studies
   - external/internal standards
   - recovery

6. Analytical results: ✓
   - related to dry matter fine soil (y/n)
   - related to dry matter total soil (y/n)
     - dry mass (%)
     - humus content
     - pH value
     - CACeff
     - recovery
     - detection limit

Figure 3: Potential input parameters database DIOXINS compartment Soil

(**bold+italic** = identification of samples with a special situation of impact, **bold**u**+✓** = s ited for evaluation(number of samples, data quality...)

The database DIOXINS subdivides the soil samples into the subcompartments „Soil terrestrial“ and „Soil subhydric“. The present evaluation is restricted to terrestrial soil. Table 1 gives a survey of the data inventory of the database DIOXINS relating to the compartment Soil with data on the number of samples and the extent of investigation.

Considering the documentation of the individual input parameters, first of all, the following main possibilities of evaluation for the compartment Soil may be derived:

- Differentiation between suspected site and background pollution
- Suspected cause
- Year of sampling
- Federal Land
- Soil horizon
- Type of area (rural area, urbanized area, agglomeration area).

Table 1: Survey of volume of database DIOXINS - compartment Soil

Institution	Name of measuring program	Year of sampling	BFLR type A = Agglommeration S = urbanized area L = rural area	Special load - = no data - AA = old plant Al = contaminated site B = not specified KS = fertilization with sewage sludge I = industrial plant V = traffic route	Land use A = arable land G = grassland W = forest S = other	Soil horizon (only terrestric) requ.= org. requirement A = min.topsoil B = B-horiozons C = C- horizons	Number of samples	PCDD/ PCDF	PCB	subcompar-tment T = terrestric S = subhydric
Bayr. Geologisches Landesamt	Organ. problematic substances (PCB and PCDD/F) in Bavarian soil	1989/1990	A S L	- AA	A G W S	Aufl. A	464	X	X	T
Bayr. Geologisches Landesamt	Permanent soil monitoring in Bavaria	1999	A S L	-	G W	Aufl A B C	178	X		T
Freie Hansestadt Bremen – Senator für Bau und Umwelt	Measuring program for investigating the dioxin contamination in the municipality Bremen	1992-1994	A	- AA AL I V	A G S	A C	102	X		T
Freie und Hansestadt Hamburg, Behörde für Arbeit, Gesundh. u.Soziales	Investigation relating to the transfer of dioxins and furans in soil/feedingstuffs	1990-1992	A	-	A S	Aufl. A	72	X		T
Hessisches Landesamt für Bodenforschung (seit 1.1.2000 Hessisches Landesamt für Umwelt und Geologie)	Permanent soil monitoring program in the Land of Hesse	1992-1997	A S L	V I	A G W S	Aufl. A B C	157	X		T
Landesamt für Umweltschutz Saarland	Soil dioxin measuring system Saar	1993		-	A G W S	-	85	X		T
Landesamt für Natur und Umwelt des Landes Schleswig–Holstein	Soil load cadastre Schleswig – Holstein	1991-1995	S L	-	A G	A	25	X		T, S
Landesanstalt für Umweltschutz Baden-Württemberg	Soil expertise for the 2nd special waste incineration plant BW	1991	A S	-	A G W	A	20	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Soil investigation (inorg./org. contaminants) S-airport	1992	A	B	G	A	3	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Weapon elimination service Sindelfingen	1991	A	-	S	A	3	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Accompanying program for immision measurement	1992	A S	-	A S	A	29	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin composting plants	1992	A	B	S	A	2	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Environment of crematories	1991-1992	A S	B	G S	A	13	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin flooded areas	1991-1993	A S	-	A G S	A	29	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Investigation of Enz meadows	1994	A	-	G	A	20	X		T

Institution	Name of measuring program	Year of sampling	BFLR type A = Agglommeration S = urbanized area L = rural area	Special load - = no data - AA = old plant Al = contaminated site B = not specified KS = fertilization with sewage sludge I = industrial plant V = traffic route	Land use A = arable land G = grassland W = forest S = other	Soil horizon (only terrestric) requ.= org. requirement A = min.topsoil B = B-horiozons C = C- horizons	Number of samples	PCDD/ PCDF	PCB	subcompar-tment T = terrestric S = subhydric
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin sport and playing grounds	1991-1993	A S	-	A G S	A	58	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Surroundings of the clinic waste incineration plant	1990	A	-	A S	A	5	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Suspicion of damage	1992	S	-	W	Aufl.	1	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxins agriculture	1988-1989	A S	-	A G S	A	77	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Surroundings of emission sources	1990	A S	B	A G W S	Aufl. A	260	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Permanent soil monitoring I, 1st sampling	1986-1987	A S	-	A G W S	Aufl. A	24	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin forest soil	1988-1989	A S	-	W		28	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin/SM load Friesenheim Island	1989	A	-	A S		15	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Investigation composting plants/humus plants	1989	S	-	G W S		13	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Permanent soil monitoring I, 2nd sampling	1996	A	-	A W	A	2	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Pollutant contents roadsides	1988-1989	A	B (eigentl. V)	S	A	24	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Basic load urban areas	1992	S	-	A G W S	A	20	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Surroundings of a car shredder	1991	A	-	S	A	5	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Incident composting plant Wieblingen	1991	A	-	A G S	A	16	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Sampling on children's playgrounds	1990-1991	A	-	S	A	7	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin investigation in the urban area of Heidelberg	1992	A	-	A G W S	A	69	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Environmental study cupola	1992-1994	A	-	G S	A	6	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Extension of the waste heating and	1990	A	-	A S	A	8	X		T

Institution	Name of measuring program	Year of sampling	BFLR type A = Agglommeration S = urbanized area L = rural area	Special load - = no data - AA = old plant AI = contaminated site B = not specified KS = fertilization with sewage sludge I = industrial plant V = traffic route	Land use A = arable land G = grassland W = forest S = other	Soil horizon (only terrestric) requ.= org. requirement A = min.topsoil B = B-horiozons C = C- horizons	Number of samples	PCDD/ PCDF	PCB	subcompar-tment T = terrestric S = subhydric
den-Württemberg	power station North, 4th waste heating boiler									
Landesanstalt für Umweltschutz Baden-Württemberg	Small gardens Friesenheim Island	1988-1989	A	-	A S	A	13	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Former industrial sites and waste disposal sites in the urban area of Stuttgart	1992/1996	A	-	S	A B C	7	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Soil expertise for UVU waste heating and power station Ludwigsburg	1992	A	-	A S	A	4	X		T
Landesanstalt für Umweltschutz Baden-Württemberg	Sediments	1993	A	-	G	_	4	X		S
Landesumweltamt Brandenburg	Program of investigation district Oranienburg	1992/1998	A	B	A G	A B C	24	X		T
Landesumweltamt Brandenburg	Investigation of transfer in the area of Eisenhuettenstadt	1993	A	AA	G S		21	X		T
Ministerium f. ländliche Räume, Landwirdmchaft, Ernährung u. Tourismus Schleswig-Holstein	Investigation of soil and vegetable for PCDD, PCDF	1992	L	-	A		7	X		T
Lower Saxon Ministery for the Environment	Investigation of the impact on soil, mussels and sediments of the area of Wilhelmshaven	1994	S	-	G S		29	X		T
Niedersächsisches Umweltministerium	Pollution of soil by dioxins in the flood land of the Elbe river	1993	A L	-	G	_	23	X		S
Sächsisches Landesamt für Umwelt und Geologie	Dioxin reference measuring program	1995	A	B I	A W		10	X		T
Staatliches Amt für Umwelt Dessau-Wittenberg (Sachsen-Anhalt)	Study complex exploration Bitterfeld	1990-1992	L	AL	A G W S		92	X		T
Staatliches Amt für Umwelt Dessau-Wittenberg (Sachsen-Anhalt)	Investigation of the surroundings of the copper mill Ilsenburg 1990	1990	S	I	S		15	X		T
Umweltamt und Gesundheidmamt der Stadt Dortmund	Dioxin problems in the Krupp Hoesch sintering plant Westfalenhuette in Dortmund	-	A	I	S		35	X		T
Umweltbehörde Hamburg, FA für Umweltuntersuchungen	Permanent soil monitoring of the Free and Hanseatic town Hamburg	1992/1995	A	B		Aufl. A	10	X		T

Institution	Name of measuring program	Year of sampling	BFLR type A = Agglommeration S = urbanized area L = rural area	Special load - = no data - AA = old plant AI = contaminated site B = not specified KS = fertilization with sewage sludge I = industrial plant V = traffic route	Land use A = arable land G = grassland W = forest S = other	Soil horizon (only terrestric) requ.= org. requirement A = min.topsoil B = B-horiozons C = C- horizons	Number of samples	PCDD/ PCDF	PCB	subcompar-tment T = terrestric S = subhydric
Umweltbundesamt-Dioxinlabor Langen	UBA measuring systems old Federal Laender	1990-1992	A S L	B	A G W	Aufl. A B	257	X	X	T
Umweltbundesamt-Dioxinlabor Langen	Measuring system new Federal Laender	-	A S L	-	A G W	Aufl. A B C	92	X		T
Universität Bayreuth	Fertilization with sewage sludge – input of PCDD, PCDF and PCB into the food chain	1990	L	- KS	A G	A	13	X	X	T
Lehrstuhl für Bodenkunde und Boden-geographie der Uni - Bayreuth	PAC/PCB in urban soil of Bayreuth	1999					25		X	T
Landesamt für Umweltschutz Sach-sen-Anhalt	Investigation of soil in the Halle-Merseburg area for PCDD/F	1993		-	A G S	-	75	X		T

Fig. 4 represents the steps of data stepsing, the number of samples resulting from it and the steps of evaluation. In this connection, it becomes obvious that notably in evaluating the data quality with regard to calculated I-TEq values without inclusion of the detection limit an essential part of the samples had to be excluded (see hereinafter). Furthermore, lacking data on soil horizons resulted in an exclusion of bigger data Inventories. However, notably the soil horizon is a central parameter of evaluation – in particular as to distinguishing between organic layer and mineral soil horizons.

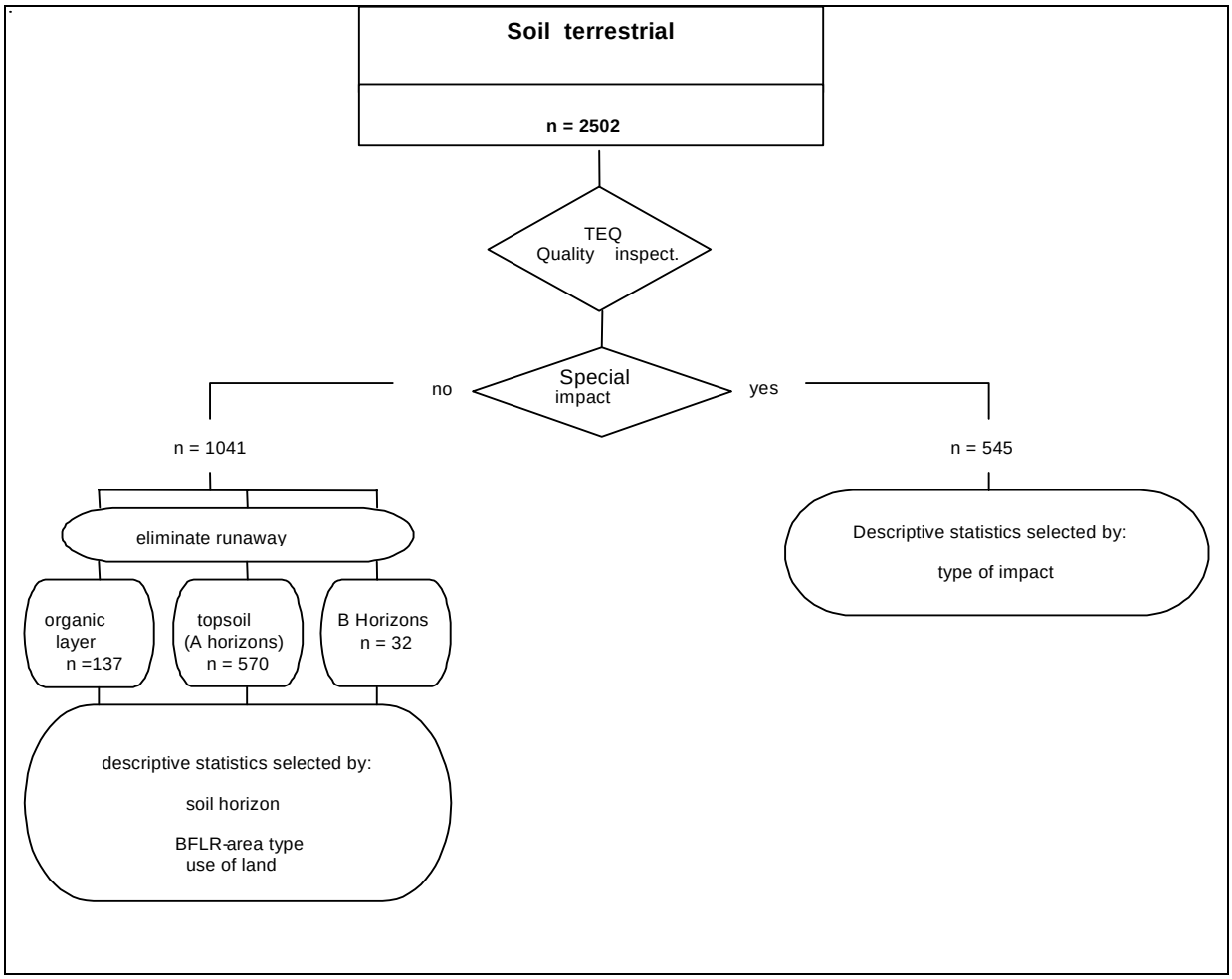

Figure 4: Steps of data stepsing and evaluation for the compartment Soil terrestrial

For the further evaluation the data inventory was, first of all, split up into samples without special impact and samples with special impact. The data pool without special impact was evaluated as to average contents and spreading with regard to temporal, spatial and use-specific differences. Evaluations of the data pool with special impact were compared with these results.

In accordance with the evaluations carried out by the Government-Laender working group on soil protection (LABO) the median values (50 percentile) and 90 percentile are in the fore also in this work for characterising background contents and their distribution, thus allowing a comparison with the results obtained there (LABO 1998) or in the framework of evaluations of data of the database DIOXINS obtained exclusively with the aid of the DRMP (UAG DRMP 2000).

4.3 General comparability of data

The comparability of the procedures applied for sampling and analysing is of central importance to the evaluation of measured results. This applies, in particular, to comprehensive data pools such as the database DIOXINS containing data of a most various origin and obtained by means of most various methods.

The database tries to consider these critical procedure indicators via the input sections “sampling” and “laboratory data”. In addition, efforts were made by the Government/Laender working group on DIOXINS, subgroup “dioxin reference measuring program”, to ensure the comparability of the procedures applied in these programs as far as possible or at least to describe the deviations. These efforts made to reach a comparability have sooner or later a direct influence on the data quality in the database DIOXINS.

In connection with the compartment Soil reference is made to the “soil data collection form” of the Federal Environmental Agency for data collection, documentation and compilation for installing permanent soil monitoring areas of the LABO ad-hoc working group on permanent soil monitoring (LABO 1999) and, in particular, to the co-operative test “dioxins in soil” of the UAG DRMP for the dioxin reference measuring program which gives a survey of the procedures applied and the deviations to be expected (Federal Environmental Agency, 1999). The evaluation of the co-operative test showed, as a rule, a good comparability of the results, thus the comparability for a joint evaluation may be considered to be given at least for the reference measuring programs.

The extraction procedure applied may be separately inquired in the database DIOXINS, for terrestrial soils, however, only data of nine measuring programs (for 378 samples) were available. When evaluating the whole data inventory considering the given spreading of the data (for the area without special impact), however, no significant deviations of individual measuring programs were to be observed which might have been interpreted as an indication to deviations in analytics.

4.4 Data quality : dioxins and calculation of I-TEq

The present report is based on the evaluation parameter I-TEq without inclusion of the detection limit. In this procedure the influence of the analysing sensitivity is to be considered. Analyses with an insignificant sensitivity, may supply here zero values (i.e. congeners not detected are considered with 0 when calculating TEq). The result is that the average values of the whole data sets will be lowered. This is the reason why data with an insufficient or lacking detection limits in calculating the parameter I-TEq (-detection limit) were excluded.

Samples were excluded if:

- more than five individual congeners were below the detection limit and the detection limits put in showed that the analyses did not correspond to the sensitivity required by the actual standard;
- values were not available for more than five individual congeners;
- for more than five individual congeners the indicated concentrations met the detection limits or the dection limit were documented instead of zero values or these detection limits did not correspond to the detection limit to be required;
- the I-TEq values could not be determined as either only data for octachlorinated congeners or only sums of homologues were available in the database; the investigation programs where these samples come from, as a rule, were carried out with targets not requiring the determination of the data of the all individual congeners.

The improvement of the analytical detection limit for dioxins is basically due to technical inovation in instruments for analysis in the early 90-ies. That is why there was expected that notably samples from investigation programs before 1992 had to be excluded.

The distribution of the excluded samples is as follows:

- before 1990: more than 75 % of all samples
- 1990 till 1992: about 50 % of all samples
- after 1992: below 15 % of all samples

Fig. 5 shows the effect of excluded samples with an insufficient detection limit by the example of topsoil samples (without further differentiation). The dioxin contents (median, 90 p.) of this data set not structured further differ e.g. always by about a factor of 2.

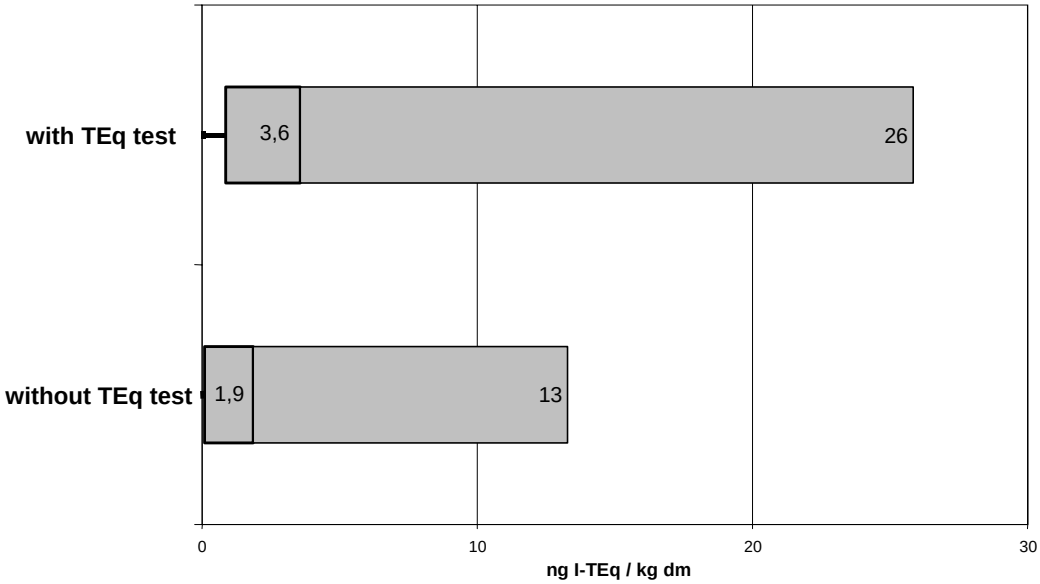

Figure 5: Comparison of PCDD/PCDF contents (median, 90 p.) in mineral topsoils with and without control of the detection limit of the data sets

4.5 PCDD/PCDF contents in terrestrial soils without special impact

4.5.1 Runaways

Fig. 6 identifies for PCDD/PCDF (I-TEq without inclusion of the detection limit) the eliminated runaways for organic layers and mineral topsoils. These samples were excluded from the further evaluation. It turns out that these data sets to be identified by means of data relating to sampling, impact etc. in the database DIOXINS as not being specially impacted contain samples which show a content by far above the usual soil loads (comp. standard values Federal Soil Protection and Contaminated Sites Ordinance (BBodSchV).

Due to the insignificant number of samples from B-horizons information on runaways was not given but just the whole data inventory was characterized (s. hereinafter).

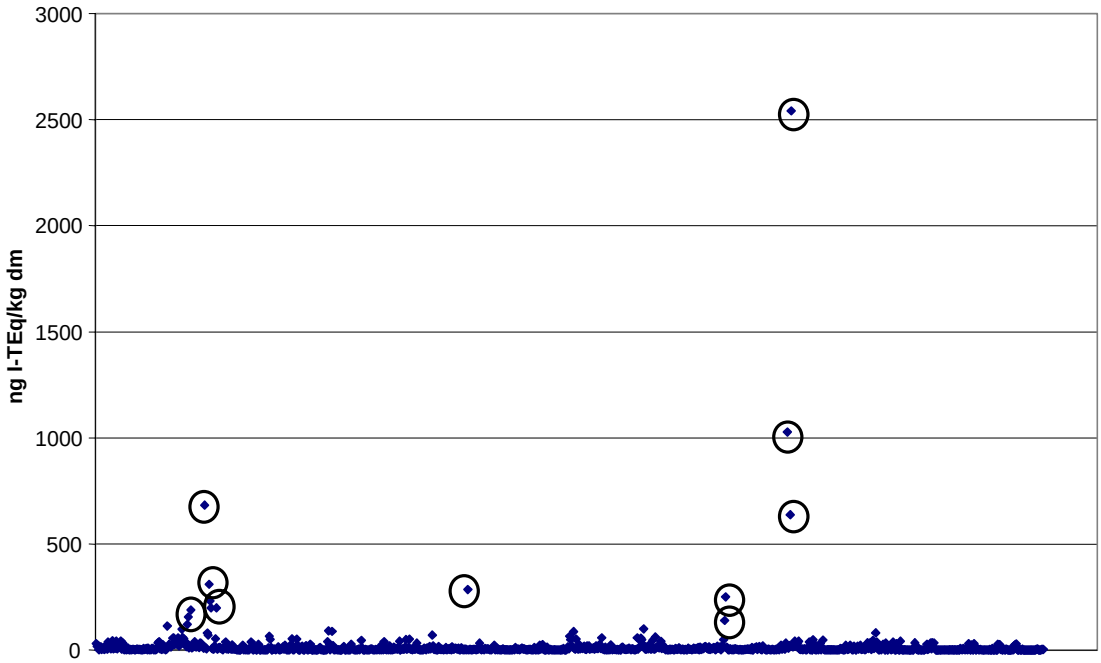

Figure 6: Runaway PCDD/PCDF (I-TEq without inclusion of the detection limit), soil terrestrial

4.5.2 Organic layers (A-horizons)

All samples from organic layers come from the use “forest and woody plants”. Not for all programs the forest use was further differentiated. Table 2 shows the programs of investigation taken into consideration.

Table 2: Measuring programs and soil use Organic layers

Institution	Name of the measuring program	Soil use
Bayerisches Geologisches Landesamt	permanent soil monitoring Bavaria	coniferous forest
Bayerisches Geologisches Landesamt	organ. problematic substances (PCB and PCDD/F) in Bavarian soil	mixed forest, coniferous forest
Freie und Hansestadt Hamburg, Behörde f. Arbeit, Gesundheit u. Soziales	investigation relating to the transfer of dioxins and furans soil/feedingstuffs	-
Hessisches Landesamt für Umwelt und Geologie	permanent soil monitoring program of the Land of Hesse	mixed forest, coniferous forest, “forest and woody plants”
Landesanstalt für Umweltschutz Baden-Württemberg	permanent soil monitoring I, 1st sampling	„forest and woody plants”
Landesanstalt für Umweltschutz Baden-Württemberg	permanent soil monitoring I, 2nd sampling	mixed forest
Landesanstalt für Umweltschutz Baden-Württemberg	dioxins forest soil	coniferous forest
Umweltbundesamt-Dioxinlabor Langen	measuring system new Federal Laender	forest and woody plants

Thus, it was only possible to evaluate the uses “coniferous forest”, “mixed forest” and the non-specific quotation “forest and woody plants” apart from the type of area (BFLR type).

When evaluating organic layers there should be considered that the results are indicated in units of dry substance weight. Yet, to compare them with other soil horizons the problems of compactness have to be considered thereby.

Fig.: 7 summarizes the descriptive statistical indicators for PCDD/PCDF of the individual uses of this compartment. Coniferous forest differs significantly in the median (U test) from the other uses. With a view of the **type of area** the differences between the “urbanized area” type with higher 90 percentile values and the “rural area” type with a higher median value may be stated. Thereby, the differences in the respective distribution of the uses are, however, to be considered. Samples of the “agglomeration” type are basically dominated by the non-specific use “forest and woody plants”, the “rural area” type basically by the use “coniferous forest”; the “urbanized area” samples involve a uniform distribution of the individual uses (see descriptions as in Table 2). Differences in the type of area are to be evaluated also against the background of this distribution.

Altogether, PCDD/PCDF contents for organic layers without special impact in the range below 50 ng I-TEq/kg dm (90 p.) or below 25 ng I-TEq/kg dm (median) may be derived from the present data set.

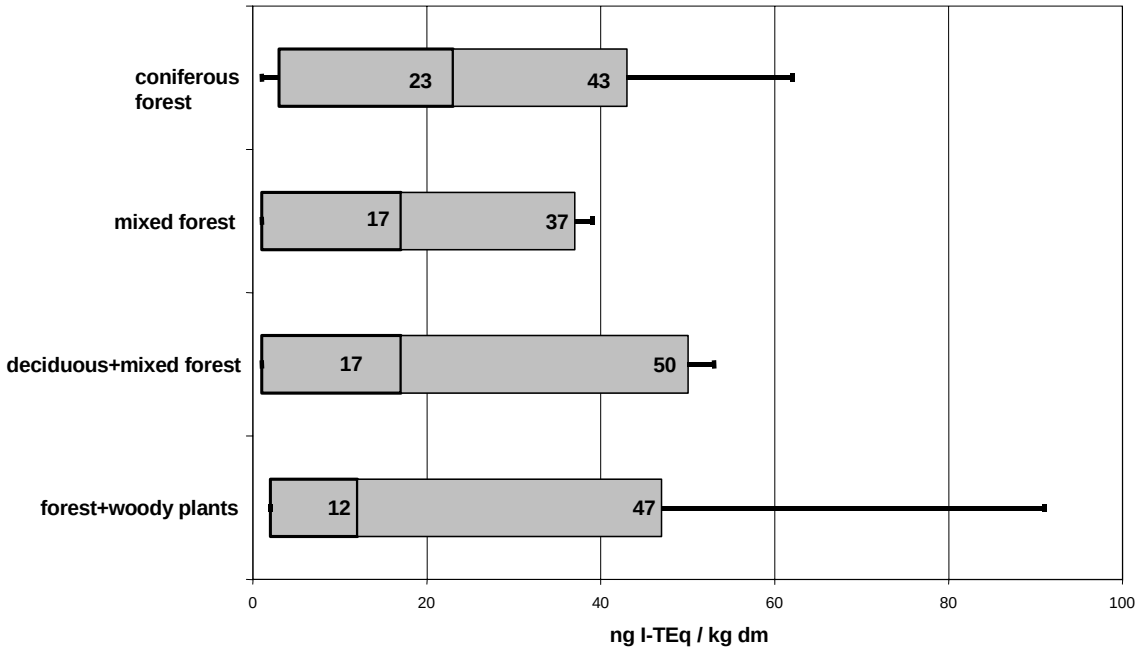

Figure 7: PCDD/PCDFcontents (I-TEq without inclusion of the detection limit) of organic layers without special impact depending on the use

The analytical results for PCDD/PCDF in the database are related to dry substance mass. Due to the essentially deviating density of organic layers, however, they represent an essentially bigger sampling area or a bigger sampling volume per mass unit and thus bigger potential pollutant input units as compared with mineral topsoils. To make organic layers and topsoils comparable the soil density may be used to obtain data related to volume and area. Strictly speaking, the compactness should be used to come to real values “related to the input areas”. However, soil densities (g/cm³) and detailed depths of withdrawal are indicated only for a few samples of layers (n=11) in the database DIOXINS. An average density of approx. 0.17 g/cm³ with the average sampling depth being about 4 cm results from them.

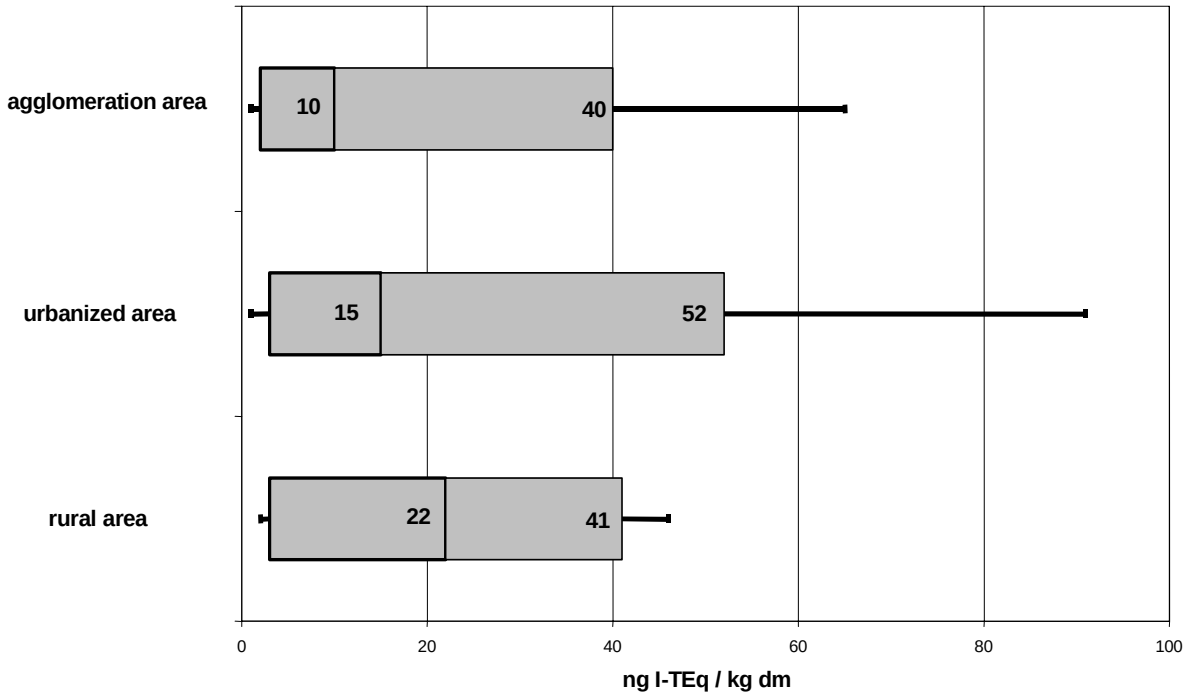

Figure 8: PCDD/PCDF contents (I-TEq without inclusion of the detection limit) of organic layers without special impact depending on the type of area

4.5.3 Mineral topsoil (A - horizons)

A comprehensive data set is available for mineral topsoils as compared with organic layers. They were classed under A horizons in a uniform way hereinafter; further, pedologically differentiated designations of the horizons were not considered. Table 3 gives a survey of the data volume relating to this subcompartment. The use partly given in greater detail is summarized as represented in Table 4. Thereby, evaluations related to use were carried out according to these conglomerated as well as to detailed uses.

Altogether 570 samples are available for this subcompartment with the uses “arable land” (n=64), “grassland” (n=281) and “forest” (n=114) dominating. Apart from the evaluation according to the type of area (BFLR type) and use of soil a classification was effected also according to the year of sampling, depth of withdrawal (horizon) and humus content. The present samples cover the period between 1985 and 1999.

Table 3: Measuring program mineral Topsoils

Institution	Name of the measuring program	Sampling	Use of soil
Bayerisches Geologisches Landesamt	Organ. problematic substances (PCB and PCDD/F) in Bavarian soil	1989/1990	Arable land, grassland, forest, parks and public gardens, sport /leisure grounds
Bayerisches Geologisches Landesamt	Permanent soil monitoring Bavaria	1999	Forest, grassland
Freie Hansestadt Bremen - Senator für Bau und Umwelt	Measuring program for investigating the dioxin contamination in the municipality Bremen	1992/1993	Arable land, grassland, sport /leisure grounds
Freie und Hansestadt Hamburg, Behörde für Arbeit, Gesundh. und Soziales	Investigation relating to the transfer of dioxins and furans soil/feedingstuffs	---	---
Hessisches Landesamt für Umwelt und Geologie	Permanent soil monitoring program of the Land of Hesse	1992-1997	Grassland, forest, arable land
Landesamt für Natur und Umwelt des Landes Schleswig – Holstein	Soil load cadastre Schleswig-Holstein	1994	Arable land, grassland
Landesanstalt für Umweltschutz Baden-Württemberg	Accompanying program for ambient air measurement	1992	Arable land, parks and public gardens, fruit trees
Landesanstalt für Umweltschutz Baden-Württemberg	Sampling on children's playgrounds	1990/1991	Children's playgrounds
Landesanstalt für Umweltschutz Baden-Württemberg	Permanent soil monitoring I, 1st sampling	1985/1986	Arable land, grassland, forest
Landesanstalt für Umweltschutz Baden-Württemberg	Soil expertise for the 2nd hazardous waste incineration plant BW	1991	Arable land, grassland, forest children's playgrounds
Landesanstalt für Umweltschutz Baden-Württemberg	Investigation of the urban area of Heidelberg for dioxins	1992	Arable land, grassland, gardens and small gardens, forest
Landesanstalt für Umweltschutz Baden-Württemberg	Incident composting plant Wieblingen	1991	Grassland, gardens and small gardens
Landesanstalt für Umweltschutz Baden-Württemberg	Environmental study cupola	1992/1994	Grassland, children's playgrounds
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin Sport and playgrounds	1991-1993	Children's playgrounds, grassland sport and leisure grounds, gardens and small gardens
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxins Flood land	1991-1993	Grassland, parks and public gardens, arable land
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxins Agriculture	1988/1990	Parks and public gardens, arable land
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxins Forest soil	1987–1989	Forest
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxins/SM-load Friesenheim Island	1988/1989	Gardens and small gardens
Landesanstalt für Umweltschutz Baden-Württemberg	Small gardens Friesenheim Island	1988/1989	Arable land, gardens and small gardens, production sites, stockyards
Landesanstalt für Umweltschutz Baden-Württemberg	Surroundings of a car shredder	1991	Gardens and small gardens

Institution	Name of the measuring program	Sampling	Use of soil
Landesanstalt für Umweltschutz Baden-Württemberg	Investigations at Enz meadows	1994	Grassland
Landesumweltamt Brandenburg	Investigations related to transfer in Eisenhuettenstadt region	1993	Grassland, children's playgrounds
University Bayreuth	Fertilization with sewage sludge – input of PCDD/F, PCB into the food chain	1990	Arable land, grassland
Federal Environmental Agency – Dioxin laboratory Langen	Measuring system new Federal Laender	1997	Arable land, grassland, forest

The PCDD/F contents in topsoils without special impact are below 26 ng I-TEq/kg dm (90 p.) or 10 ng I-TEq/kg dm (median). The uses were combined to superior uses as represented in Table 4.

Table 4: Summary of soil uses

Superior soil use	Detailed soil use
arable land	arable land fodder plants cereals maize fallow land vegetable root crops
grassland	permanent grassland meadow pasture lawn and meadows parks and public gardens
forest	deciduous forest coniferous forest mixed forest forest and woody plants

Fig. 9 gives the statistical indicators. Accordingly, PCDD/PCDF contents are below 11 ng I-TEq/kg dm (90 p.) for mineral topsoils of grassland and arable land or about 20 ng I-TEq/kg dm (90. P) for forest. The medians are below 5 ng I-TEq/kg dm for all uses.

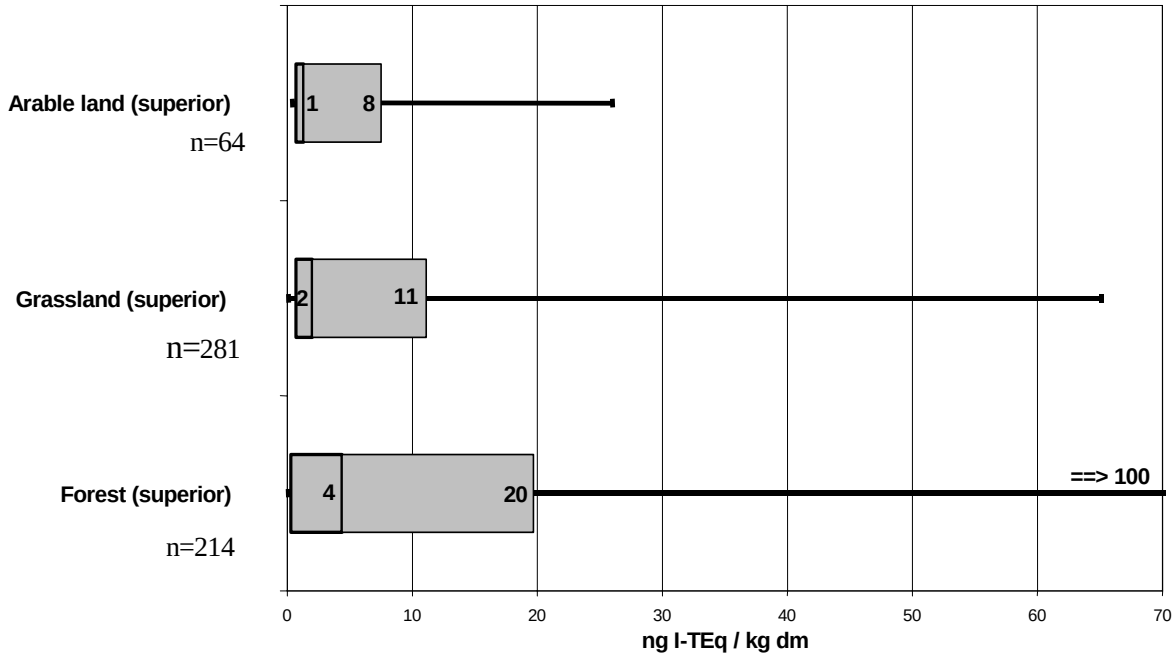

Figure 9: PCDD/PCDF contents (I-TEq without inclusion of the detection limit) in mineral topsoils without special impact depending on the uses

In addition, the data set for forest was evaluated according to detailed uses. The volume of the data set solely for deciduous forest was not sufficient for it. Thus, it was considered together with mixed forest. Samples of deciduous and mixed forests differed significantly from samples of coniferous forests. The third use “forest and woody plants” is a non-specific assignment and therefore not further evaluated in this connection (Fig. 10). The evaluation according to detailed uses of grassland did not show significant differences.

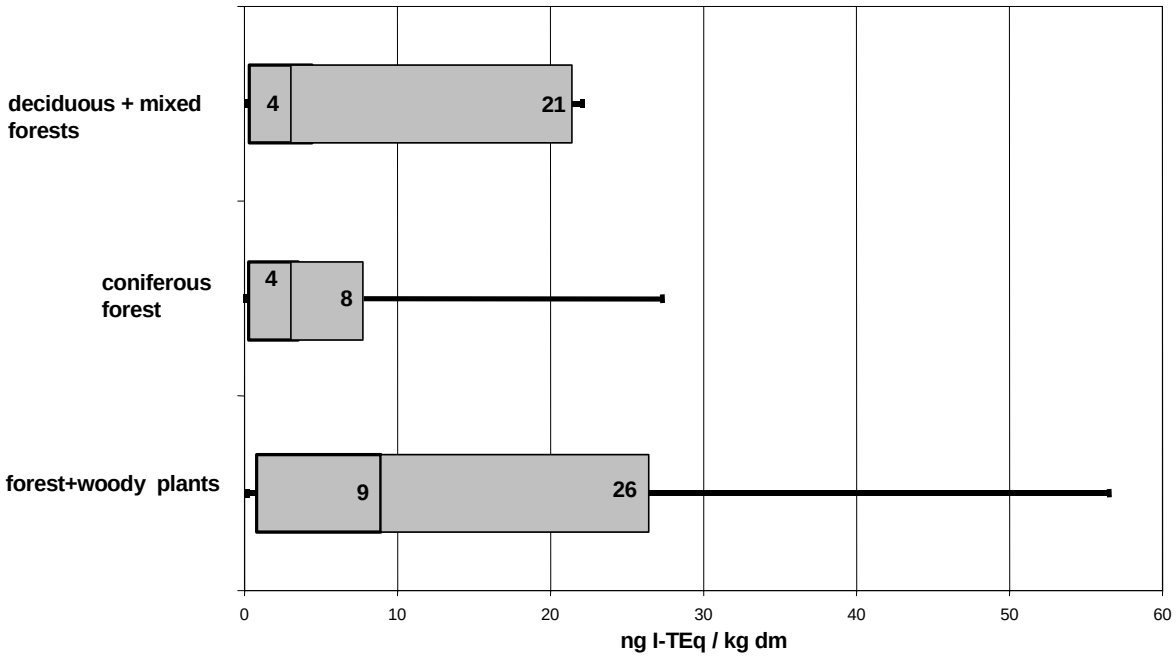

Figure 10: PCDD/PCDF content (I-TEq without inclusion of the detection limit) in mineral topsoils without special impact for various uses of forest

The evaluation of the data set classified according to the **type of area** (BFLR type: agglomeration, urbanized, rural areas) does not give information about significant differences (Fig. 11).

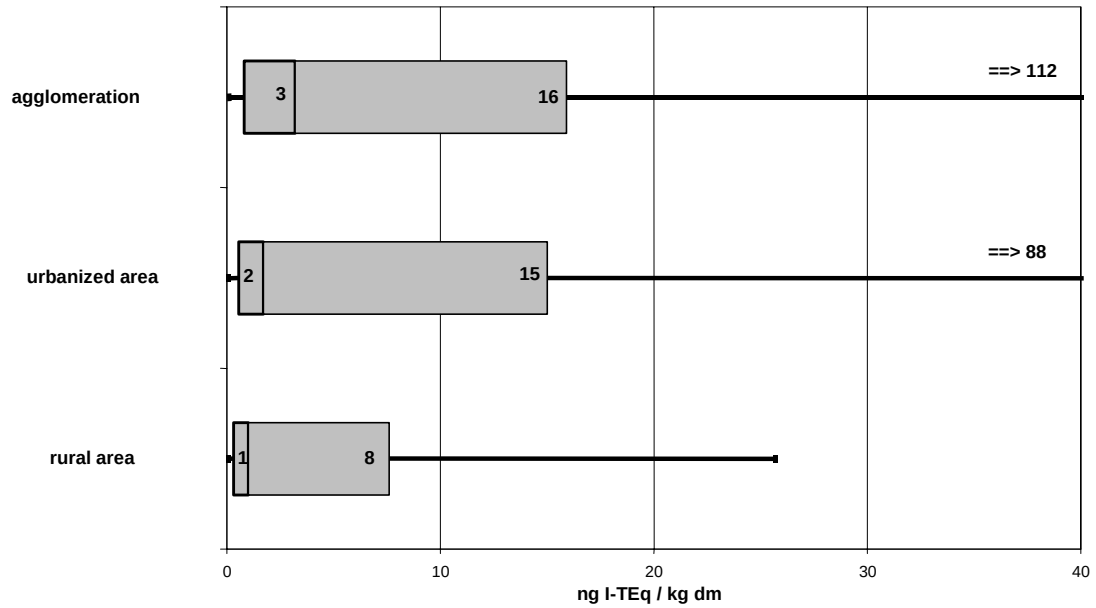

Figure 11: PCDD/PCDF content (I-TEq without inclusion of the detection limit) in mineral topsoils without special impact in various types of areas

4.5.4 B-horizons

Contaminant contents in samples from B-horizons from background areas might indicate potential transfers of contaminants to deeper “soil layers”. Especially from the viewpoint of sustained soil protection significant temporal trends would be of importance.

Altogether only 31 samples without special impact were available for evaluation. That is why a further differentiation was renounced. Thereby, the average PCDD/PCDF contents were below 2 ng I-TEq/kg dm (90 p.).

Table 5: Measuring program B-horizons

Institution	Name of the measuring program
Bayerisches Geologisches Landesamt	Permanent soil monitoring Bavaria
Hessisches Landesamt für Umwelt und Geologie	Permanent soil monitoring program of the Land of Hesse
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin forest soils
Landesamt für Natur und Umwelt des Landes Schleswig - Holstein	Soil load continuons Schleswig-Holstein
Federal Environmental Agency – Dioxin laboratory Langen	Measuring system new Federal Laender

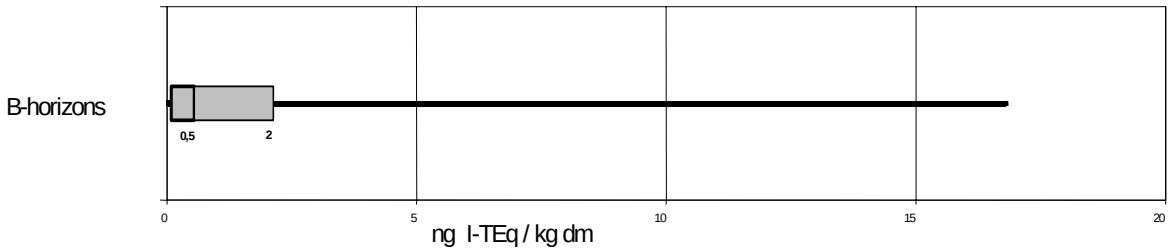

Figure 12: PCDD/PCDF content (ng I-TEq/kg dm) in B-horizons without special impact

4.5.5 Summary Terrestrial soil without special impact

The evaluation carried out is based on the consideration of individual characteristic input parameters from the database DIOXINS. Though it was possible to detect significant differences as regards the investigated parameters (deciduous/mixed forests and coniferous forest) in individual cases the data set altogether does not allow a differentiated evaluation of the PCDD/PCDF contents. That is why the compartment “Terrestrial soil” may be considered inter-parametrically.

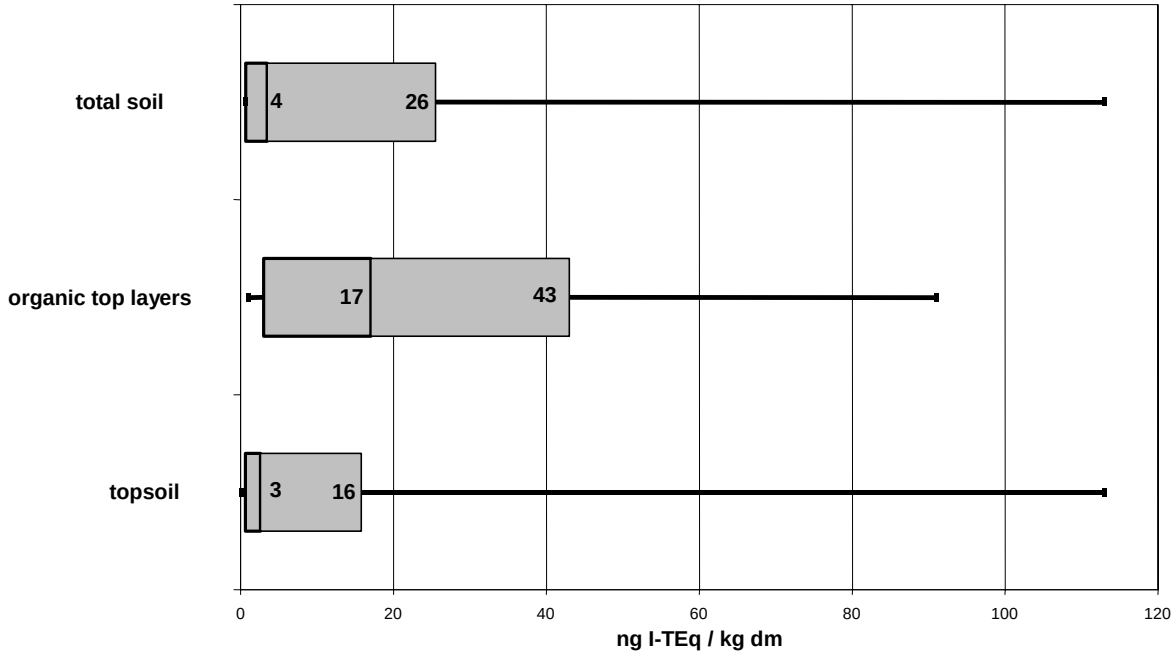

Figure 13: PCDD/PCDF content (ng I-TEq/kg dm) without inclusion of the detection limit in terrestrial soil

The contamination of soils by PCDD/PCDF in ng I-TEq/kg dm (without inclusion of the detection limit) is always below 20 ng I-TEq/kg dm related to the median contents; this involves also the organic layers. Taking the 90 percentile as criterion it is below 30 ng I-TEq/kg dm for any soil; hereby topsoils are below 20 ng I-TEq/kg dm, however, the organic layers reach nearly 50 ng I-TEq/kg dm (Fig. 13).

The Federal Soil Protection and Contaminated Sites Ordinance (BbodSchV) gives an action level of 100 ng I-TEq/kg dm (children's playgrounds), 1.000 ng I-TEq/kg dm (residential areas, parks and leisure grounds) and 10.000 ng I-TEq/kg dm (industrial and commercial properties) (without information relating to the inclusion of the detection limit) for the soil-human health pathway. The contents represented in Fig. 13 (median, 90 p.) are below these measure values.

4.6 Summary compartment Soil

The results obtained by this evaluation may be compared with a report (LABO, 1998) prepared by the LAENDER working group on soil protection. The PCDD/PCDF concentrations in soil given therein, classified according to type and Federal Land, are summed up in Table 6.

Though LABO classificates the soils in a different way than the database DIOXINS there are similar results. With regard to the evaluation by LABO and this evaluation in the case of soils a classification into urban/rural or rural area/urbanized area/agglomeration seems to be not very appropriate.

Table 6: PCDD/PCDF concentrations in soil, classified by Federal Laender (LABO 1998);

Federal Land	n	50 percentile (ng TEq/kg)	90 percentile (ng TEq/kg)
Background values for Baden-Wuerttemberg			
Arable land, topsoil, type III		1.0	5.0
Grassland, topsoil, rural area	50	3.7	36.6
Forest, layer, rural area	20	16.7	53.0
Background values for Bavaria			
Arable land, grassland, topsoil, type II/III	115	0.2	1.1
Forest, topsoil, type II/III	41	0.3	3.3
Forest, layer, type II/III	50	4.6	30.0
Background value Brandenburg			
Arable land, topsoil, type 0	16	1.0	2.0
Grassland, topsoil, type 0	11	2.0	2.0
Forest, layer, type 0	14	24.0	49.0
Background values Hesse			
Topsoils without differentiation of use, yet involving arable land + grassland + forest topsoil+forest-layer, type 0	72	11.3	46.1
Topsoils without differentiation of use, yet all of them Ah, type 0	52	5.0	22.0
Arable land, topsoil, type 0	23	1.0	1.3
Background values Lower Saxony			
Arable land, topsoils, urban	14	2.9	3.7
Arable land, topsoils, rural	45	1.4	2.6
Arable land, topsoils, rural	18	3.8	4.9
Background values North Rhine-Westphalia			
Grassland, topsoils, type 0	157	5.0	8.9
Grassland, topsoil, high settlement density	10	7.4	18.0
Grassland, topsoil, average settlement density	53	6.3	11.0
Grassland, topsoil, low settlement density	112	4.7	7.9
Background values Saarland			
Topsoils, type 0	69	2.3	9.4
Topsoils, compaction area	34	3.5	12.8
Topsoils, fringe areas, compaction area	22	1.8	3.5
Topsoils, rural	13	1.6	2.7
Arable land, topsoil, type 0	15	1.6	2.7
Grassland, topsoil, type 0	14	2.0	10.5
Forest, topsoil, type 0	37	3.5	10.4
Background values Schleswig-Holstein			
Topsoils, type 0	90	0.5	1.8
Background values Thuringia			
Without differentiation of use Topsoils, type 0	17	1.1	3.1
Arable land, topsoils, type 0	12	1.1	2.1

Typ 0 = not specificd, Typ II = urban area, Typ III = rural area

5 COMPARTMENT AIR – AMBIENT AIR

5.1 Introduction

The pathway air is attached major importance in distributing PCDD/PCDF in environment. So, this compartment forms the connection between source (e.g. incineration) and compartments such as feedingstuffs or biota which, on their turn, represent an important pathway of input into the food chain, thus ultimately having a share in the impact on humans. That is why air pollution gives information on a type of “unavoidable basic impact” in a location. In addition, the time of sampling gives a defined reference and thus the exact time when a potential impact occurred. The present chapter deals with the compartment Ambient air.

5.2 Data inventory and quality

A survey of parameters which may be put into the database DIOXINS for a more detailed description of the samples from the present compartment is reflected in Fig. 14. Parameters which may be relevant to the evaluation (temporal, spatial, impact) basically required are pointed out hereby. The possibility of evaluating these parameters, however, presupposes a sufficient number of inputs into the database. If this was given it will be also shown.

Considering the documentation of individual input parameters, first of all, the following main possibilities of evaluation for the compartment Air – ambient air may be derived:

- gaseous phase/ particle phase separation
- year of sampling
- date of sampling (seasonal trend)
- type of area (rural, urbanized, agglomeration areas)
- special impact (y/n)

Table 7 gives a survey of the actual data inventory for this compartment in the database DIOXINS and of characteristic sample properties used in the course of evaluation.

Compartment Air-ambient air:

1. Reason for the investigation:

environmental monitoring (y/n)
permanent monitoring (y/n)
approval procedure (y/n)
investigation of contaminated sites/soil
research project (y/n)
legally prescribed investigation (y/n)
investigation of incident (y/n)
measuring program /name
others

2. Target of investigation:

determination of background contamination rural (y/n)
determination of background contamination urban (y/n)
determination of background contamination agglomeration (y/n))
consideration of transfer/pathways
connection with other samples
connection with other measuring programs

3. Location data:

community indicator/BFLR type ✓
easting/northing value
height amsl
dioxin-relevant industrial area (y/n) - distance
dioxin-relevant former industrial plant (y/n) - type
Transport routes – type distance

4. Sampling:

date beginning ✓
date end ✓
amount of precipitation
average relative humidity
average air temperature
average wind velocity
gaseous phase (y/n) ✓
particle phase (y/n) ✓
sample of suspected site /y/n)
tracking sample (y/n)
sample quantity
sampling according to VDI 3498 (y/n)
other sampling
deviations
transport of samples (transport vessel/conditions/time))

5. Laboratory data

storage time
storage temperature
regulation for the preparation of samples
regulation for analysing
number IS (PCDD/, PCB)
quantity IS
confidence interval
participation in co-operative tests
external/internal quantification
recovery

6. Analytical results: ✓

recovery
detection limit

Figure 14: Potential parameters of input into the database DIOXINS

compartment Air-ambient air(***bold+italic*** = identification of samples with special impact,
bold+✓ = suited for evaluation (number of samples, data quality...)

Tabelle 7: Survey of measuring program compartment Air - ambient air

Nr	Institution	Name of the measuring program	Year of sampling	BFLR type A = agglommeration. S = urbanized area L = rural area - = no data	Special impact **- = no data** **I = industrial plant** **B = not specified** **S = incident** **V = traffic**	Sample matrix G = gaseous phase S = dust phase - = not specified	Number of samples	PCDD/ PCDF	PCB
1	Bayr. Staatsministerium f. Landesentwicklung u. Umweltfragen	Dioxin measurements in the ambient air in Bavaria	1992 – 1993	A, S, L	- , I	G + S	273	X	
2	Hessisches Landesamt für Umwelt und Geologie	Hesse dioxin reference measuring program	1989 – 1998	A, S	-, I	G + S	812	X	X**)
3	Hessisches Landesamt für Umwelt und Geologie	Ecosystem-related biomonitoring program of the region Biebesheim 1992-1994	1992, 1993	A, S	B	S	58	X	
4	Landesumweltamt Nordrhein-Westfalen	Investigations of biota, emissions, depositions and ambient air of the Land of NRW	1991	S	B	G + S	1	X	X
5	Sächsisches Landesamt für Umwelt und Geologie	Ambient air in the urban area of Riesa	1998	A	I	G + S	12	X	
6	Sächsisches Landesamt für Umwelt und Geologie	Ambient air measuring program of the Free State of Saxony	1997, 1998	A	B	G + S	36	X	
7	Thüringer Landesanstalt für Umwelt und Geologie	Chloroaromatics hydrocarbon dioxin measuring programm of Thuringia	1993-1999	S	-, V	G + S	102	X	
8	University Bayreuth	Atmospheric input pathways and behaviour of PCDD, PCDF, PAC in maize crops	1994	L	-	G + S	7	X*)	
9	University Bayreuth	Environmental impact by the atmospheric input of emissions from a waste incineration plant	1992, 1993	S	-	G + S	64	X	
10	University Bayreuth	Investigation of the atmospheric pollution, far-reaching transport, deposition of PCDD/F, coplanar PCB	1995, 1996	A, L	-	G + S	50	X	
11	University Bayreuth	Investigation of the atmospheric input of PCDD/F in fodder plants	1996	S, L	-	G + S	60	X*)	X
12	University Bayreuth	Investigation of emission pathways of polychlorinated dioxins and furans in chemical cleaning plants	1992	L	-	G + S	1	X	
13	Federal Environmental Agency	UBA measuring systems old Federal Laender	1994	L	-	-	32	X	
14	Federal Environmental Agency	UBA measuring systems new Federal Laender	1996, 1997	L	-	-	6	X	

*) only sums of homologues **) PCB 77, PCB 105, PCB 126, PCB 169 in selected samples

5.3 Evaluation procedure

First of all, two basic influencing factors have to be taken into account for the evaluation of ambient air data. Methodical differences in sampling, i.e. considering the gaseous phase and/or the particle phase, may lead to deviating results with regard to the gas/particle distribution. Furthermore, ambient air of PCDD/PCDF are subject to seasonal fluctuations due to seasonally changing sources (domestic fuel etc.) which are, in addition, intensified e.g. by frequently occurring inversion weather conditions. The procedure applied for the evaluation of ambient air data is represented in Fig.15. First of all, the data set was separated into samples without special impact and samples with data relating to special impact. Each of these evaluations was carried out seasonally separated. The evaluations refer only to PCDD/PCDF.

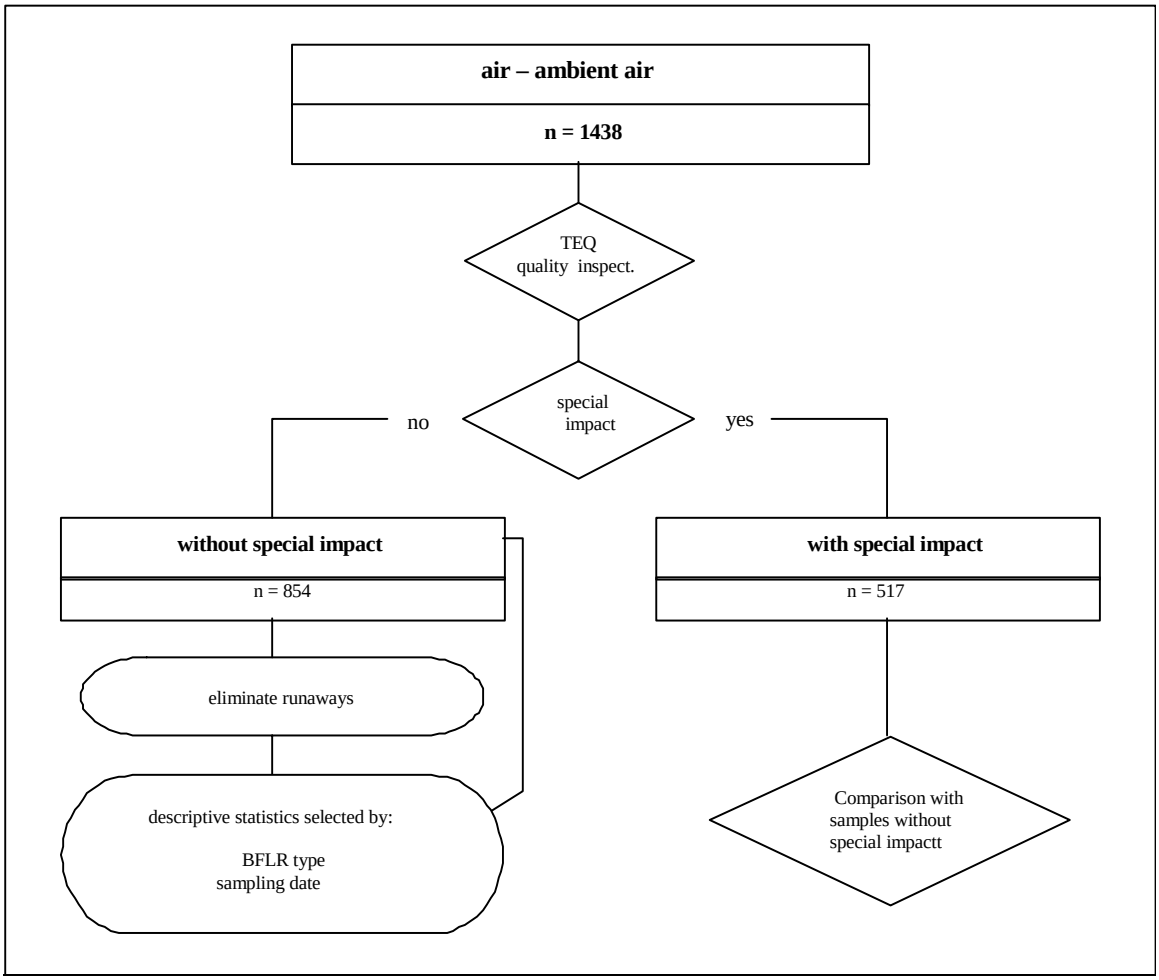

Figure 15: Evaluation scheme compartment Air - ambient air (PCDD/PCDF)

5.4 Runaways

Fig. 16 identifies, first of all, 6 runaways which were eliminated from the data set without special impact before the evaluation Thus, samples with ambient air concentrations below 250 fg I-TEq/m³ were considered in the evaluation.

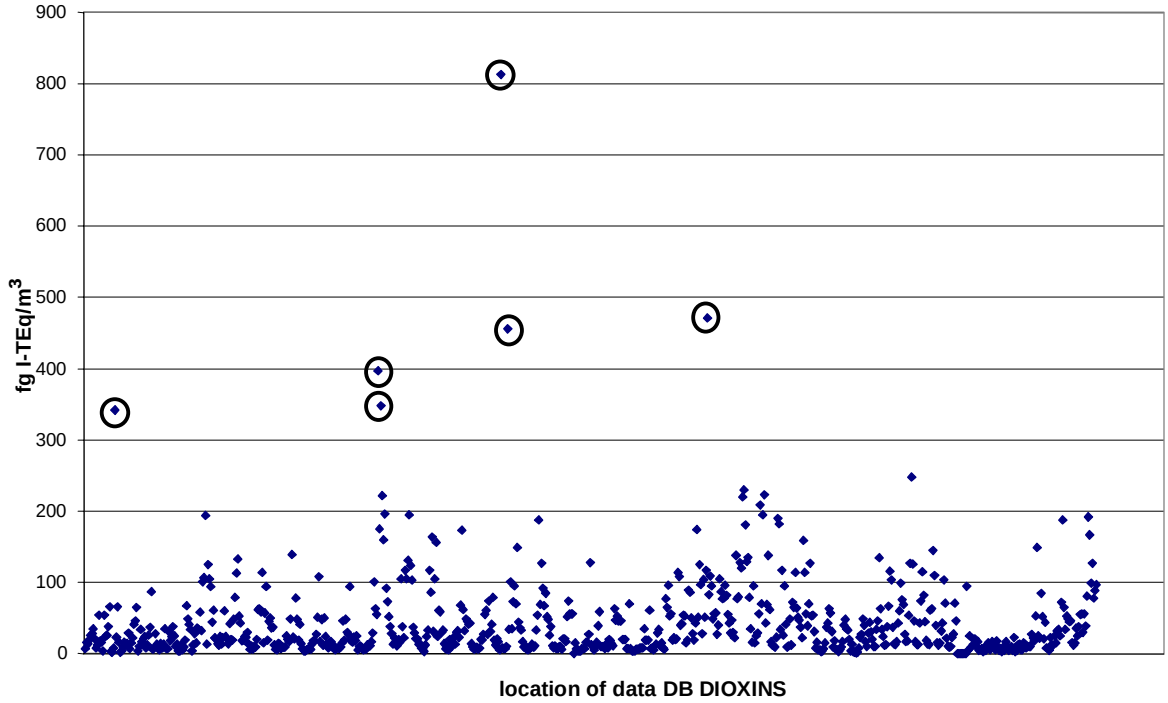

Figure 16: Runaways PCDD/PCDF in the imission data set of the database DIOXINS for samples without impact

5.5 Seasonal variations, general trend and average ambient air concentrations

The changed situation of sources between winter (heating period = October till March) and summer (= April till September) and inversion weather conditions result in seasonally differing situations of impact. That is why this basic variation of the ambient air data has to be considered in all further evaluations.

Hereinafter the seasonal course of the PCDD/PCDF ambient air concentrations from samples without special impact is represented, i.e., on the one hand, as monthly averages (Fig. 17). and, on the other hand, as an arithmetical mean during the heating period (October till March) and out of the heating period (April till September) (Fig. 18). Here, the seasonal variations are clearly visible.

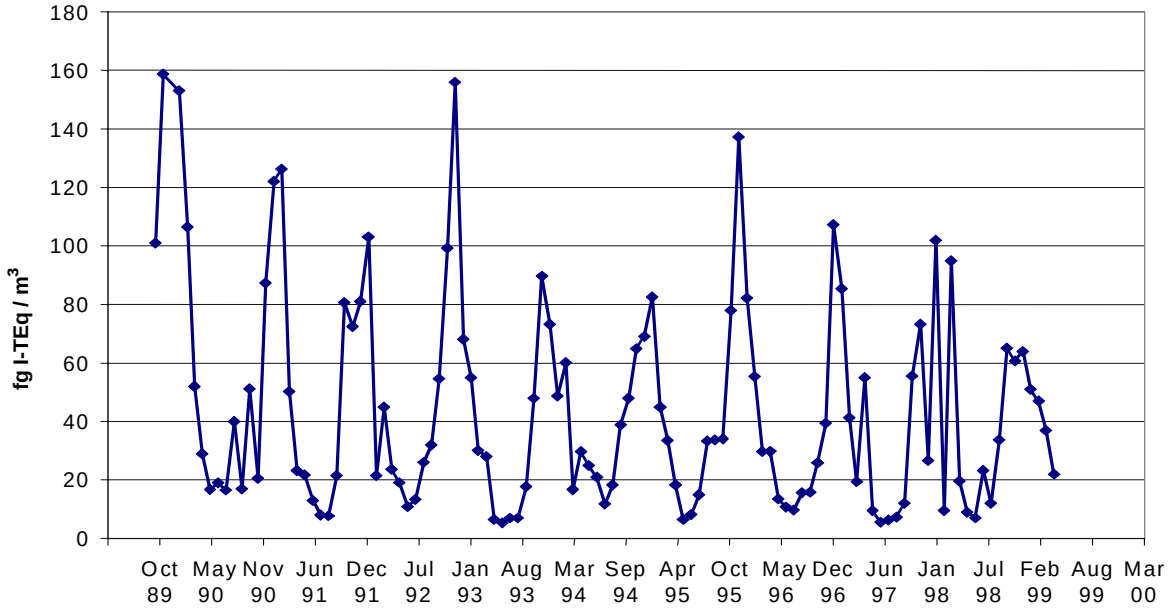

Figure 17: Seasonal variations of the PCDD/PCDF ambient air concentrations from samples without special impact (monthly averages)

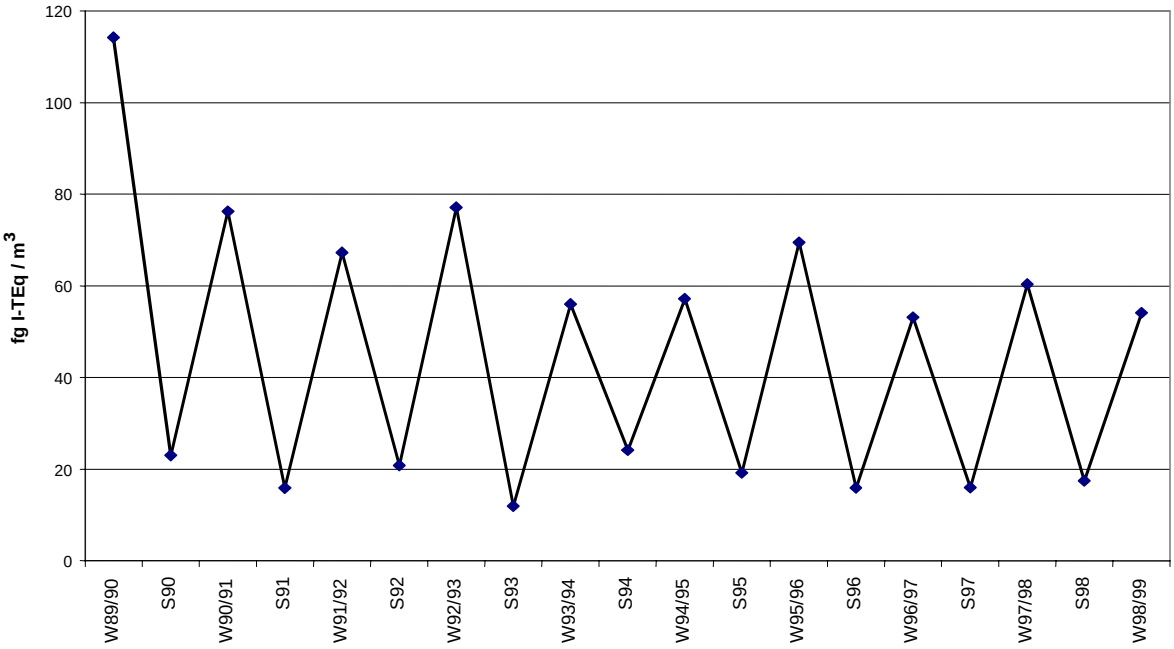

Figure 18: Seasonal variations of the PCDD/PCDF ambient air concentrations from samples without special impact (semiannual averages: W = October–March, S = April-September)

When considering the monthly averages differences between winter and summer maxima occur in the range between a factor of 7 and 30, for the seasonal averages they are approximately between a factor of 2 and 5. Furthermore, a decline of the ambient air concentrations is clearly recognizable notably when considering the winter maxima. Hereby, the peak impacts of the latest data sets were nearly reduced by half as compared with winter 1989/1990.

With the exception of winter 1995/1996 the impacts based on seasonal averages since winter 1993/1994 may be regarded to have remained nearly constant. Thus, this period is suited for the calculation of average ambient air concentrations.

5.6 Methodical differences in ambient air sampling

As regards ambient air sampling the database DIOXINS gathers the methodically recorded ambient air compartments (gas and particle phase, only gaseous phase, only particle phase). For all three variants data sets are available in the database, however for the data sets as of 1994 only for the particle phase and gas with particle phase.Fig. 19 shows the comparison between various sampling strategies (consideration of gaseous and/or particle phase) to determine the ambient air loads.

Here, the whole data set as well as data as of 1994 were included. In the data set considering only the particle phase a measuring program shows partly significantly increased contents as compared with other measuring programs. That is the reason for a separate evaluation of this special data sets.

It is to be expected when considering the gaseous and the particle phases together the highest contents will be obtained. Yet Fig. 19 does not confirm this hypothesis. All three sampling variants show comparable results, in particular after excluding a measuring program with partly increased contents (particle (selec.).

Due to the results of this evaluation hereinafter a differentiation according to the sampling methods applied is not made.

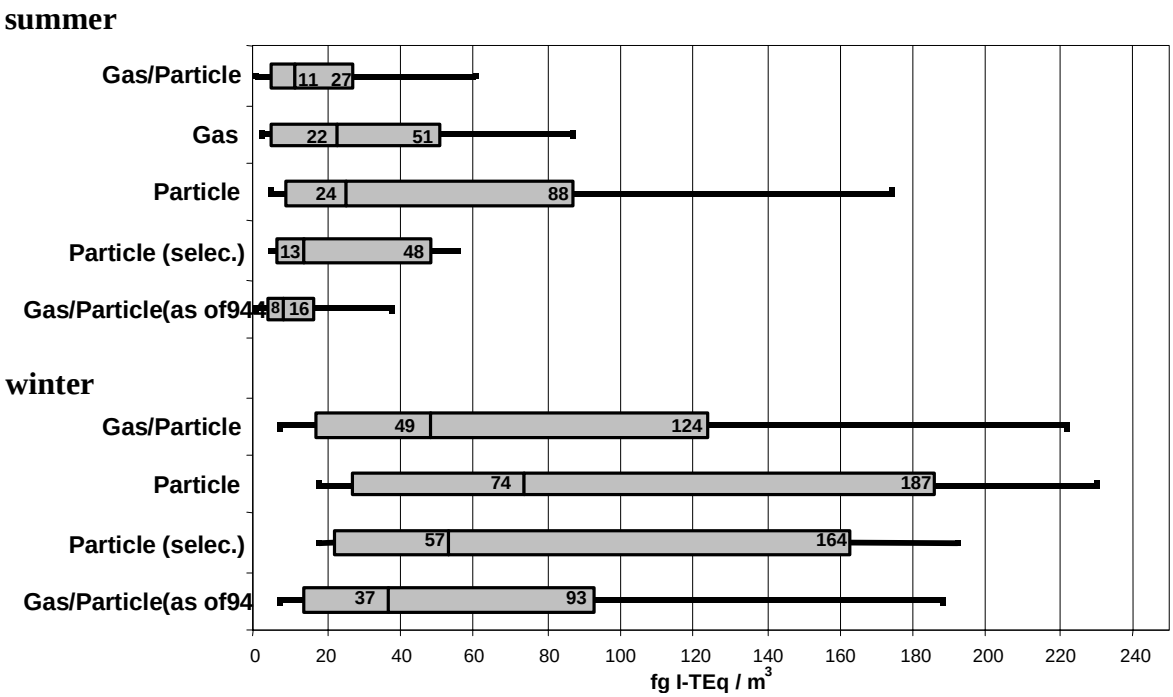

Figure 19: Comparison of various sampling strategies (consideration of gas and/or particle phase) to determine ambient air loads

5.7 Differentiation of the ambient air concentration according to the type of area

Apart from considering temporal trends, seasonal effects and methodical differences the database DIOXINS allows to differentiate ambient air concentrations according to the type of area (rural area, urbanized area, agglomeration) in accordance with the classification by settlement structural area types of the former Federal Institute for Areal Studies and Regional Planning (BFLR 1994). Fig. 20 shows this on the basis of the whole data inventory, Fig.:21 for samples as of 1994.

Thereby, the whole data inventory does not show significant differences between the area types in the summer months (April – October) and the samples of the area type “rural area” in winter. Deviating thereof the area types “agglomeration” and “urbanized area” appear with higher contents in winter (November – March) which are both comparable.

When using data as of 1994 which proved to be nearly constant in considering the temporal development of seasonal minima/maxima this information was confirmed. Only “agglomeration” and rual area” show still signficantly lower contents in summer.

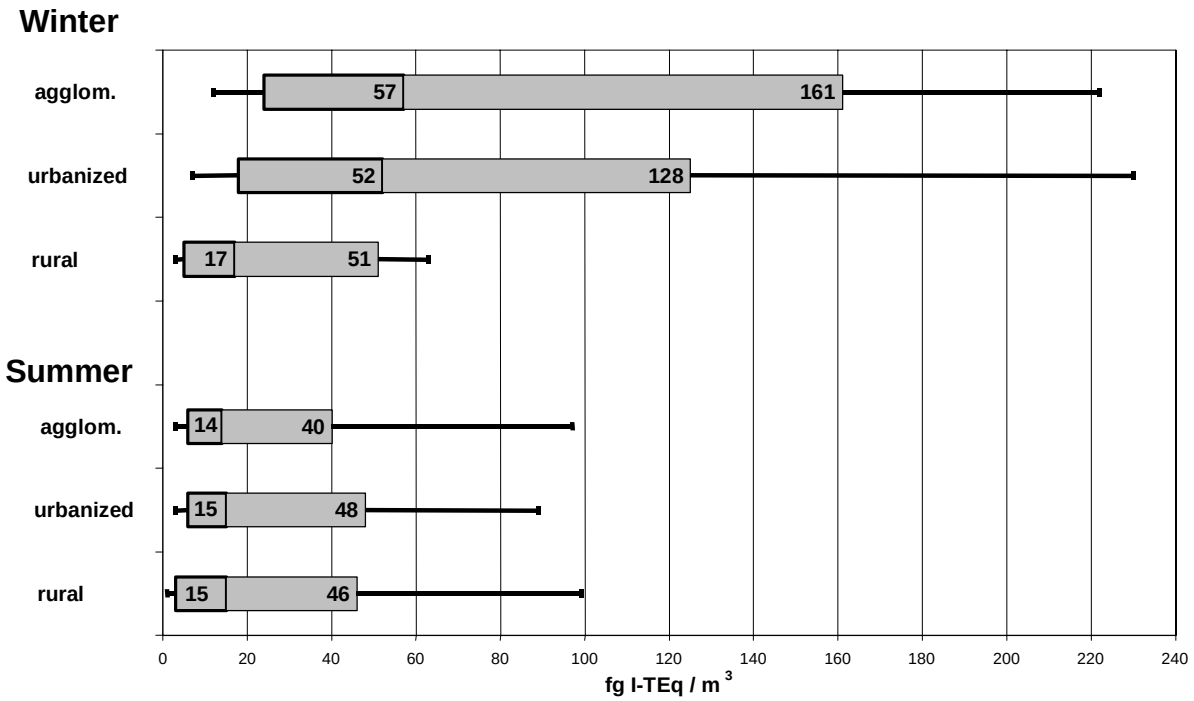

Figure 20: Differentiation of ambient air loads according to area types (agglomeration, urbanized, rural areas)

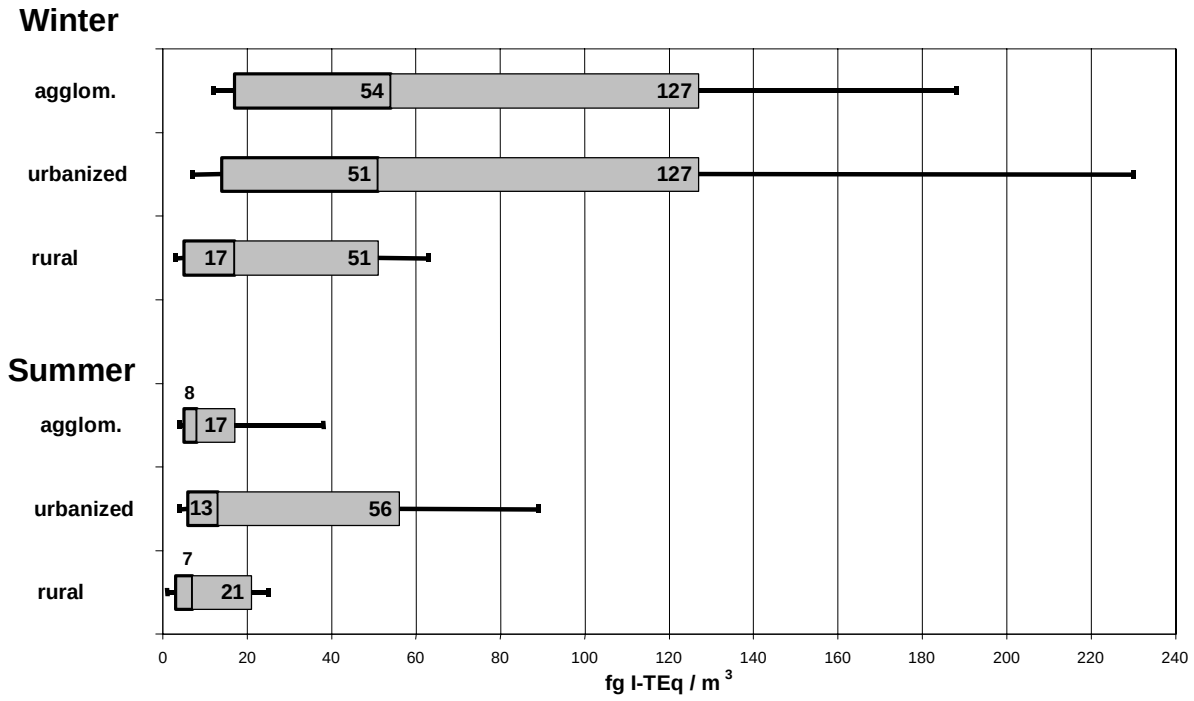

Figure 21: Differentiation of ambient air loads according to area types (agglomeration, urbanized, rural areas, data as of 1994)

Thereof may be derived that in rural areas and areas close to cities without special impacts up to 130 –160 fg I-TEq/m³ (90 p.) have to be expected with the average contents (median) being in the range of 50 fg I-TEq/m³ . The 90 percentile totals approx. 50 fg I-TEq/m³ for rural areas in winter and for all types of areas in summer whereas average contents (median) in the range below 20 fg I-TEQ/m³ may be derived. In addition, the data set for agglomeration and rural areas suggests a further reduction of the ambient air concentrations in summer as of 1994.

5.8 Summary compartment Air ambient air

With altogether 1440 samples the compartment has the third biggest data set in the database DIOXINS after foodstuffs and soil. The data of ambient air samples without special impact prove the seasonal variations of the ambient air concentration and a temporally declining trend of the peak load in winter since the early 90-ies. Since that time the load declined by about half, yet has remained at a nearly constant level since 1994.

Loads up to 160 fg I-TEq/m³ (90 p) have to be expected in areas close to cities and in agglomerations without special impact in winter. The average contents (median) are in the range of 50 fg I-TEq/m³. The value of 50 fg I-TEq/m³ stands for higher contents (90 p.) for rural areas in winter and for all types of areas in summer, whereas average contents (median) in the range below 20 fg I-TEq/m³ may be derived. In addition, the data set for agglomeration and rural areas suggests a further reduction of ambient air concentrations in summer.

6 COMPARTMENT AIR – DEPOSITIONS

6.1 Introduction

Apart from the ambient air concentrations deposition rates are further characteristic parameters for describing air pollution by PCDD/PCDF or its potential transition from the compartment Air e. g. to the compartments Biota (plants) and Soil. Thus, they describe a basic input into these compartments at a given location. In addition they can provide the basis for further estimating inputs into other compartments (e.g. air – dust – wastewater – sewage sludge pathways). As has been already described for ambient air by means of

deposition data the connection between source (e.g. incineration) and input pathway for the food chain may be established.

6.2 Data inventory and quality

Fig. 22 shows a survey of the parameters which were put into the database DIOXINS to describe samples from the present compartment in greater detail.

Parameters relevant to evaluations (temporal, spatial, impacts) required basically are pointed out thereby. However, a possibility of evaluating these parameters requires a sufficient number of inputs into the data base. If this was given it is shown.

With regard to the documentation of the individual input parameters the following possibilities of evaluation may be derived for the compartment Air-depositions:

- type of deposition (total, dry)
- year of sampling
- date of sampling (seasonal trend)
- type of area (rural area, urbanized area, agglomeration)
- special impact (y/n)

Table 8 gives a survey of the data inventory for this compartment in the database DIOXINS and of characteristic properties of samples used in the course of evaluation.

Compartment Air-ambient air:

1. Reason for the investigation:

- environmental monitoring (y/n)
- permanent monitoring (y/n)
- approval procedure (y/n)
- research project (y/n)
- legally prescribed investigation (y/n)
- ***investigation of incident (y/n)***
- measuring program /name
- others

2. Target of investigation:

- ***determination of background contamination rural (y/n)***
- ***determination of background contamination urban (y/n)***
- ***determination of background contamination agglomeration (y/n))***
- consideration of transfer/pathways
- connection with other samples
- connection with other measuring programs

3. Location data:

- ***community indicator/BFLR type*** ✓
- easting/northing value
- height amsl
- dioxin-relevant industrial area (y/n) – distance
- dioxin-relevant former industrial plant (y/n) – type
- transport routes – typ- distance

4. Sampling:

- date beginning ✓
- date end ✓
- amount of precipitation
- average relative humidity
- average air temperature
- average wind velocity
- **dry deposition** /y/n)
- **wet deposition (y/n)**
- **total deposition (y/n)**
- **rainwater (y/n)**
- **crown drip deposition (y/n)**
- **suspected sample (y/n)**
- **tracking sample (y/n)**
- sample quantity
- sampling according to VDI 2119 (y/n)
- other sampling
- number of Bergerhoff vessels
- number of Loebner-Liesegang vessels
- geometry of arrangement
- deviations
- transport of samples (transport vessel/conditions/time))

5. Laboratory data

- storage time
- storage temperature
- regulation for the preparation of samples
- regulation for analysing
- number IS (PCDD/, PCB)
- quantity IS
- confidence interval
- participation in co-operative tests
- external/internal quantification
- recovery

6. **Analytical results:** ✓

- recovery
- detection limit

Figure 22: Potential input parameters database DIOXINS compartment Air - depositions (***bold+italic*** = identification of samples with special impact, **bold+✓** = suited for evaluation (number of samples, data quality ...)

Table 8: Survey of the measuring program compartment Air - depositions

Nr	Institution	Name of the measuring program	Year of sampling	BFLR type A = agglommeration S = urban area L = rural area - = no data	Special impact **- = no data** **I = industrial plant** **B = not specified** **S = indicdent** **V = traffic**	Sample matrix N = wet dep. T = dry dep. G = total dep. - = nnit specified	Proben-anzahl	PCDD/ PCDF	PCB
1	Freie Hansestadt Bremen – Senator für Bau und Umwelt	Measuring program for the investigation of the dioxin contamination in the municipality of Bremen	1992	A	B	G	30	X	
2	Freie und Hansestadt Hamburg, Behörde f. Arbbeit, Gesundh. u. Soziales	Investigation of the transfer of dioxins and furans soil/feedingstuffs	1990, 1991	A	-	G	30	X	
3	Hessisches Landesamt für Umwelt und Geologie	Hesse dioxin reference measuring program	1989–1998	A, S	-, B	G	398	X	
4	Hessisches Landesamt für Umwelt und Geologie	Ecosystem–related biomonitoring program in the region of Biebesheim 1992-1994	1992, 1993	A, S	B	G	36	X	
5	Landesumweltamt Brandenburg	Investigation of transfer in the region of Eisenhuettenstadt	1993	A	I	G	4	X	
6	Landesumweltamt Nordrhiein-Westfalen	Investigation of biota, emissions, depositions and ambient air of the Land of NRW	1991	S	B	G	1	X	X
7	Landesumweltamt Nordrhiein-Westfalen	Investigation of deposition of the Land of NRW	1991, 1993-1999	A	-	G	49	X	X
8	Ministerium für Umwelt und Forsten Rheinland-Pfalz	Deposition measuring program of the Land of Rhineland-Pfalz	1993, 1994	A, S, L	-	G	22	X	
9	Thüringer Landesanstalt für Umwelt und Geologie	Chloraromatics dioxin measuring program Thuringia	1993–1999	S	-, V	T	102	X	
10	Umweltbehörde Hamburg, FA für Umweltuntersuchungen	Investigation of deposition, water and sediments for the Hamburg dioxin reference measuring program	1996	A	-	G	24	X	
11	Federal Environmental Agency	UBA measuring systems old Federal Laender	1994	L	-	-	34	X	
12	Federal Environmental Agency	UBA measuring systems new Federal Laender	1996, 1997	L	-	-	16	X	
13	University Bayreuth	Atmospheric input pathways and behaviour of PCDD, PCDF, PAC in a maize crop	1994	L	-	G	5	X*)	
14	University Bayreuth	Investigation of the atmospheric pollution, far-reaching transport, deposition of PCDD/F, coplanar PCB	1995, 1996	L, A	-	G	101	X	X

*) only sums of homologues

6.3 Evaluation procedure

Potential methodical differences in sampling (wet deposition, dry deposition, total deposition) have to be considered in evaluating the deposition data. As ambient air of PCDD/PCDF depositions are subject to seasonal variations due to seasonally changing situations of the sources (domestic fuel, weather conditions etc.).

The procedure adopted in evaluating the deposition data is represented in Fig. 23. First of all, the data set was divided into samples without special impact and into samples with data on special impact.

The data set without special impact was included in the further evaluation. These evaluations were carried out seasonally separated and refer only to PCDD/PCDF.

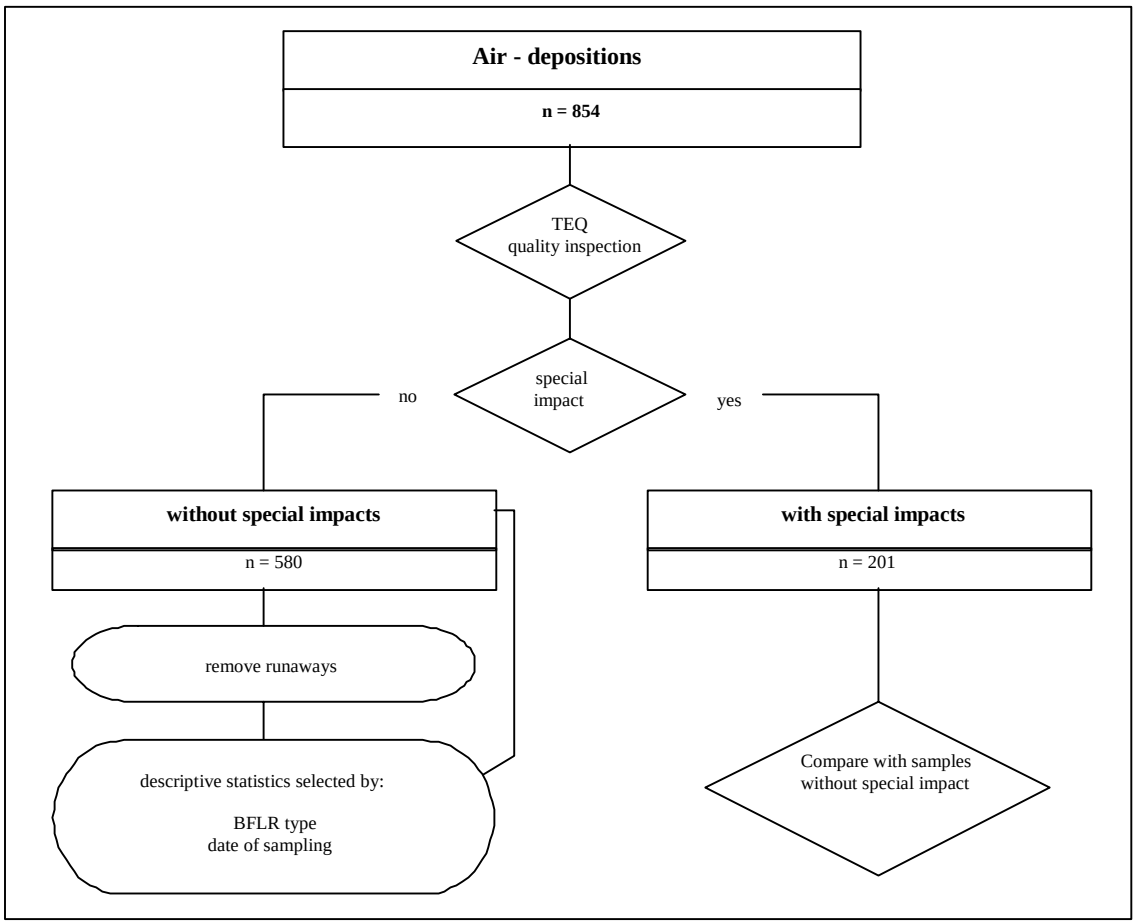

Figure 23: Evaluation scheme compartment Air - depositions (PCDD/PCDF)

6.4 Runaways

Fig. 24, first of all, identifies 6 runaways eliminated from the data set without special impact before carrying out the evaluation. Thus, samples with deposition concentrations below 150 pg I-TEq/m² and day, for the majority of samples below 50 pg I-TEq/m² and day, were available for evaluation.

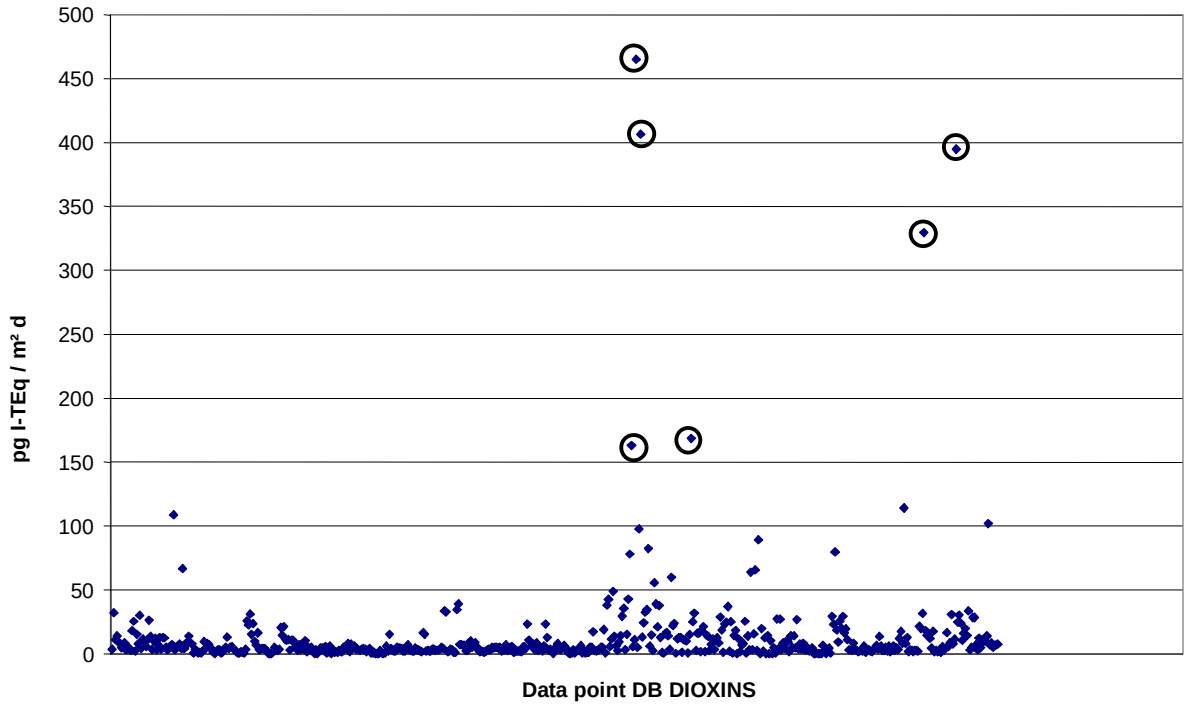

Figure 24: Runaways PCDD/PCDF in the deposition data set of the database DIOXINS for samples without impact

6.5 Seasonal variations, general trend and average deposition concentrations

As mentioned the chapter on ambient air changed situations of sources between winter (heating period) and summer result in seasonally various impacts. They should then be also reflected in the deposition impacts. Hereinafter, the seasonal course of the PCDD/PCDF deposition concentrations from samples without special impact is represented.

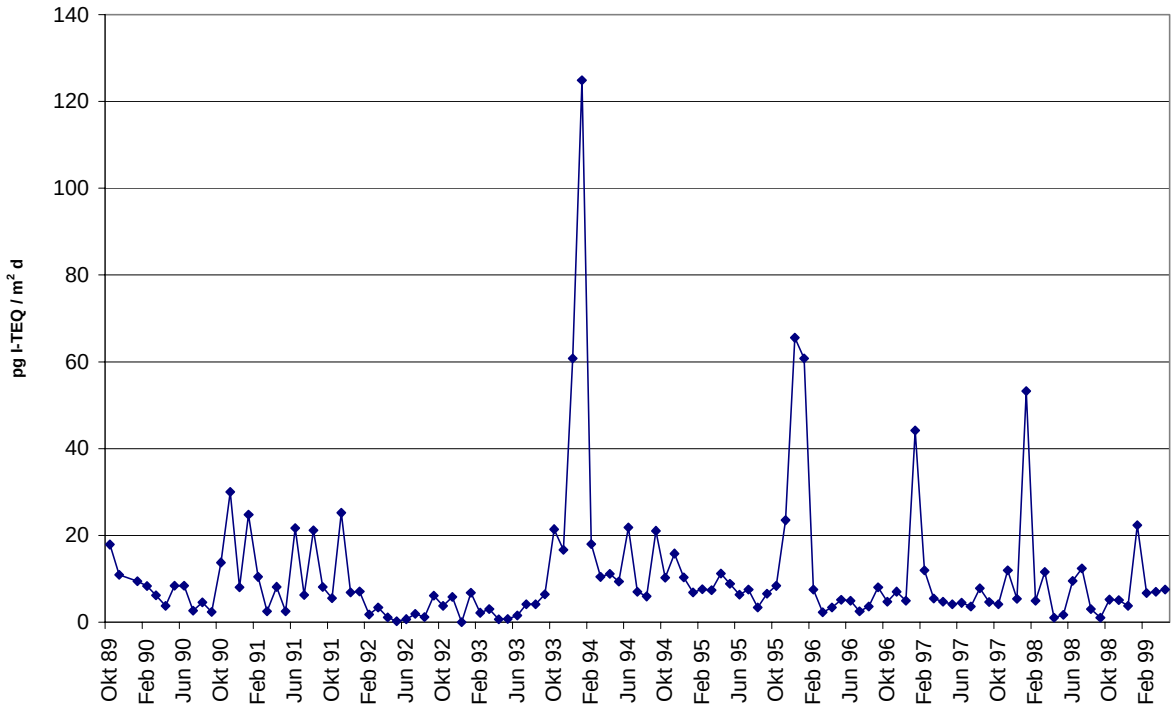

Figure 25: Seasonal variations of the PCDD/PCDF deposition concentrations from samples without special impact (monthly averages)

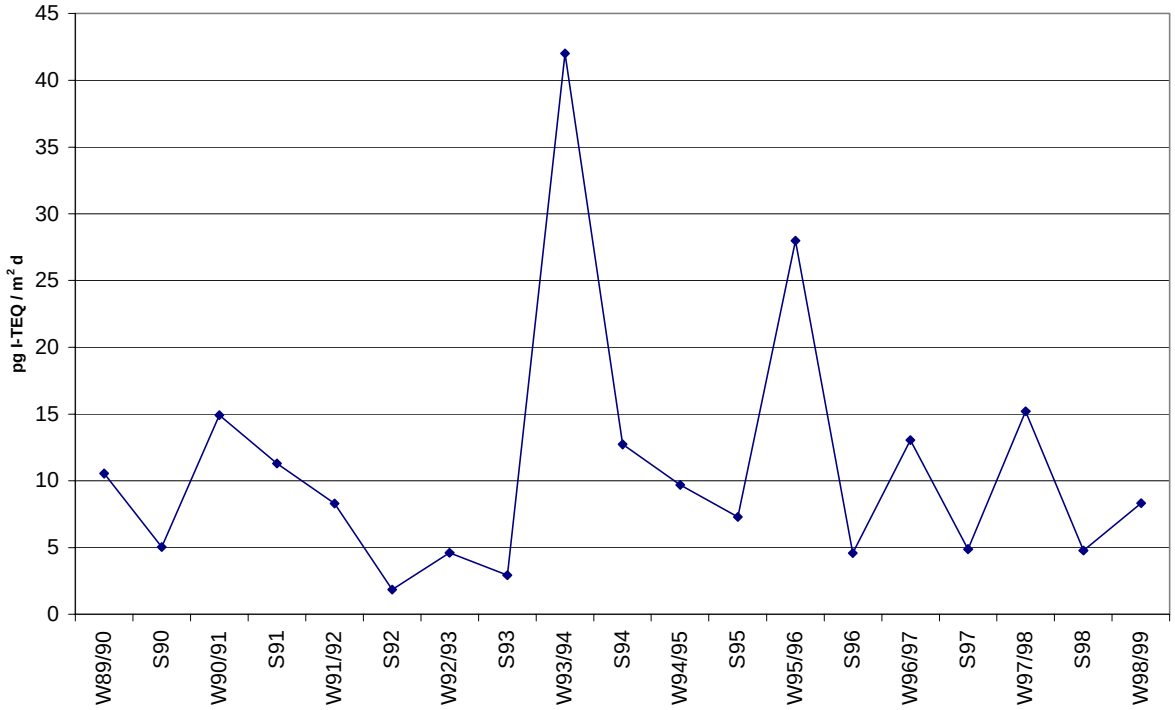

Figure 26: Seasonal variations of PCDD/PCDF deposition concentrations from samples without special impact (semiannual averages: W = October–March, S = April–September)

Fig. 26 shows averages during the heating period (October till March = winter (W)) and out of the heating period (April till September = summer (S)). However, in contrast with the ambient air data these variations have not been clearly visible before winter 1994. The peak impacts to be detected as of 1994 show a similar trend as the ambient air data.

Considering the monthly averages differences in the range of a factor of 5 till 15 between winter and summer maxima arise, for seasonal averages they amount approximately to a factor between 1.5 and 10. A decline of the deposition concentrations, primarily in considering the winter maxima, may be clearly recognized. Thereby, the peak impacts of the latest data sets of the monthly averages declined by about a factor of five as compared with winter 1993/1994. Excluding the extreme values of winter 1993/94 and 1995/96 from the total consideration (winter 1989/90 till winter 1998/99) the values are comparatively constant.

When considering seasonal averages only from 1997 on there may be spoken of an approximately constant level, thus this period 1997 – 1998 is suited for assessing the present average deposition concentrations.

6.6 Methodical differences in deposition sampling

As to deposition sampling the database DIOXINS comprises the methodically covered compartments (total deposition, wet deposition, dry deposition, crown drip deposition, snowfall). Primarily data on the total depositions are available, thus the data set should be altogether well comparable in this respect (all with the Bergerhoff gauger).

6.7 Differentiation of the deposition concentration by the type of area

Apart from considering temporal trends, seasonal effects and methodical differences the database DIOXINS provides the possibility to differentiate deposition concentration by the type of area (rural area, urbanized area, agglomeration) in accordance with the classification by settlement structural area types of the BFLR. Fig. 27 shows them on the basis of the whole data inventory, Fig. 28 for samples between 1996 and 1998.

Thereby, the whole data inventory do not show significant differences between the area types in the summer half-year and “rural area” type in winter, with the exception of the increased 90 p. of the “urbanized area” type.

For the winter samples the “agglomeration” is of the size of the “urbanized area” in summer, the “urbanized area” shows increased contents as 90 percentile also in winter. In summer the 90 P. is below 10 pg in agglom. and rural areal and below 25 pg in urbanized area. For the winter half-year contents (90 p.) below about 20 pg I-TEq/m² d (urbanized area 43 pg I-TEq/m² d) may be expected. Considering the medians, in general, values below 10 pg I-TEq/m² d, in summer even below 5 pg I-TEq/m² d, are to be expected.

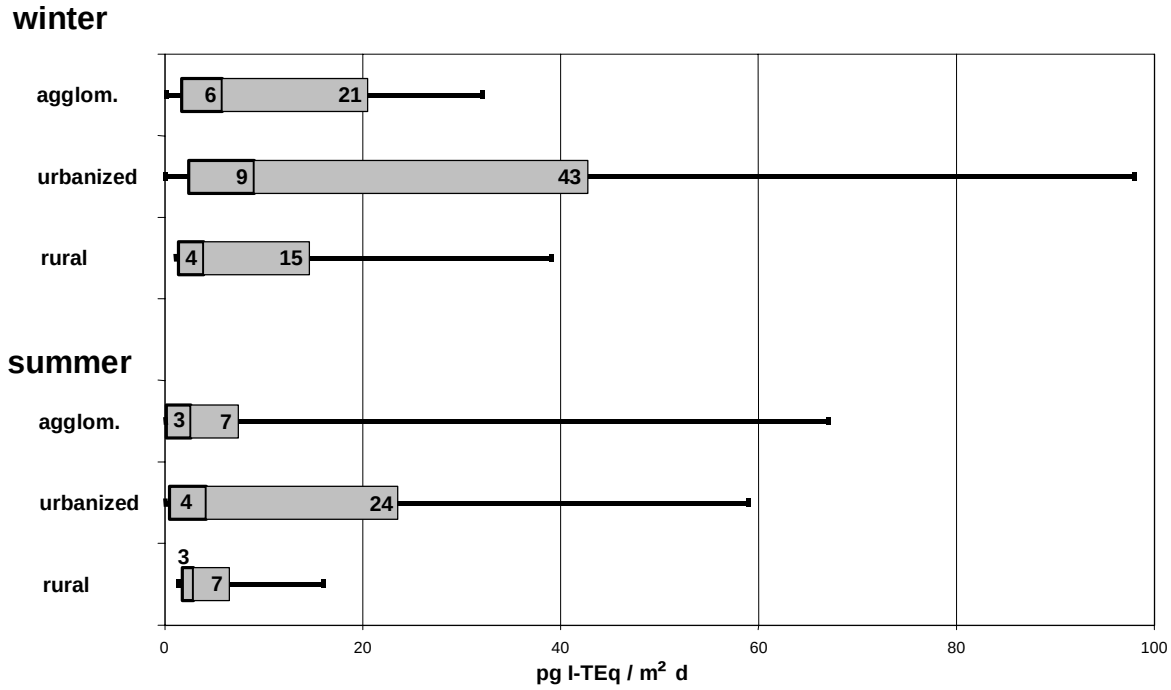

Figure 27: Differentiation of deposition impact by area types (agglomeration, urbanized area, rural area, whole data inventory)

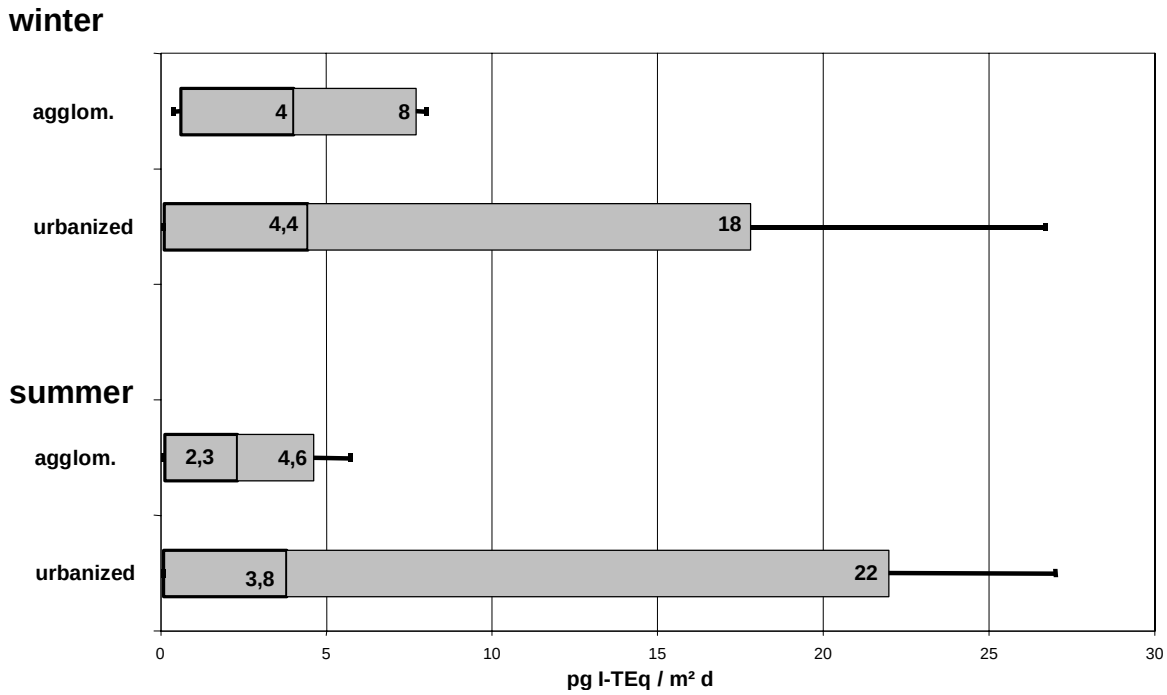

Figure 28: Differentiation of the deposition impact by area types (agglomeration, urbanized area, data 1996-1998)

When using data between 1996 and 1998 the data set is only sufficient for considering the agglomeration and the urbanized area. The increased contents in the “urbanized area” type as compared with the “agglomeration” type are confirmed.

It is possible to derive – without special impact – concentrations in deposition equally below 10 pg I-TEq/m² (in summer even below 5 pg I-TEq/m² d (90. p.). In agglomeration areas and in areas close to cities the results are approximately below 22 pg I-TEq/m² d (90 P.) throughout the year. Considering the medians yearly contents below 5 pg I-TEq/m² d are to be expected.

6.8 Summary compartment Air- depositions

The data of deposition samples without special impact document the seasonal variations as they were detected also in ambient air concentrations in recent data sets. They show a temporally declining trend of the peak impacts in winter. Thereby, the peak impacts of the latest data sets of the monthly averages declined approximately by a factor of five as compared with winter 1993/1994.

Excluding the extreme values of winter 1993/94 and 1995/96 from the whole consideration (winter 1989/90 until winter 1998/99) the values are, however, comparatively constant.

With the exception of “urbanized areas” with impacts (90 p.) below 10 pg I-TEq/m² d special impacts are not to be expected (urbanized areas <20 pg I-TEq/m² d, in winter <40 pg I-TEq/m² d).

7. COMPARTMENT WILD PLANTS AND ANIMALS (BIOTA)

7.1 Introduction

The compartment Biota comprises wild plant and animal organisms as well as organisms raised under standardized conditions and then released into open land in the course of biomonitoring procedures.

Thus, the pollutant contents detected in this compartment are, on the one hand, connected with the compartments Foodstuffs or Feedingstuffs and thus directly with the input into the human food chain (pathway: plant/animal – feedingstuffs/foodstuffs – impact on humans). That is why they may form the basis for further assessments of inputs into these further compartments. In this function the compartment Biota according to source and first spreading steps (e.g. air, water...) is an important input pathway into the human food chain.

Furthermore, through their “biomonitoring function” organisms give information on local pollutant contents in the individual environmental compartments where the organisms were taken from. For instance, fish is in a close connction with waters – water plants or sediment loads – depending on food and living conditions. Plant bioindicators are notably in connection with air pollution, and in this function, are to be regarded as a further supplement to the chapters ambient air and depositions.

Pollutant contents in biota describe the pollutant contents actually taken over from other compartments. Thus, they may be used for first assessments of the transfer rates, bioconcentration factors etc.

7.2 Data inventory and quality

Parameters put into the database DIOXINS to describe samples from the present compartment in greater detail are listed in Fig. 29. Parameters which may be relevant to the evaluations (temporal, spatial, impacts) basically required are pointed out thereby. The possibility of evaluating these parameters, however, presupposes a sufficient number of inputs into the database. If this was given it is shown.

The following main possibilities of evaluation may be, first of all, derived for the compartment Biota with regard to the documentation of individual input parameters:

- biota type (plant/animal species)
- year of sampling
- date of sampling (seasonal trend)
- type of area (rural area, urbanized area, agglomeration)
- special impact (y/n)

Compartment Biota:

1. Reason for the investigation:

- environmental monitoring (y/n)
- permanent monitoring (y/n)
- approval procedure (y/n)
- research project (y/n)
- legally prescribed investigation (y/n)
- ***investigation of incident (y/n)***
- measuring program /name
- others

2. Target of investigation:

- ***determination of background contamination rural (y/n)***
- ***determination of background contamination urban (y/n)***
- ***determination of background contamination agglomeration (y/n))***
- consideration of transfer/pathways
- connection with other samples
- connection with other measuring programs

3. Location data:

- ***community indicator/BFLR type*** ✓
- easting/northing value
- height amsl
- dioxin-relevant industrial area (y/n) – distance
- dioxin-relevant former industrial plant (y/n) – type
- **transport routes – type- distance**

4. Sampling:

- **date** ✓
- **plant (y/n)** ✓
- **animal (y/n)**
- **microorganism (y/n)**
- **name**
- **part**
- sex
- **age**
- individual sample (y/n)
- mixed sample (y/n)
- **suspected sample (y/n)**
- tracking sample (y/n)
- sample quantity
- transport of samples (transport vessel/conditions/time))

5. Laboratory data

- storage time
- storage temperature
- regulation for the preparation of samples
- regulation for analysing
- number IS (PCDD/F, PCB)
- quantity IS
- confidence interval
- participation in co-operative tests
- external/internal quantification
- recovery

6. **Analytical results:** ✓

- fresh weight (y/n)
- dry weight (y/n)
- recovery
- detection limit

Figure 29: Potential input parameters database DIOXINS compartment Biota (***bold+italic*** = Identification of samples with special impacts, **bold+✓** = suited for evaluation (number of samples, quality of data...)

Table 9 gives a survey of the actual data inventory for this compartment in the database DIOXINS as well as of characteristic sample properties used in the course of evaluation.

Table:9 Survey of measuring program compartment Biota

Nr.	Institution	**Name of the measuring program**	**Year of sampling**	BFLR type **A = a**gglommeration S = urbanized area L = rural area - = no data	Special impact **- = no data** **I = industrial plant** **B = not specified** **S = incident** **V = traffic**	Sampling matrix F = spruce G = green cabbage W = Welsh ryegrass Gr = grass non-spec. Pf = plant non-spec. Fi = fish Mi = mussels - = not specified	**Number of samples**	PCDD/ PCDF	PCB
1	Bayrisches Landesamt für Umweltschutz	Biomonitoring (Welsh ryegrass, green cabbage) in 6 locations	1996-1997	L, A	-, V	W, G	65	X*)	
2	Bayrisches Landesamt für Umweltschutz	Bioindication of airborne dioxins and furans by means of spruce needles	1992-1998	A, S, L	-	F	163 **)	X**)	
3	Chemisches Landesuntersuchungsamt Oldenburg	Dioxin reference monitoring program	1995-1998	-	-	-	60	X	
4	Freie Hansestadt Bremen – Senator für Bau und Umwelt	Measuring program for the investigation of dioxin contamination in the municipality of Bremen	1993	A	B	G	6	X	X
5	Freie und Hansestadt Hamburg, Behörde f. Arbeit, Gesundh. u. Soziales	R&D biomonitoring of the Elbe-river	1994	A, L	-	Fi ***)	14	X	
6	Hessisches Landesamt für Umwelt und Geologie	Ecosystem-related biomonitoring program in the region of Biebesheim 1992-1994	1992, 1993	A, S	B	F, G	64	X	X
7	Landesumweltamt Brandenburg	Transfer investigation in the area of Eisenhuettenstadt	1993	A	-, I	Gr, Fi	11	X	
8	Landesumweltamt Nordrhein-Westfalen	Investigation of biota, emissions, depositions and ambient air of the Land of NRW	1994	A	I	Gr	1	X	
9	Ministerium für Umwelt, Natur und Forsten des Landes Schleswig-Holstein	Investigation of soils and vegetable for PCDD/PCDF	1994	-	-	Pf	11	X	
10	Niedersächsisches Umweltministerium	Investigation of the pollution of soils, mussels and sediments from the region of Wilhelmshaven	1994	S	I	Mi	13	X	

*) only sums of homologues, no individual congeners, only I-TEq incl ½ detection limit; **) for n = 82 (= 1992–1994) no individual congeners indicated, only I-TEq incl ½ detection limit; ***) bream

7.3 Evaluation procedure

The procedure for evaluating biodata is represented in Fig 30. First of all, the individual species have to be considered separately for the evaluation of biotadata (other accumulation properties, wax content, fat content etc.). In this case, spruce needles. green cabbage and Welsh ryegrass samples offer themselves for a more detailed evaluation. Individual considerations of fish and mussels complete this chapter.

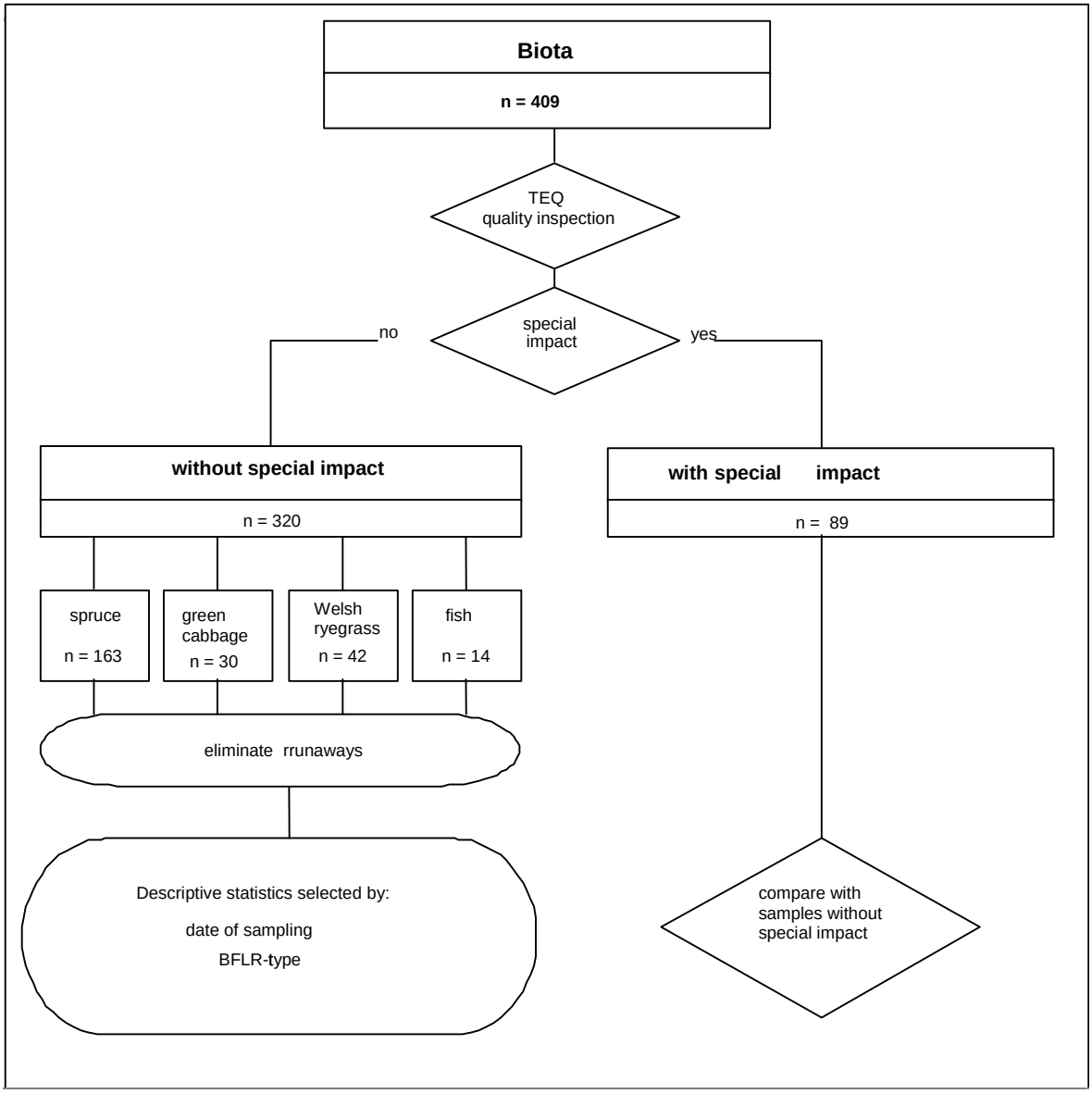

Figure 30: Evaluation scheme compartment Biota (PCDD/PCDF)

The data set, on its turn, was divided into samples without special impact and into samples with data on special impacts. The data set without special impact was used for the further evaluation, the data set with data on special impacts is compared with these data in the respective chapter of the biota type (species). The evaluations refer only to PCDD/PCDF.

7.4 Spruces

The by far most comprehensive data set on biota is available for spruces. Only samples from the program “Bioindicators of airborne dioxins and furans with spruce needles” (Bayr. Landesamt für Umweltschutz) are available with the marking “without special impact”. They may be subsequently compared with data from the program “Ecosystem-related biomonitoring program in the region of Biebesheim 1992-1994“ (Hessisches Landesamt für Umwelt und Geologie) marked as being subjected to special impact in the surroundings of a waste incineration plant.

Fig. 31 shows the whole data inventory from the program carried out in Bavaria. Between end of October/beginning of November or beginning of April samples were taken over the whole period. Fig. 31 shows up to 4 times higher contents in individual measuring points in April reflecting higher ambient air impacts throughout the winter.

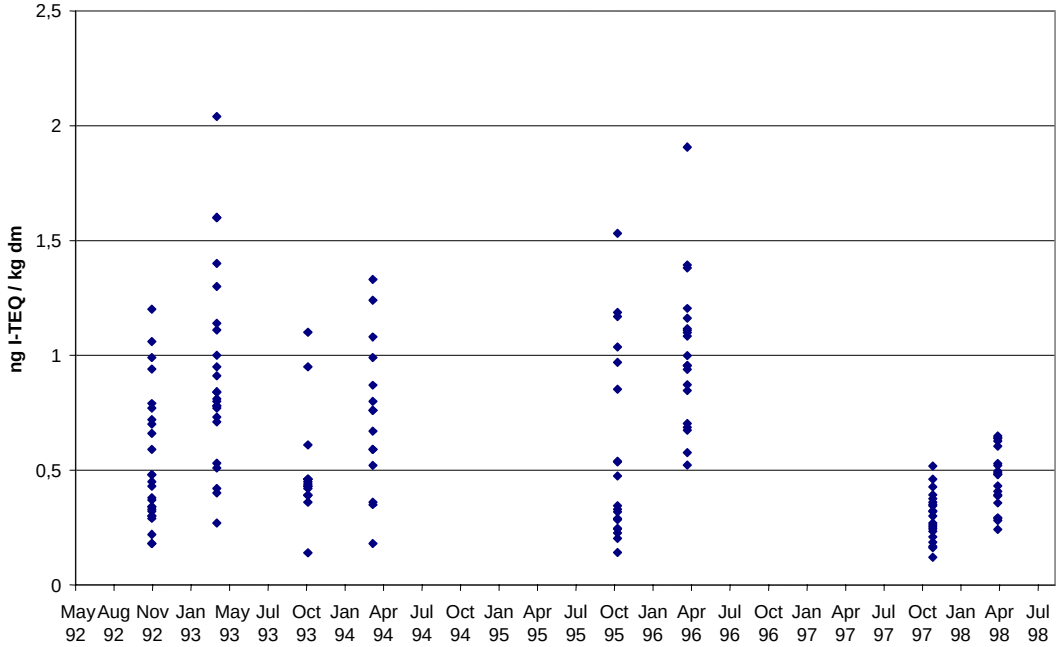

Figure 31: Survey of measured data “Bioindication of airborne dioxins and furans with spruce needles (Bayr. Landesamt für Umweltschutz)

Fig. 32 shows the statistical indicators of the whole data inventory. Information on the temporal development cannot be given as the sampling locations changed in the periods of investigation. If the data sets of 1997 and 1998 are used for deriving actual impacts from sampled spruce needles in Bavaria contents in the range of about 0,5 ng I-TEq/kg dm (90 percentile) have to be expected.

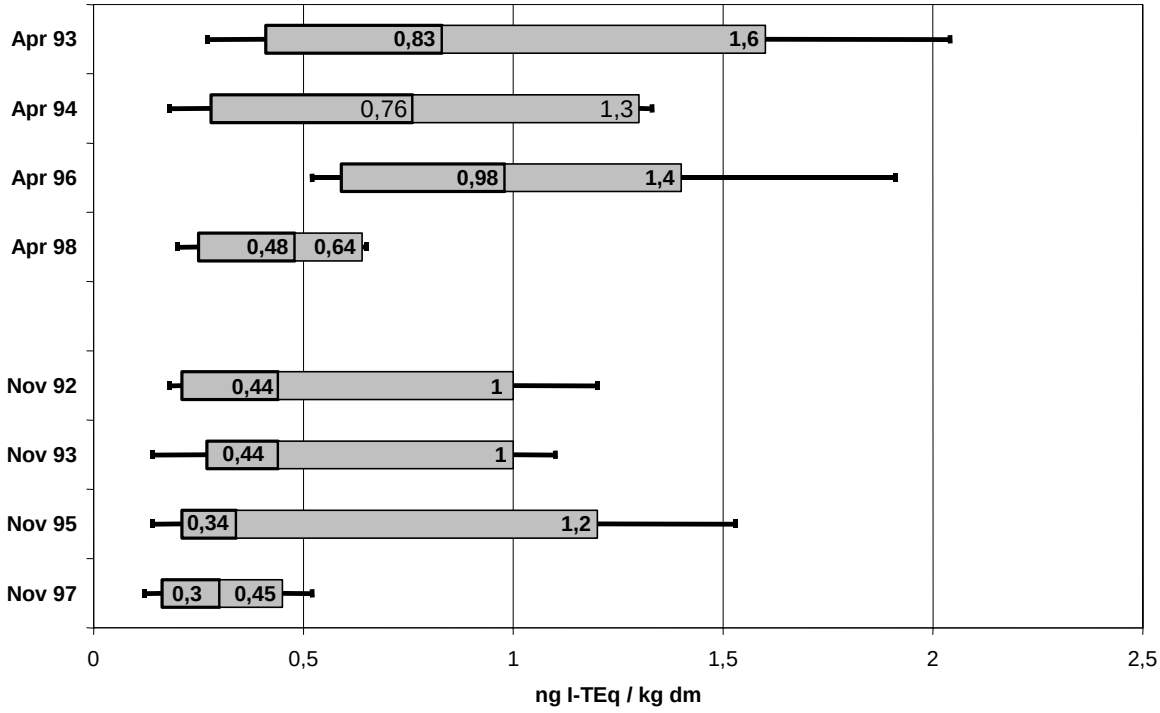

Figure 32: PCDD/PCDF contents in spruce needles “Bioindication of airborne dioxins and furans in spruce needles” (Bayr. Landesamt für Umweltschutz)

Beside the data of the program of investigation described the database DIOXINS contains spruce needles from the program “Ecosystem-related biomonitoring program in the region of Biebesheim 1992-1994“ (Hessisches Landesamt für Umwelt und Geologie). These samples are marked by “special impact (waste incineration plant)”. Fig. 33 shows the PCDD/PCDF contents. They were predominantly detected in the agglomeration area type and in some samples in the urbanized area type. With median contents of 1.7 ng I-TEq/kg dm (90 percentile = 2.3 ng I-TEq/kg dm) for 1993 they are slightly higher than the Bavarian contents. Apart from the “impact” also the remaining structure of emission sources in the respective area of investigation has to be considered. It is not possible to derive from the data set to difter “special impact” or differences in the regionally typical contents.

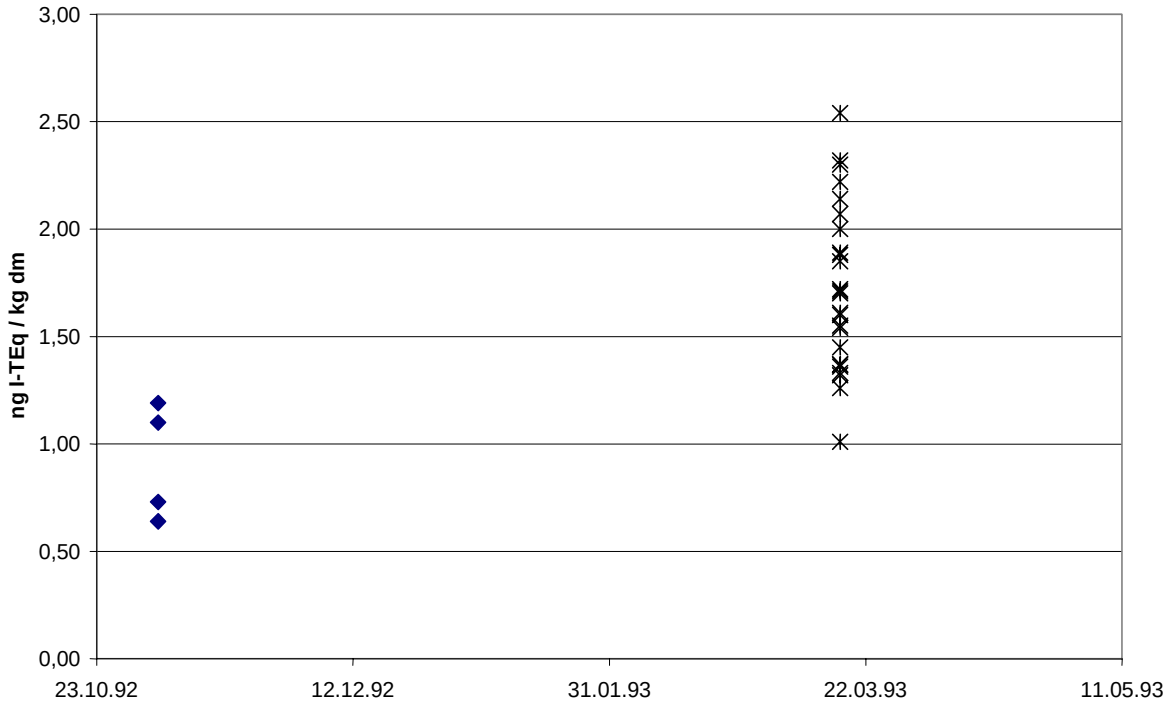

Figure 33: Survey of measured data on spruces “Ecosystem-related biomonitoring program in the region of Biebesheim 1992-1994“ (Hessisches Landesamt für Umwelt und Geologie)

7.5 Green cabbage

Green cabbage and Welsh ryegrass cultures are typical standardized bioindication cultures for monitoring ambient air impacts as also described in the VDI standards for bioindication of airborne pollutants. Data on green cabbage from the measuring program without special impact are only available for rural areas and agglomerations in Bavaria (August/September or October/November 1996/1997). The data set comprises altogether only 24 samples. They are confronted with 34 green cabbage samples from the Hesse “Biebesheim program” and six samples from the agglomeration Bremen (March 1993) with special impact.

With one exception, the data from late summer and autumn of 1996/1997 are below 1 ng I-TEq/kg dm (August/September, as a rule, below 0,25 ng I-TEq/kg dm, October/November, as a rule, between 0,4 and 0,6 ng I-TEq/kg dm) (Fig. 34). Thus, the seasonally caused increase towards the winter is clearly visible. Thus, green cabbage samples are in concentration ranges as they were detected also for spruce needles, as a rule, after eight weeks of exposition. The less comprehensive data set did not show differences as regards the area type (rural, agglomeration).

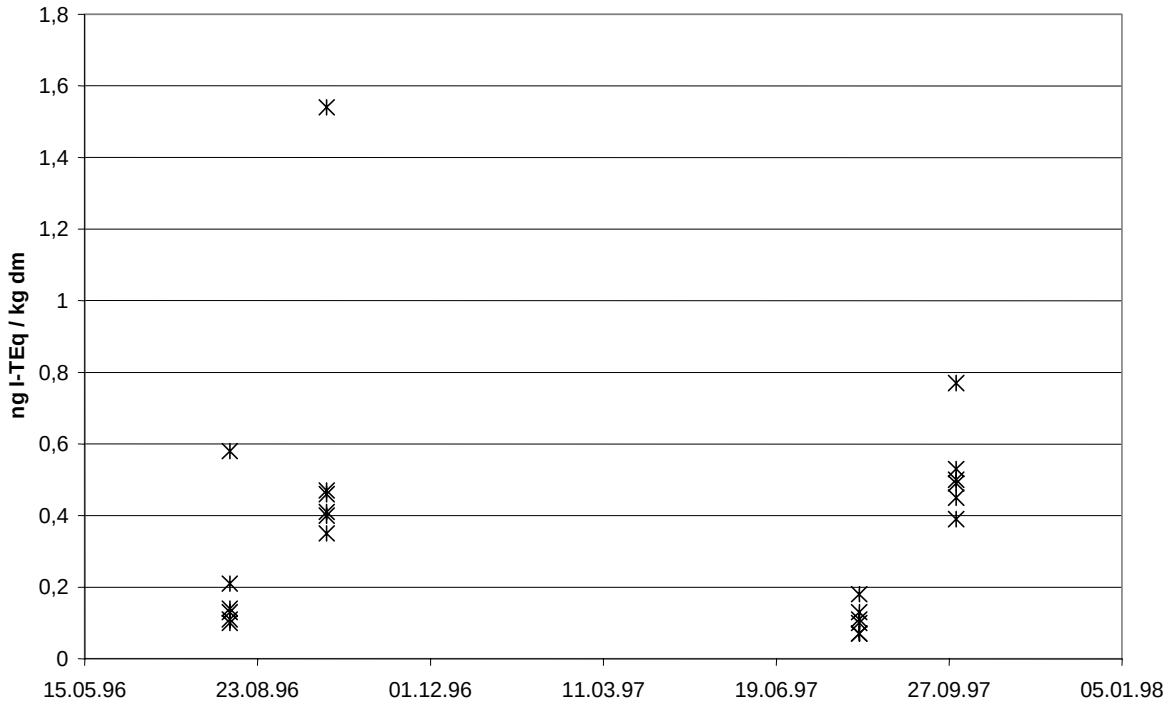

Figure 34: Survey of green cabbage data without special impact (Bavaria)

The green cabbage samples from the Hesse “Biebesheim program” with special impact (November 1992) are represented for all mesasuring points in Fig. 35. With one exception, the concentrations vary between 0,5 and 1,5 ng I-TEq/kg dm (median 0.9 ng I-TEq/kg dm, 90 percentile 1,3 ng I-TEq/kg dm) and thus, on average, above the contents from Bavaria in 1996/1997. This can have various causes, the most important of which might be that the exposure duration of the Hesse program totalled five months (sowing early in June, harvesting mid-November 1992), whereas later biomonitoring programs have duration of only eight weeks. As the ambient air concentrations are declining in many locations in Germany it is quite possible that in 1992 not yet all emission-reducing measures were successful and thus also in the early 90-ies higher concentrations were detected in the biomonitors. Finally, comments as to an actual impact can be made as comparative data from this year are not available in the database DIOXINS.

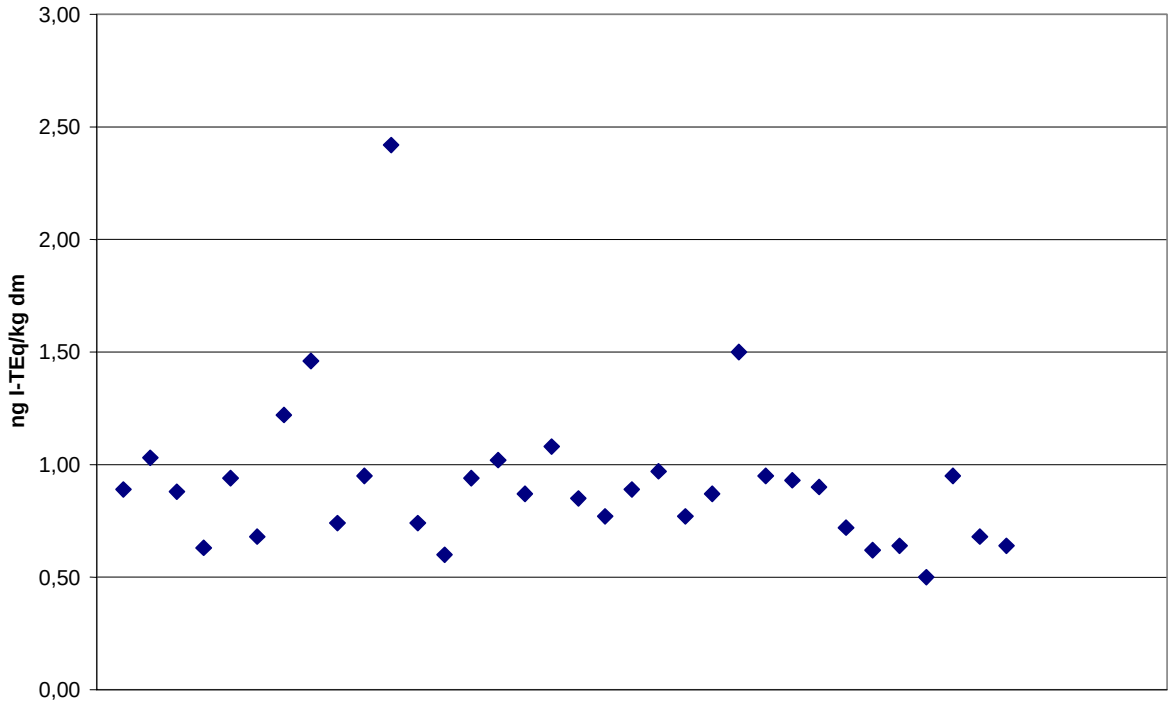

Figure 35: Survey of green cabbage data with special impact: “Ecosystem-related biomonitoring program in the region of Biebesheim 1992-1994“ (Hessisches Landesamt für Umwelt und Geologie)

The six green cabbage samples from the agglomeration Bremen (1993, non-specific special impact) show PCDD/PCDF contents between 1,2 and 2,6 ng I-TEq/kg dm and are thus, on average, in the upper range of the contents in the region Biebesheim.

7.6 Welsh ryegrass

Welsh ryegrass cultures are also equally used as standardized biomonitoring cultures for the detection of ambient air impacts. Due to a reduced growth towards autumn they are predominantly used in the summer period (May till September) whereas green cabbage is typically used in late summer and primarily in autumn.

Investigations of Welsh ryegrass are only available from the research program “Biomonitoring (Welsh ryegrass, green cabbage) in six locations“ (Bayr. Landesamt für Umweltschutz) (n = 42, 1996/1997). With one exception, the PCDD/PCDF contents are below 1.5 ng I-TEq/kg dm (median 0,5 ng I-TEq/kg dm, 90 percentile 1 ng I-TEq/kg dm) for 1996. The few data of 1997 are below 0,5 ng I-TEq/kg dm. Due to only two sampling years and the small data set volume it is not possible to speak here of a secure trend towards a reduction of the ambient air impact in this period – detected via Welsh ryegrass biomonitoring. The insignificant data set does not show differences as to the area type (rural, agglomeration). Data sets on Welsh ryegrass samples with special impact were not available.

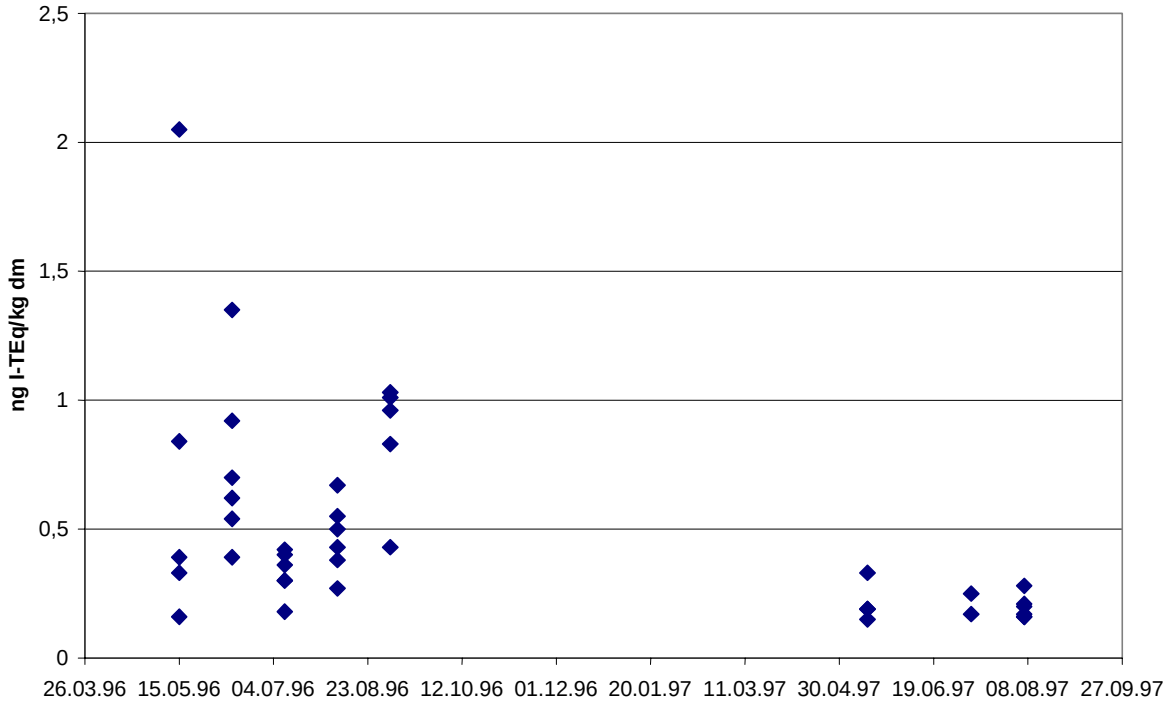

Figure 36: Survey of Welsh ryegrass data without special impact (Bavaria)

The program “Investigation of transfer in the region of Eisenhuettenstadt” (Landesumweltamt Brandenburg) involves nine samples marked non-specifically as cabbage/grass and with special impact (“dioxin-relevant former industrial plant”). These samples of 1993 show PCDD/PCDF contents between 1. 5 and 7.7 ng I-TEq/kg dm . Due to Welsh ryegrass data or other comparative grass/cabbage data from this sampling year lacking comments on the extent of impact of these samples cannot be made.

7.7 Fish, mussels

In contrast with the plant bioindicators of the types spruce, green cabbage and Welsh ryegrass reflecting ambient air impacts animal bioindicators such as fish or mussels shall be considered under aspects as discharge monitoring or impact on the animal food chain (thus connection with the compartment Foodstuffs).

The data set on biota fish with the marking “without special impact” involves only 14 samples from the program “R&D biomonitoring of the Elbe river” (Freie und Hansestadt Hamburg. Behörde für Arbeit, Gesundheit und Soziales, 1994). Two samples from the program “Investigation of transfer in the region of Eisenhuettenstadt” (Landesumweltamt Brandenburg) with the marking “dioxin-relevant former industrial plant” are added as special impact.

Fig. 37 shows the PCDD/PCDF contents in breams from the Elbe-river in 1994. With one exception, the values are below 5 ng I-TEq/kg of fresh weight. Indications to changes of the contents along the river or in individual sections of the river did not appear. The extreme value of about 13 ng I-TEq/kg of fresh weight cannot be explained in greater detail.

The two samples from the region of Eisenhuettenstadt marked with “special impact” show contents of 42 (perch, 1993) or 60 ng I-TEq/kg dm (fish not specified, 1993).

Here the differing data in fresh weight (bream) and dry weight (perch or fish not specified) should be taken into account. Considering the various fat contents of bream with approx. 5.5 % and of perch approx. 0.8 % and the average water content of about 80 % (Deutsche Forschungsanstalt für Lebensmittelchemie, 1991) fat-related contents of about 20-90 pg I-TEq/g fat for bream (based on 1-5 ng I-TEq/kg of fresh weight) and about 100 pg I-TEq/g of fat for perch (based on 42 ng I-TEq/kg dm) are obtained thereof in a first approximation. Thus, the sample from Eisenhuettenstadt definitely extends to the concentration range of breams from the Elbe river

In addition, the data set marked with “special impact” contains 13 samples of mussels from the program “Investigation of impact from soil, mussels and sediments from the area of Wilhelmshaven” (Lower Saxon Ministry for the Environment, 1994); as special impact industrial discharges are mentioned. The PCDD/PCDF contents are here between 0.55 and 0.96 ng I-TEq/kg of fresh weight. Considering a fat content of mussels of about 1.3 % fat-related contents of about 42-74 pg I-TEq/g of fat result from it and thus, on the other hand, values as indicated for fish in the database DIOXINS.

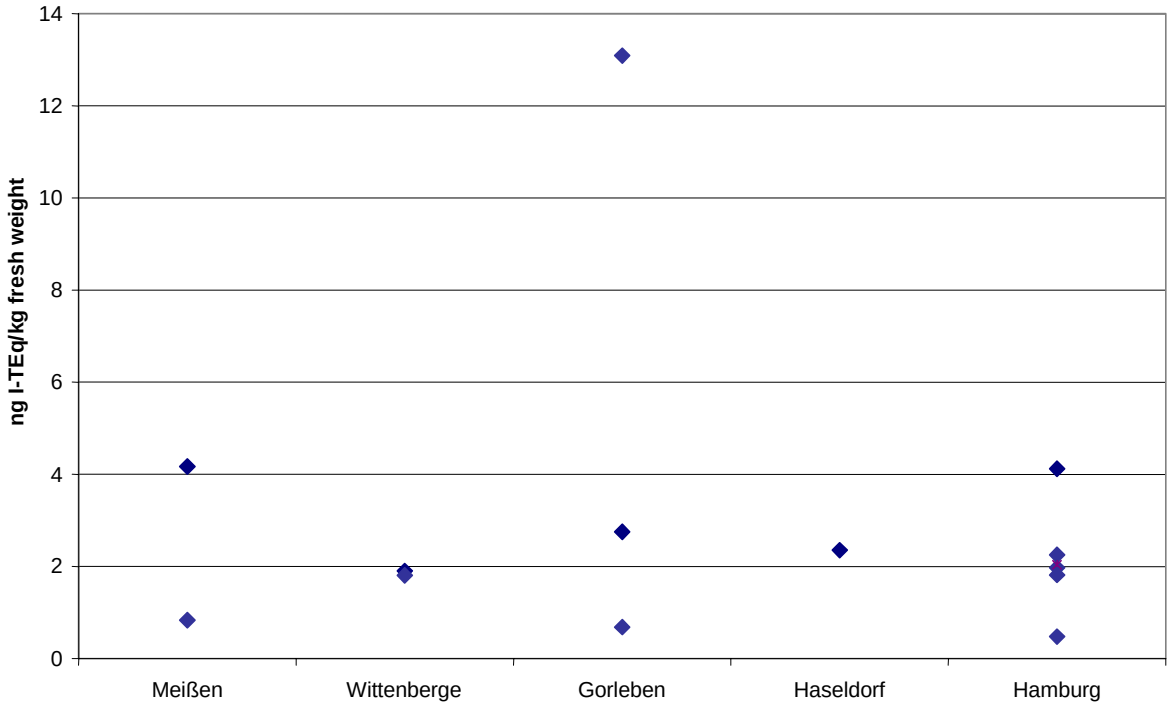

Figure 37: Survey of fish samples (bream) without special impact from the Elbe river (1994)

7.8 Summary Biota

The data set for the compartment Biota contains samples for the species spruce needles, green cabbage, Welsh ryegrass, fish (bream, perch) and mussels.

According to the ambient air concentrations spruce needles show trends towards a higher content in April – i.e. after exposition in winter. Furthermore, a decline of the PCDD/PCDF contents (90 percentile) by more than half of it becomes obvious in spruce needles in later measurement series (1997/1998) as compared with data from 1992 – 1996. PCDD/PCDF loads in spruce needles from Bavaria from the years 1997 and 1998 are in the range of about 0.5 ng I-TEq/kg dm (90 percentile).

The PCDD/PCDF contents for Welsh rygrass from Bavaria are below 1.5 ng I-TEq/kg dm (median 0,5 ng I-TEq/kg dm, 90 percentile 1 ng I-TEq/kg dm) for 1996, with one exception. The few data from 1997 are below 0.5 ng I-TEq/kg dm.

Green cabbage cultures from Bavaria show PCDD/PCDF contents, as a rule, below 1 ng I-TEq/kg dm late in summer and autumn 1996/97 (August/September, as a rule, below 0.25 ng I-TEq dm, October/November, as a rule, between 0.4 – 0.6 ng I-TEq/kg dm).

Only in exceptional cases the data set volume allows to make comments on area-typical differences. Thereby, differences were not to be stated for the compartment Biota.

PCDD/PCDF contents for a few breams, as they were detected for samples from the Elbe river in 1994, are below 5 ng I-TEq/kg of fresh weight, with one exception. Indications to changes of the contents along the rivercourse or in individual sections were not detected. Derivations as regards fat-related contents furnished values between 20 and 90 pg I-TEq/g of fat. Mussels show PCDD/PCDF contents between 0.55 and 0.96 ng I-TEq/kg of fresh weight. Related to the fat content loads as in fish are obtained.

8. COMPARTMENT WASTE

8.1 Introduction

This compartment is envisaged to include measured data from the comprehensive compartments Waste for utilization and Waste for disposal, thus recording important sinks of dioxins and PCB. Sinks from various pathways may e.g. be:

- sink sewage sludge: pathway: domestic use – sewage

  pathway: industrial stepses - wastewater

  pathway: emission (plant, traffic ...) – deposition – effluents (road...) - wastewater

- sink waste from waste gas purification plants:

  pathway: industrial stepses – waste gas purification

  pathway: disposal (energetic utilization, thermal treatment) waste gas purification plants

- sink wastes from production and processing.

Apart from that, wastes are basic materials for further stepses/uses (recycling etc.) and, in this connection, will be again relevant as pollutant sources. As can be seen from the examples of sink functions this compartment is furthermore relevant to the balancing of pollutant flows.

The compartment Waste consists mainly of the matrix sewage sludge. All further matrices are shortly evaluated at the end of this chapter.

With the Ordinance on Sewage Sludge (AbfKlärV) appropriate limits as to placing sewage sludge on soil used for agriculture or horticulture are available for this matrix. They are connected with the targets of soil protection and the targets of health-related consumer protection (obtaining harmless foodstuffs and feedingstuffs). They total 100 ng I-TEq/kg dm for PCDD/PCDF, for the sum of six selected PCB (# 28, 52, 101, 138, 153, 180) 200 µg/kg dm per individual component. Thus, the evaluation for the matrix sewage sludge is connected with these existing limits apart from considering temporal and regional trends.

8.2 Data inventory and quality

A survey of parameters which may be put into the database DIOXINS to describe samples from the present compartment in greater detail is given in Fig. 38. Thereby, parameters which may be relevant to the evaluations (temporal, spatial, impacts) required basically are pointed out. However, a possibility of evaluating these parameters requires a sufficient number of inputs into the data base. It is equally shown hereinafter if this was given.

With regard to the documentation of the individual input parameters the following main possibilities of evaluation may be derived thereof for the compartment Waste:

- type sewage sludge
- year of sampling
- area type (rural, urbanized, agglomeration areas)
- I-TEq (-detection limit)

Table 10 gives a survey of the actual data inventory on the compartment Waste in the database DIOXINS and on the characteristic sample properties used in the course of evaluation.

440 of the altogether 466 samples of this compartment are from sewage sludge; further data sets of waste matrices do not exist to an extent relevant to evaluation.

The data set “sewage sludge” is, temporally and regionally regarded, very inhomogeneous. Only the data set of the Umlandverband Frankfurt/Main (Association of the surrounding area of Frankfurt/Main) provides the possibility of making comments on the temporal trends for identical locations. The comprehensive research program “Nationwide analysis of sewage sludge for PCDD/PCDF” completes the research spectrum in regional respect, yet for a big part of samples detailed explanations (sampling location, BFLR type) are lacking. Data on the sampling date are not available for this measuring program and for the programs “Chloroaromatics dioxin measuring program Thuringia” and “Investigation of potential sources of PCDD/PCDF in sewage sludge” which excludes an evaluation with regard to temporal trends.

Furthermore, the program “Chloroaromatics dioxin measuring program Thuringia” includes only TEq values incl. detection limit for PCDD/PCDF, however no data on individual congeners are not available in the database. Thus, the PCDD/PCDF data from this program were not included in the evaluation.

Compartment Waste:

1. Reason for the investigation:

environmental monitoring (y/n)
permanent monitoring (y/n)
approval procedure (y/n)
investigation of former industrial sites/soil
research project (y/n)
legally prescribed investigation (y/n)
investigation of incident (y/n)
measuring program /name
others

2. Target of investigation:

determination of substance flows (y/n)
consideration of transfer/pathways
connection with other samples
connection with other measuring programs

3. Location data:

community indicator/BFLR type ✓
type of plant
steps
operator
target product
rated capacity
number of charges
residues/wastes
efficiency
operating state
waste quantity
residue quantity
starting material/content
others
existing waste pretreatment plant
sewage plant type

4. Sampling:

date
sampling strategy
industrial waste (y/n)
ash (y/n)
waste oil (y/n)
slag (y/n)
filter dust
type (fíbrous filters/electrostatic filters)
filter sludge
sewage sludge from a biol. sewage plant
other purification methods
municipal waste domestic waste
municipal waste ash of domestic fuel
municipal waste fire residue
other municipal waste
sewage sludge of a biol. sewage plant
sewage sludge of a municipal sewage plant ✓
compost
dump goods
rumble
earth excavation
excavated earth

municipal sewage
dump leakage water
industrial effluents
road effluent
sampling at a wastewater discharge location
sampling behind the pretreatment plant
sampling at the inflow to the sewage plant
effluent from the municipal sewage plant

individual sample (y/n)
mixed sample (y/n)
suspected sample (y/n) + cause

sampling quantity
sample area
transport of samples (transport vessel/conditions/time))

5. Laboratory data

storage time
storage temperature
regulation for the preparation of samples
regulation for analysing
number IS (PCDD/, PCB)
quantity IS
confidence interval
participation in co-operative tests
external/internal quantification
recovery

6. **Analytical results:** ✓

recovery
detection limit

Figure 38: Potential input parameters database DIOXINS compartment Waste/Recoverable materials/Residues(**bold** = considered for evaluation, **bold+✓** = suited for evaluation (number of samples, data quality...))

Table 10: Survey of measuring program Waste

No.	Institution	Name of measuring program	Year of sampling	BFLR type A = agglommeration. S = urbanized area L = rural area - = no data	Special impact - = no data I = industrial plant B = not specified S = incident	Type of waste A = ash F = filter dust I = industrial waste KS = sewage sludge S = other	Number of samples	PCDD/ PCDF	PCB
1	Landesumweltamt Brandenburg	Investigation of transfer in the region of Eisenhuettenstadt	1993/1994	A, L, -	-	KS, I + F	18	X	
2	Staaatliches Amt für Umwelt Dessau-Wittenberg (Saxony-Anh.)	Investigation of the surroundings of copper mill Ilsenburg 1990	1990	S	-	A + F	3	X	
3	Thüringer Landesanstalt für Umwelt	Chloroaromatics dioxin measuring program Thuringia	--	S, L	-	KS	51	X ***)	X
4	University Bayreuth	Sewage sludge surrounding area of Frankfurt/Main	1991, 1993-1997	A, S	-	KS	192	X	X
5	University Bayreuth	Precipitation-dependent input of PCDD and PCDF into an urban drainage systems	1991, 1992	A, L	-	KS, S **)	53	X	
6	Federal Environmental Agency	Occurence of PCDD/PCDF in cases of fire	-	A	- (S)	F	1	X	
7	Federal Environmental Agency	Investigation of potential sources of PCDD/F in sewage sludge	-	A, S, L	-	KS	30	X	
8	Federal Environmental Agency	Nationwide analysis of sewage sludge for PCDD/PCDF	- *)	A, -	-	KS	118	x	

*) 1990 oder before

**) Gully sediments, canal sediments, sediments rain retention basin, road dust

***) PCDD/PCDFnot considered in evaluation

8.3 Evaluation procedure

The procedure adopted for evaluating sewage sludge data is represented in Fig. 39

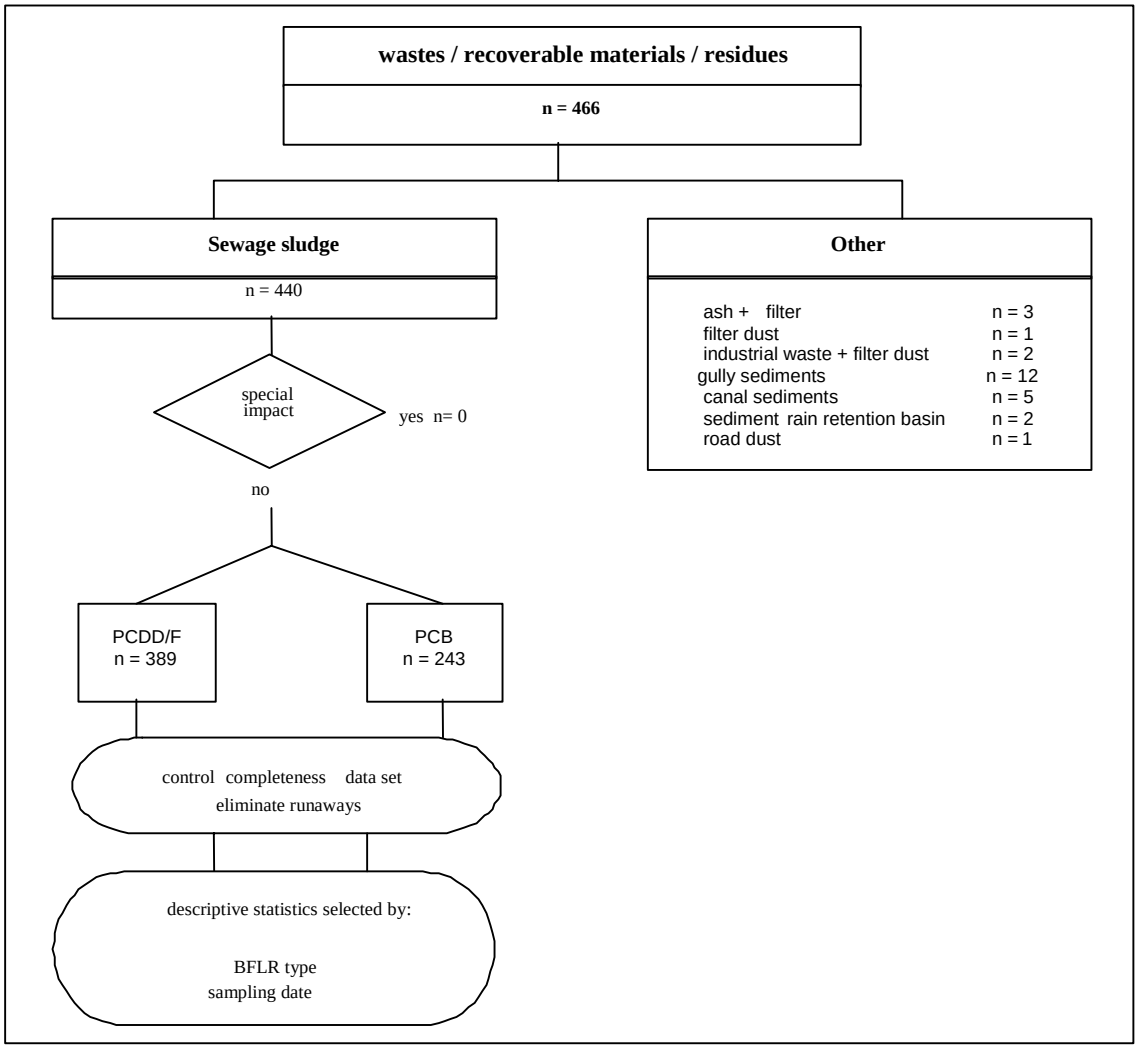

Figure 39: Evaluation scheme compartment Waste

A classification into sewage sludges not specially impacted and samples from areas subjected to strong impact (in this case e.g. special discharges) was not made. First of all, all data are therefore to be classified as background contents. Considerations of runaways, however, have to be considered.

8.4 PCDD/PCDF contents in sewage sludge

8.4.1 Runaways

Fig. Fig.:40 shows the variation range of measured data and identifies runaways. These samples were not considered for a further evaluation with regard to background contamination.

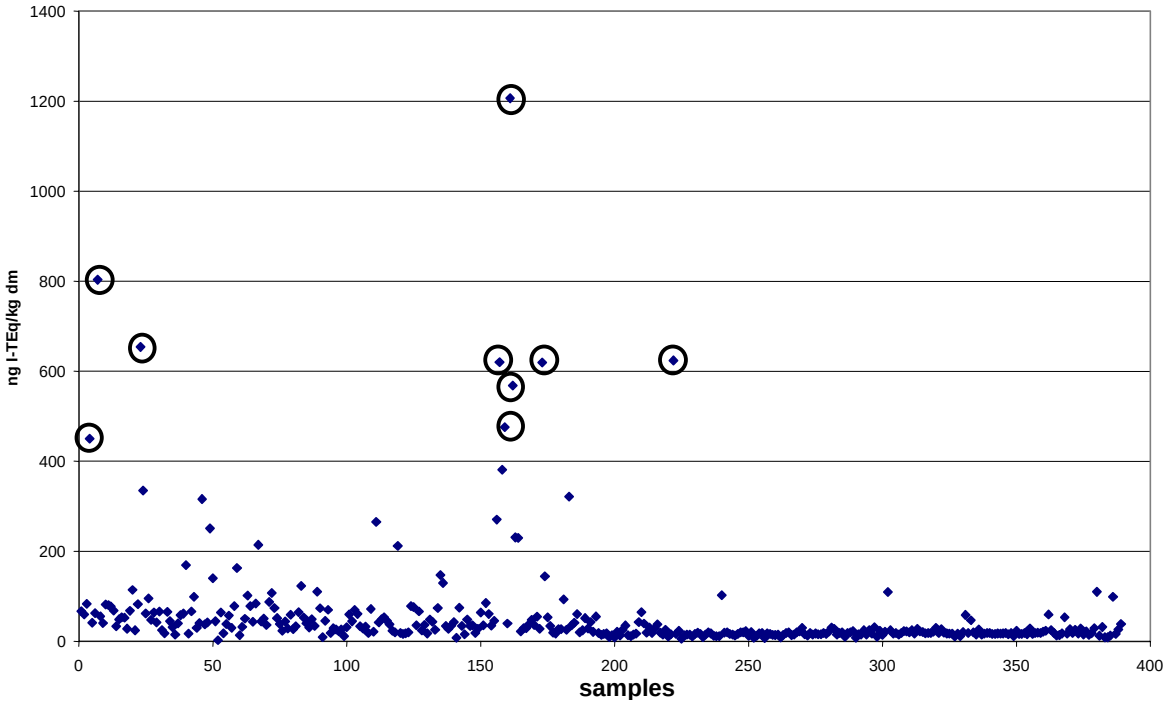

Figure 40: Distribution of PCDD/PCDF content in sewage sludge samples and runaways (I-TEq, -detection limit)

Runaways come from the programs no. 1, 4 und 7 (Table 10) and – if a sampling date is available – from the period 1991 until 1994. The samples come from agglomeration and rural areas.

8.4.2 Temporal trend

The data set comprises samples of the period 1990 till 1997, program no. 8 (without date of sampling), in addition, it contains data determined before 1991.Fig. 41 shows the temporal

course of the PCDD/PCDF contents in sewage sludge. Hereby, program no.8 was equated with 1990.

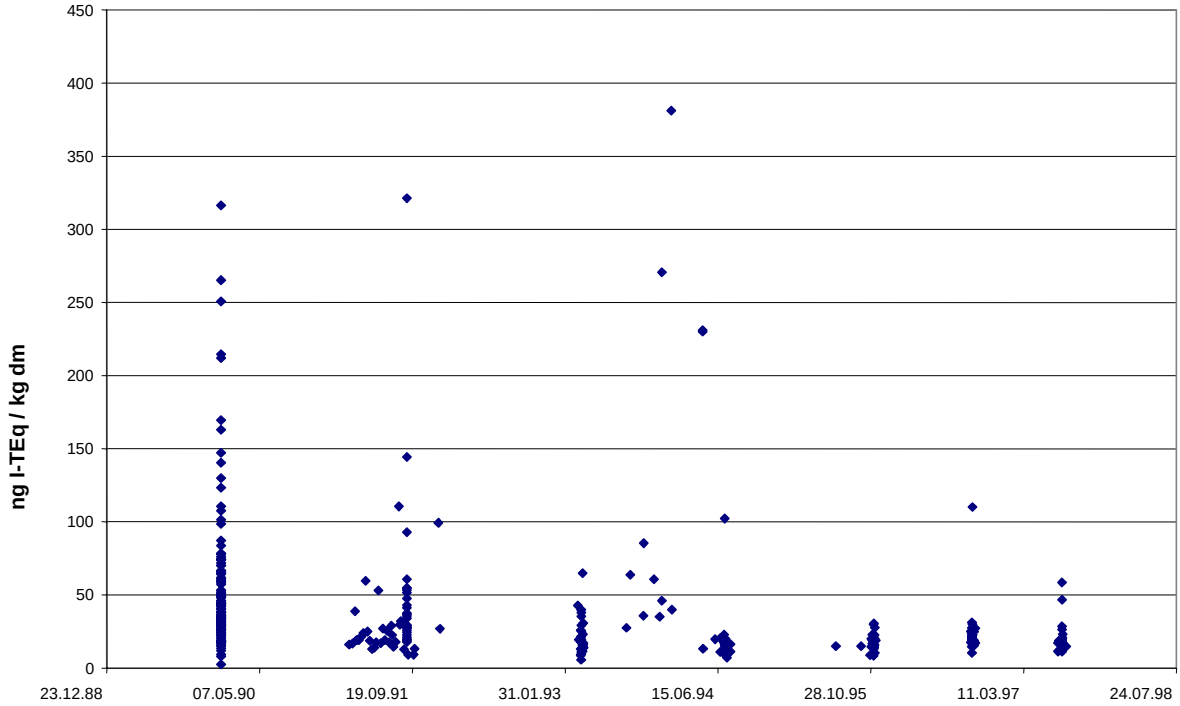

Figure 41: Temporal course of PCDD/PCDF content in sewage sludge in the period 1990-1997 (I-TEq, -detection limit)

Fig. 42 shows a nearly continuous decline of the average content of about 42 ng I-TEq/kg dm in 1990 (median) to about 17 ng I-TEq/kg dm in 1997. Furthermore, some runaways are recognizable within the individual yearly data sets.

The data set of the Umlandverband Frankfurt allows to follow the temporal trend over seven years in nearly identical plants. Here, the trend may be recognized still more clearly (Fig. 42). Following the procedure so far adopted in defining the content without special impact on the basis of the 90 percentile background contents below 30 ng I-TEq/kg dm and thus by far below the limit of the Sewage Sludge Decree (Fig. 41) have been obtained as of 1995. The cases of exceeding the limit in the present data sets are also declining.

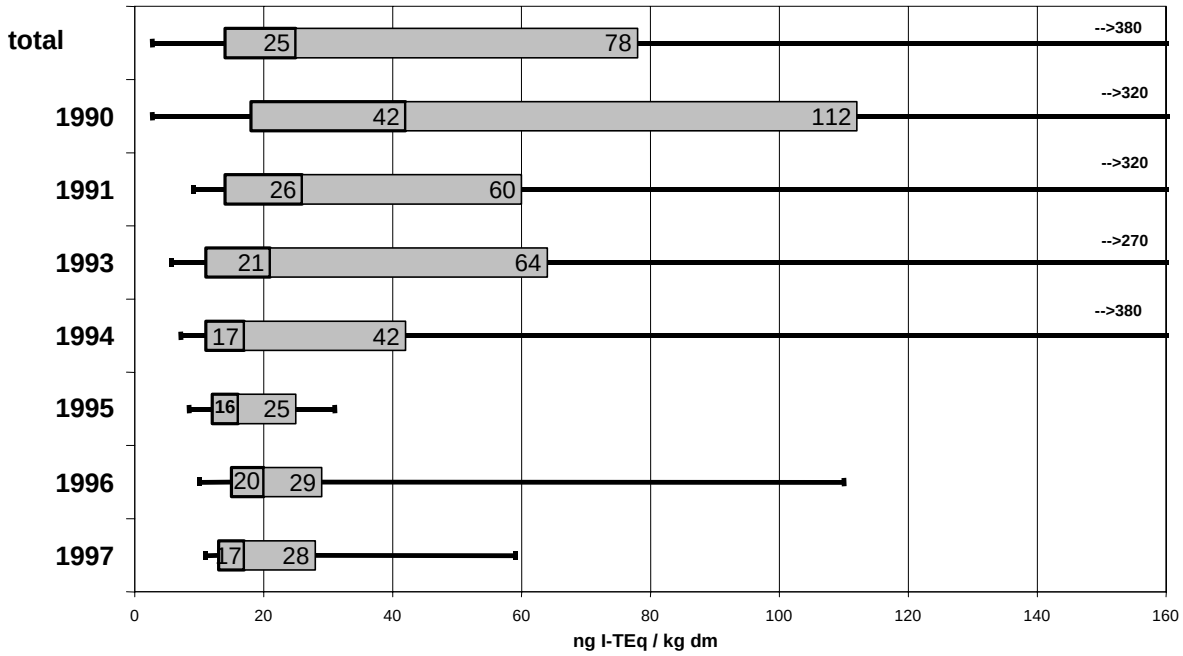

Figure 42: Statistical indicators of the temporal trend of PCDD/PCDF contents in sewage sludge in the period of 1990-1997 (I-TEq, -detection limit)

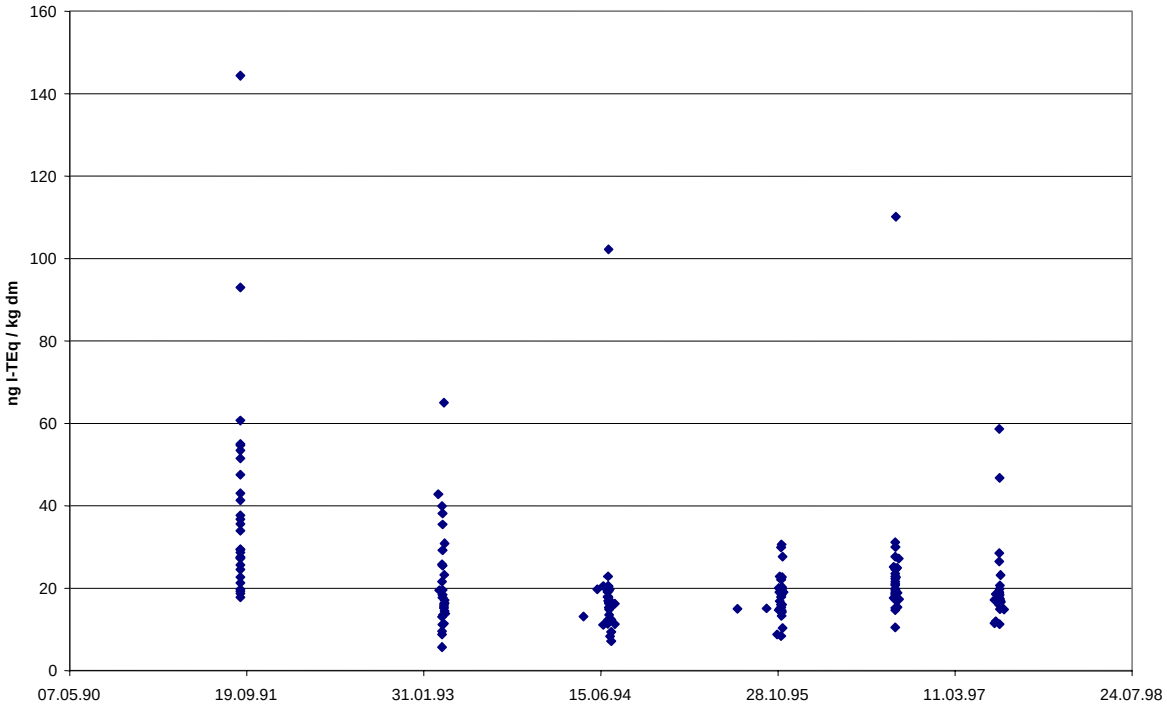

Figure 43: Temporal course of the PCDD/PCDF content in sewage sludge of the Umlandverband Frankfurt in the period 1991-1997 (I-TEq, -detection limit)

8.5 Summary compartment Waste/Residues/Recoverable materials

Following the compartments Foodstuffs, Soil, Ambient air and Deposition the compartment Waste/Residues/Recoverable materials with altogether 466 samples has the fifth biggest data set in the database DIOXINS. It concentrates nearly exclusively on sewage sludge. A differentiation between samples from areas without special impact and samples coming possibly from impacted areas was not made. After eliminating the runaways all samples were summed up; they have to be classified as background contents.

The sewage sludge data allow an evaluation with regard to temporal changes of the PCDD/PCDF and PCB contents. Since the early 90-ies a decline of the contents is shown for both pollutant groups. For PCDD/PCDF a continuous decline of the average contents of more than 40 ng I-TEq/kg dm in 1990, to below 20 ng I-TEq/kg dm in 1997 is obvious. The PCDD/PCDF contents based on the 90 percentile from samples without special impact with clearly less than 30 ng I-TEq/kg dm are by far below the limit of the Sewage Sludge Decree. These background values were derived predominantly from data of agglomerations – in particular from the agglomeration Rhine-Main. Due to data lacking with the aid of the database DIOXINS no further information can be given on other area types.

Apart from sewage sludge a few data from the area industrial waste, filter dust and wastewater sediments are available, the volume of which, however, does not allow a further evaluation.

9 COMPARTMENT SUBSTANCES/PREPARATIONS/PRODUCTS

9.1 Introduction

With the compartment Substances/Preparations/Products the database DIOXINS covers an area which may be relevant with regard to the production of dioxin (sources) as well as to potential exposition pathways (distribution). These are, on the one hand, all substances such as chemicals polluted by PCDD/PCDF already by the production procedure. This is the case primarily for chlorophenols and their derivates or, if alkaline extraction steps for preparation were applied, in synthesis stepses which explains e.g. the pollution of chlorobenzenes by dioxin/furan. Finally by the application of chlorinated metal catalysts a PCDD/PCDF pollution may reach the final product or halogen-free chemicals may be halogenated by radicals present in the steps in the presence of chlorine (NATO/CCMS 1988). Some chemicals contaminated by PCDD/PCDF such as biocides are used in consumer and industrial products. By means of the dioxin/furan pattern the source of dioxin contamination may be identified. In addition, this compartment is suited to identify production procedures connected with a production of dioxin. Furthermore, it is possible to thus document how changes in the production steps (legally prescribed or by voluntary self-obligation) or prohibitions of starting materials, aggregates etc. may affect the PCDD/PCDF contamination of products.

PCDD/F-measured data of products may also furnish supplementary information on the release and distribution of dioxins and are helpful information on dioxin inventories, exposure, assessment etc.

9.2 Data inventory and quality

Fig. 44 gives a survey of parameters which may be put into the database DIOXINS from the present compartment to describe samples in greater detail.

Compartment Substances/Preparations/Products:

1. Reason for the investigation:

- environmental monitoring (y/n)
- permanent monitoring (y/n)
- approval procedure (y/n)
- research project (y/n)
- legally prescribed investigation (y/n)
- ***investigation of incident (y/n)***
- measuring program /name
- others

2. Target of investigation:

- determination of background contamination rural (y/n)
- determination of background contamination urban (y/n)
- determination of background contamination agglomeration (y/n)
- consideration of transfer/pathways
- connection with other samples
- connection with other measuring programs

3. Location data:

- ***community indicator/BFLR type*** ✓
- sampling at the producer (y/n)
- sampling at the manufacturer (y/n)
- sampling at the consumer (y/n)
- sampling at the wholesale (y/n)
- sampling at the importer (y/n)
- others

4. Sampling:

- date
- product name
- product type
- producer
- address
- individual sample/mixed sample
- **suspected sample (y/n)**
- transport of samples (vessel, conditions/ duration)

5. Laboratory data

- storage period
- storage temperature
- regulation for the preparation of samples
- regulation for analysing
- number IS (PCDD/, PCB)
- quantity IS
- confidence interval
- participation in co-operative tests
- external/internal quantification
- recovery

6. **Analytical results:** ✓

- recovery
- detection limit

Figure 44: Potential input parameters database DIOXINS compartment Substances/Preparations/Products (***bold+italic*** = identification of samples with special impact, **bold+✓** = suited for evaluation (number of samples, data quality...))

In particular, the distribution pattern for the individual congeners and the indication of the level of impact on various preparations and products as well as the representation are very important to the evaluation.

Table 11 gives a survey of the actual data inventory for this compartment in the database DIOXINS and of characteristic sample properties. Altogether 251 samples are available. With the exception of a PCB sample only data on PCDD/PCDF are available for all samples. A big part of the samples from the reaearch program “Dioxins and furans in textiles and leather” was investigated not for all individual congeners in accordance with the target of the program, thus partly only octachlorinated congeners or sums of homologues were determined. These data were not further considered here due to the data set being incomplete. Furthermore samples from the program “Investigation of potential environmental hazards in plastic burning” represent a special case in the database DIOXINS. These samples are obviously samples from burning experiments where PCDD/PCDF contents were analyzed and equally not considered.

Table 11 Survey of measuring program compartment Substances/Preparations/Products

Nr	Institution	Name of the measuring program	Year of sampling	Type of product, preparation	Number of samples	PCDD/PCDF	PCB
1	Federal environmental agency	Investigation of potential environmental hazards in plastic burning	1990	Chemicals, consumer goods, materials	57	X	-
2	Senatsverwaltung für Stadtentwicklung des Landes Berlin	Investigation of pesticides from the GDR production for PCDD/F	1990	Chemicals	6	X	-
3	University Bayreuth	Dioxins and furans in textiles and leather	1991-1996	Textiles, leather	148	X	X
4	University Bayreuth	Environmental pollution by dioxins	1994	Chemicals	5	X	-
5	University Bayreuth	Investigation of emission pathways of polychlorinated dioxins and furans in chemical cleaning plants	1992	Textiles	18	X	-

9.3 PCDD/PCDF contamination of chemicals

The database DIOXINS contains PCDD/PCDF concentrations of pentachlorophenols of a various production, trichlorobenzene and pesticides from the GDR production. Table 12 shows the respective PCDD/PCDF concentrations in pg I-TEq/g. In the case of data on spray lindan und buctril A there should be considered that the detection limits per congener were about 20 or 50 pg/g.

Table 12: Survey of PCDD/PCDF in chemicals

Chemical	I-TEq (pg/g)
PCP	79524
PCP	414390
PCP	2311860
PCP	1853045
trichlorobenzene	23
selest	4660
spray hormin	2295
spray hormit	3849
spray lindan	5
tricilin 25	7149

9.4 PCDD/PCDF concentrations in textiles

A program for investigating chemical cleaning plants investigated PCDD/PCDF contents in new textiles after use and after chemical cleaning. Fig.:45 shows the PCDD/PCDF concentrations for these samples and the effect of use and cleaning. In the first case working pants worn in a car workshop are concerned where possibly the presence of engine oil and other oils or waste gases could have contributed to producing very high PCDD/PCDF concentrations. The second pants were worn during assembly work at a conveyor line. The results of these two measurements give rise to the suspection of effects on the workplace. However, the figures show, too, that by chemical cleaning the biggest part of the dioxin contamination is eliminated: from the first working pants approx. 90 % (decline of 112 to 17 pg I-TEq/g) and also from the surface material and the pants on the right of Fig. 45. This proves the cause for the partly very high dioxin concentrations in the distillation residues of chemical cleaning plants.

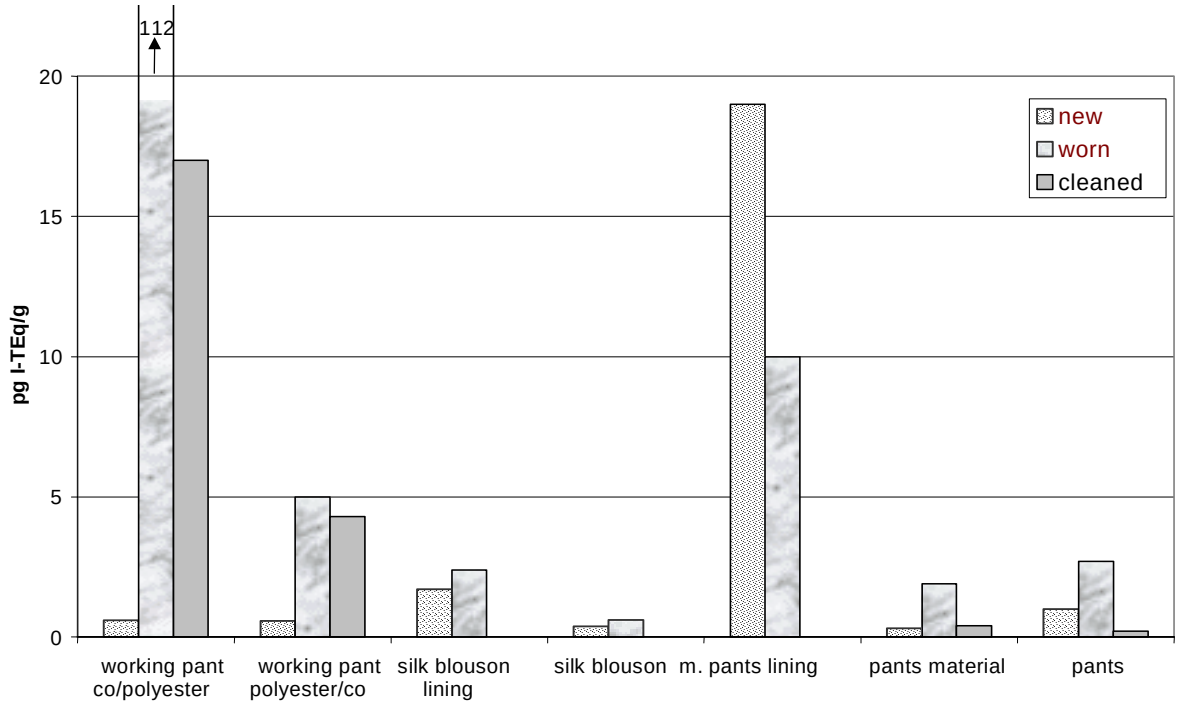

Figure 45: PCDD/PCDF concentrations in textiles (new, worn and cleaned)

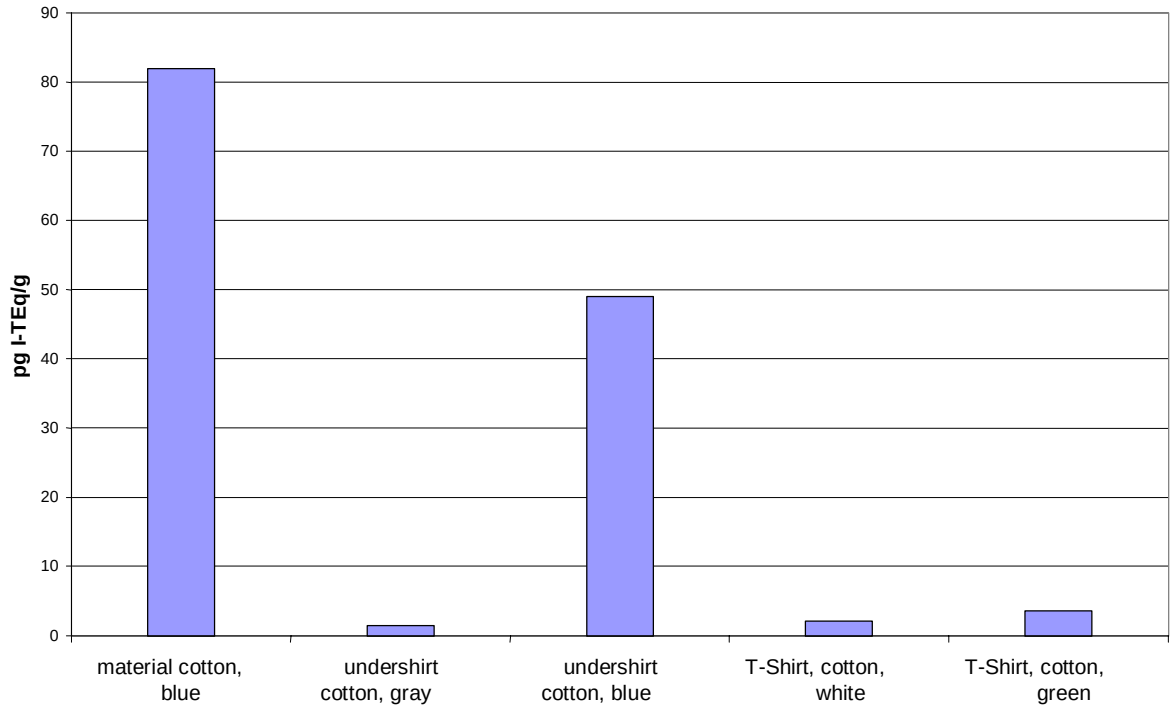

Figure 46: PCDD/PCDF contents in cotton textiles

Fig. 46 shows the results of PCDD/PCDF analyses of cotton textiles and Fig. 47 the range of dioxin concentrations in leather materials and products. As leather was not cleaned or washed before being sold as e.g. textiles the comparatively higher PCDD/PCDF concentrations are not surprising.

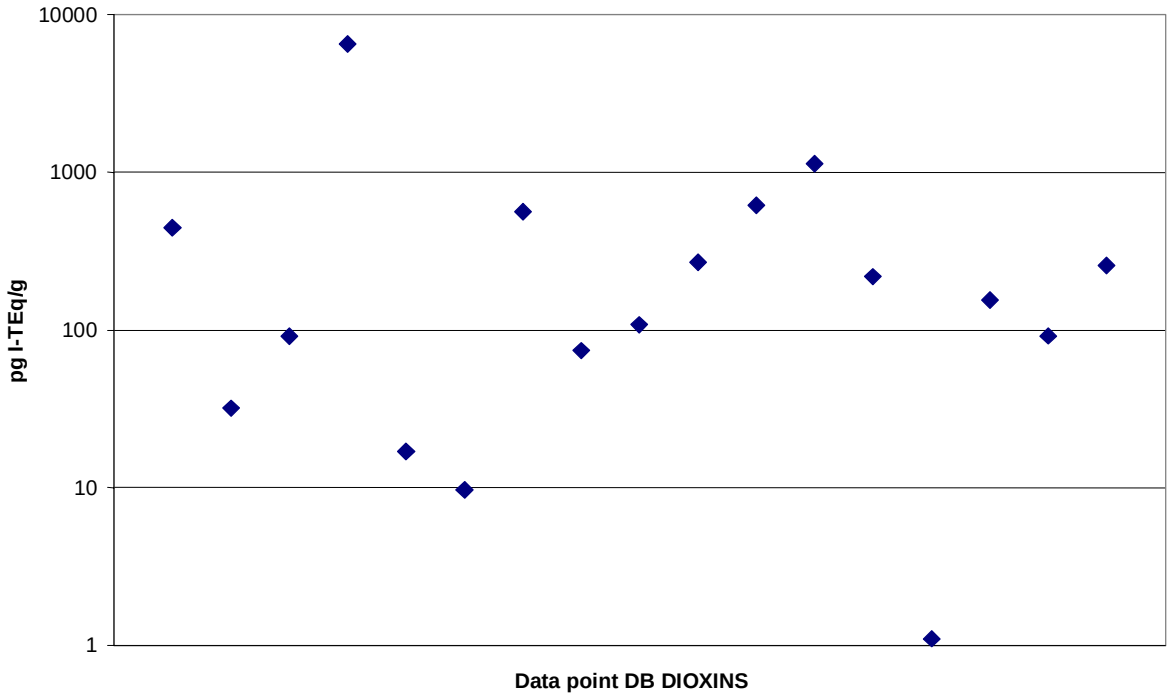

Figure 47: PCDD/PCDF contents of leather materials and products

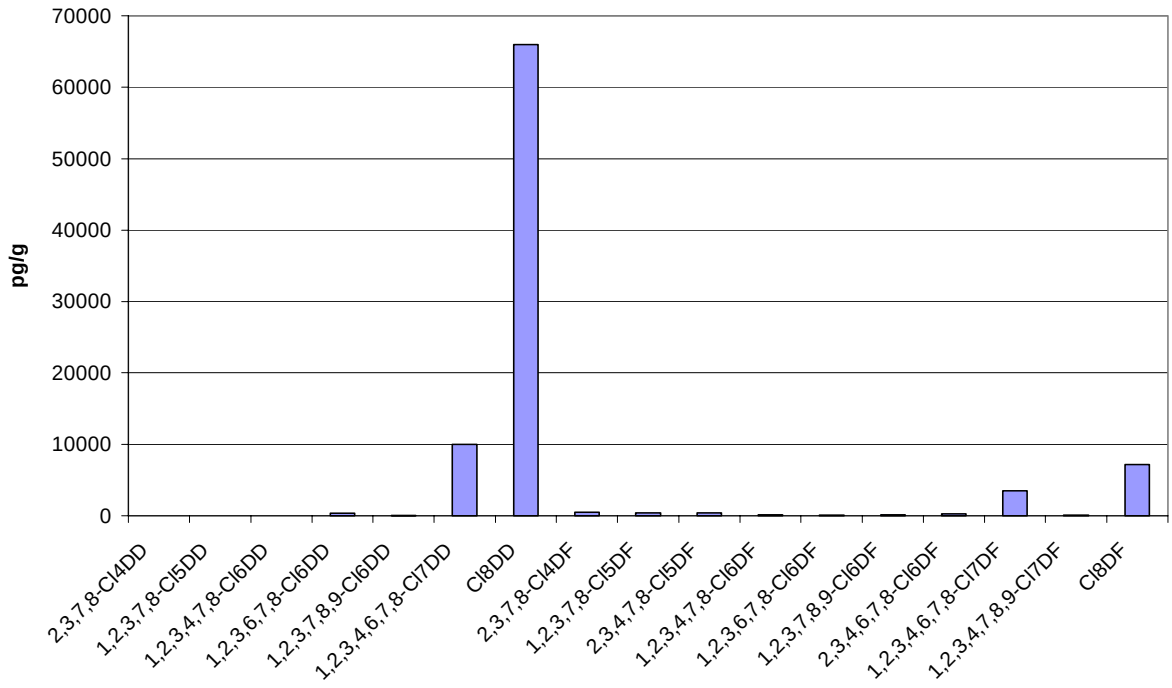

Figure 48: Average congener pattern in impacted leather samples

PCDD/PCDFcontents in leather have to be considered in connection with the use of chemicals. Congener patterns in leather can then give information on the dioxin source. Fig 48 shows an average congener pattern in leather samples. In low impacted samples also deviating patterns may be stated (Fig. 49).

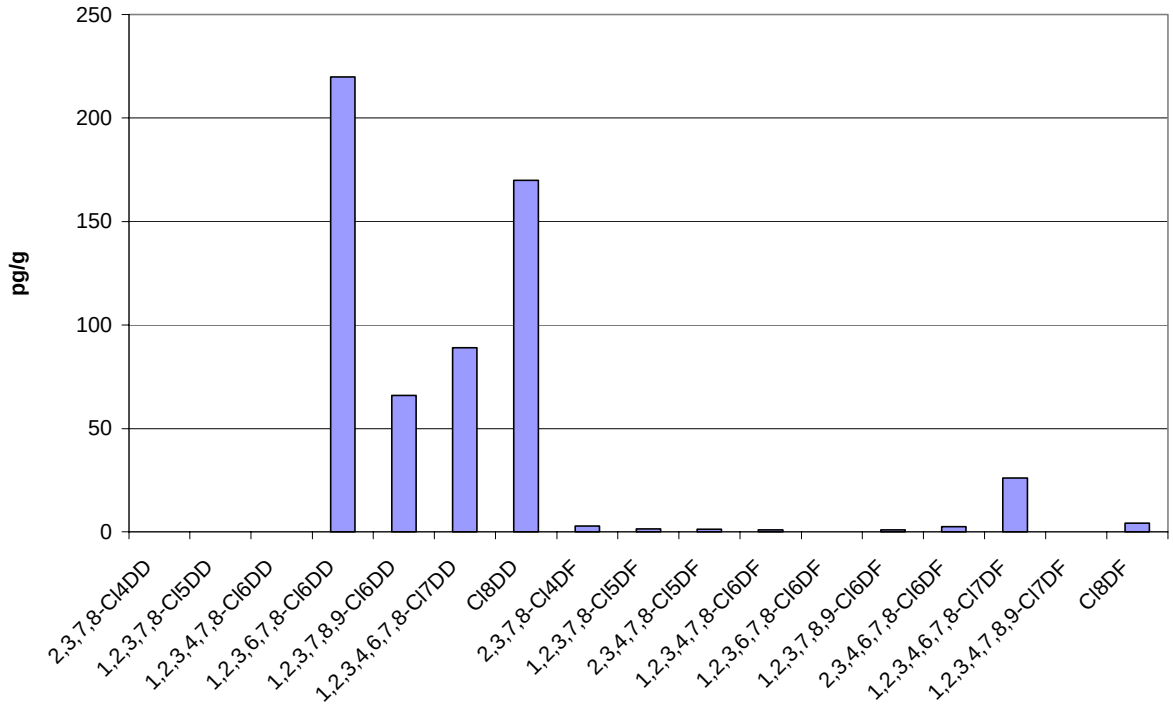

Figure 49: Example of a congener pattern in an low impacted leather sample

9.5 Summary Substances/Preparations/Products

The data for the compartment Substances/Preparations/Products cover a comparatively short period between 1992 and 1996. The samples contained therein are closely connected with the investigations of products and consumer goods treated with chemicals where high PCDD/PCDF concentrations were detected. These are mostly chlorochemicals such as chlorophenols or chlorobenzenes. Though the production of these chemicals has been prohibited in Germany already for many years (PCP decree of 1989) or stopped by a self-obligation of chemical industry evidence for the use of these chemicals in consumer goods is still to be found until the mid-90-ies. Examples of textiles and leather samples are a proof of the necessity of monitoring.

10 SUMMARY

10.1 Objective of the database DIOXINS

Taking as a basis a Bundesrat decision (Bundesrats-Drucksache 140/90), the Federal and Laender Ministers for the Environment decided at their 34th Conference at the beginning of the 90s to set up a Government/Laender working group on DIOXINS. This working group was commissioned inter alia with initiating and coordinating measuring programs, and documenting and evaluating data. In order to accomplish this task, a central database DIOXINS was set up at the Federal Environmental Agency. The transfer of environmental data on pollution from dioxins - as part of an administrative agreement on data exchange in the environmental sector between the government and the Laender - serves primarily

- as an overview of the level of environmental pollution,
- as an indispensable basis for environmental goals and setting priorities,
- as a technical basis in order to derive implementable and scientifically founded benchmarks and limit values, and not least
- as a means of observation the success of political activities.

10.2 Procedure

As a first step, the Government/Laender working group on DIOXINS elaborated data collection forms for documenting the data for the various compartments. The forms can be obtained from the Federal Environmental Agency. Data were primarily collected by the Laender, the results were transferred to the data collection forms and forwarded to the operators of the database. The Federal Environmental Agency is responsible for running the database in cooperation with the former Federal Institute for Consumer Health and Veterinary Medicine.

The focus of the 3rd report of the Government/Laender working group on DIOXINS is on the evaluation of the **117 measuring programs** now contained in the database DIOXINS **with a total of 9,469 datasets**. The compartments covered were as follows: foodstuffs – 3,504, feedingstuffs – 128, sediments - 212, biota - 409, water discharges - 11, dust - 97, ambient air – 1,438, emission - 205, deposition - 254, indoor air - 1, soil terrestrial – 2,525, soil subhydric - 44, products/mixtures - 235, wastes/recoverable materials/residues – 406.

Food and human data were not contained in this report. These data can be found **in the 4th report** of the Government/Laender working group on DIOXINS.

Evaluation of the data was performed along the lines: information on projects and number of samples, and corresponding to the goals of the database, for temporal and spatial trends as well as for non-impacted and impacted areas, respectively. Statistics of the results for all compartments include the median, 10 and 90 percentiles as well as minimum and maximum concentrations. Numbers are given in I-TEq as the common basis (without inclusion of the detection limit for non-quantifiable congeners when calculating the TEq). In individual cases, PCD data were also evaluated.

10.3 Results

The data for **ambient air** samples “without special impact” proved seasonal variations and a temporal declining trend in the peak concentrations during wintertime **since the early 1990s**. The **concentrations** went down by **half**, but have leveled off since approximately 1994. At sampling locations close to cities or in **urban centers**, the concentrations range from **130 to 160 fg I-TEq/m³ in winter (90 P)**; **medians** are around **50 fg I-TEq/m³**. **Rural areas** in winter as well as all other regions in summer have 90 percentile concentrations of **50 fg I-TEq/m³** and **medians <20 fg I-TEq/m³**. In most cases, ambient air samples labeled “special impact” do not differ from the non-impacted samples.

The data for the **deposition** samples “without special impact” confirmed the seasonal variations and the temporal declining trends of the peak winter concentrations as obtained for the ambient air samples. The **peak concentrations** of the most recent data sets have **reduced by a factor of about five when compared with maximum concentrations during the winter 1993/94**. In urban centers the 90 percentile is <20 pg I-TEq/m² d with winter concentrations <40 pg I-TEq/m² d; all other samples have a 90 percentile <10 pg I-TEq/m² d. Higher concentrations could not be confirmed for samples with the parameter “special impact“.

Within the **compartment Biota**, **spruce needles used as biomonitors** generally had **higher concentrations** after **winter exposure** (sampling in spring) when compared to the samples taken **in autumn**. Furthermore, a decrease of approximately 50% (90 percentile) in the PCDD/PCDF content was shown in the more recent samples (1997/1998) as compared to samples from 1992-1996. PCDD/PCDF concentrations in pine needles were around 0.5 ng I-TEq/kg dm (90 percentile) in 1997 and 1998.

In 1996, PCDD/PCDF concentrations in **Welsh ryegrass** from Bavaria were below 1.5 ng I-TEq/kg dm with a **median of 0.5 ng I-TEq/kg dm** (one exception). The few individual data from 1997 gave results below 0.5 ng I-TEq/kg dm.

2,500 samples were used for an evaluation of the **compartment Soil**. The contamination in soils showed a **median of below 20 ng I-TEq/kg dm**. The 90 percentile for all types of soils was <30 ng I-TEq/kg dm; top soils were <20 ng I-TEq/kg dm, and organic layers <50 ng I-TEq/kg dm. The Federal Soil Protection and Contaminated Sites Ordinance prescribes action levels to protect the pathway soil-humans: the maximum permissible concentration is 100 ng I-TEq/kg dm for children's playgrounds, 1,000 ng I-TEq/kg dm for residential areas, parks and leisure areas, and 10,000 ng I-TEq/kg dm for industrial and commercial areas. All the median concentrations and the 90 percentiles of all soil contained and assessed in the DB DIOXINE are below the respective action levels.

The compartment Waste/Recoverable materials/Residues is dominated by **sewage sludge** results. **PCDD/PCDF and PCB concentrations** in municipal sewage sludge have **declined since the early 1990s**. For PCDD/PCDF, the 90 percentile has been on the same level since 1995/97 and for PCB since 1993. Samples with “no special impact” have a **PCDD /PCDF concentration (90 P) <30 ng I-TEq/kg dm.** All other data sets in this compartment (industrial residues, filter dusts, sediments from wastewater discharges) were too small in number for evaluation.

The data for the **compartment Products/Preparations /Products** contain samples treated with dioxin-containing chemicals. With regard to the sometimes high PCDD/PCDF concentrations identified in textiles and leather, these were primarily cases of imported goods from countries where chemicals that have been banned for years in Germany (*e.g.* PCP or chloranil-based dyestuffs) are still in use and are being exported.

10.4 Conclusions and Outlook

The priority task of elaborating an overview of the level of pollution from dioxins has been fulfilled with the aid of the DIOXINE database. The evaluations of approx. 10,000 data sets were carried out primarily with regard to temporal and spatial trends, and with differentiation between impacted areas and non-impacted areas.

The evaluation of the data collected over the past ten years documents the success of the environmental protection measures that have entered into force to reduce the input of dioxins into the environment. New inputs of dioxins into the environment have been drastically reduced.

The median levels that were determined for the individual compartments could be used as comparative values in order to make statements on possible environmental impacts in the case of incidents.

The Government/Laender working group on DIOXINS proposes continuing the measuring programs in the Laender. The consequence of this continuation is that the work of the database DIOXINS will also be sustained; the data will be published.

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12 APPENDIX

12.1 Measuring programs and executing institutions (up to September 2000)

The following institutions carried through the measurement and research projects indicated in in Table 13. The results have been collected in the database DIOXINS as of September 2000. In addition, numerous further measurements were carried out the data of which are not yet covered by the database DIOXINS as the measurements had to fulfill specific targets, sampling varied in the past and thus the results are comparable only to a limited scale.

Table 13 Survey of projects database DIOXINs (up to September 2000)

Institution	Measuring program
Bayrisches Geologisches Landesamt	Permanent soil monitoring Bavaria Organ. problematic substances (PCB and PCDD/F) in soils of Bavaria
Bayrisches Landesamt für Umweltschutz	Biomonitoring (Welsh ryegrass, green cabbage) in 6 locations Bioindication of airborne dioxin und furan by means of spruce needles
Bayrisches Staatsministerium für Landesentwicklung und Umweltfragen	Dioxin measurement in the ambient air in Bavaria
Bundesanstalt für Milchforschung	Dioxin research department BML
Bundesinstitut für gesundheitlichen Verbraucherschutz und Veterinärmedizin	Ilsenburg
Chemische Landesuntersuchungsanstalt Freiburg	Official foodstuff monitoring 1993 - 1995
Chemische Landesuntersuchungsanstalt Stuttgart	Official foodstuff monitoring 1993 - 1995
Chemisches Landesuntersuchungsamt Oldenburg	Dioxin reference measuring program Investigation in the surroundings of the former hazardous waste dump
Freie Hansestadt Bremen - Senator für Bau und Umwelt	Measuring program for the investigation of dioxin contamination in the municipality of Bremen

Freie und Hansestadt Hamburg Behörde f. Arbeit, Gesundheit u. Soziales	Investigation of the transfer of dioxins and funrans soil/feedingstuffs R&D Biomonitoring of the Elbe river
GAA Itzehohe, Lufthygienische Überwachung Schleswig - Holstein	Deposition measurements of PCDD + PCDF in 5 measuring points in Schleswig-Holstein
Hessisches Landesamt für Umwelt und Geologie	Hesse dioxin reference measuring program Permanent soil monitoring program of the Land of Hesse
Hessisches Ministerium des Innern und für Landwirtschaft, Forsten und Naturschutz	Hesse milk investigation program
Landesamt für Natur und Umwelt des Landes Schleswig-Holstein	Water monitoring Schleswig-Holstein Soil impact cadastre Schleswig-Holstein
Landesamt für Umwelt, Naturschutz und Geologie Mecklenburg-Vorpommern	Investigation of pollutants in surface sediments
Landesamt für Umweltshutz Saarland	Dioxin soil measuring system Saar
Landesamt für Umweltshutz Sachsen-Anhalt	Emission measurement reports of the Land of Saxony-Anhalt Dioxin measuring program of the Land of Saxony-Anhalt Small furnace emission investigation LAU Investigation of soil in the region Halle-Merseburg for PCDD/F

Landesanstalt für Umweltschutz Baden-Württemberg	Small gardens Friesenheim Island Extension of the waste heating and power station North, 4th waste heating boiler Environment study cupola Dioxin investigation in the urban area of Heidelberg Sampling in children's playgrounds Incident Composting plant Wieblingen Environment of the clinic waste incineration plant Environment of a car shredder Soil expertise for the UVU waste heating and power plant Ludwigsburg Former industrial sites and waste disposal sites in the municipality of Stuttgart Investigations in the meadows of the Enz-river Soil expertise for the 2nd hazardous waste incineration plant BW Soil investigation (inorg./org. pollutants) S-airport Weapon elimination service Sindelfingen Accompanying program for ambient air measurement
Landesanstalt für Umweltschutz Baden-Württemberg	Dioxin composting plants Environment of crematories Dioxin flood land Sediments Basic impact urban regions Dioxin sport and playgrounds Suspicion of cases of damage Dioxins agriculture Environment of sources of emission Permanent soil monitoring I, 1st sampling Dioxins forest soil Dioxins/SM load Friesenheim Island Investigations composting/humus plants Permanent soil monitoring I, 2nd sampling pollutant content roadsides

Landesumweltamt Brandenburg	Investigation program district Oranienburg Transfer investigations region of Eisenhuettenstadt
Landesumweltamt Nordrhein-Westfalen	Deposition investigation of the Land of NRW Investigation of biota, emissions, depositions and ambient air of the Land of NRW Water quality monitoring
Landesuntersuchungsamt für das Gesundheitswesen Südbayern	Official food quality surveillance hen's eggs 1995 - 1999 Official food quality surveillance 1996 - 1999 Official food quality surveillance collective milk 1998 Official food quality surveillance collective milk Official food quality surveillance collective milk 1992 Official food quality surveillance collective milk 1996 Official food quality surveillance hen's eggs 1992 - 1999 Official food quality surveillance -meat Food quality surveillance South Bavaria
Ministerium f. ländliche Räume, Landwirtschaft, Ernährung u. Tourismus Schleswig-Holstein	Investigation of soil and vegetable for PCDD, PCDF
Ministerium für Umwelt und Forsten Rheinland-Pfalz	Deposition measuring program of the Land of Rhineland-Pfalz
Ministerium für Umwelt, Natur und Forsten des Landes Schleswig-Holstein	Investigation of soil and vegetable for PCDD/PCDF
Chemisches Landes- und Staatliches Veterinäruntersuchungsamt Münster	Dioxin reference program
Niedersächsisches Umweltministerium	Investigation of the impact on soil, mussels and sediments from the region Wilhelmshaven Dioxin contamination of soil in the flood land of the Elbe-river
Sächsische Landesanstalt für Landwirtschaft	Official feedingstuffs surveillance Ambient air measuring program of the Free State of Saxony
Sächsisches Landesamt für Umwelt und Geologie	Dioxin reference measuring program

Senatsverwaltung für Stadtentwicklung des Landes Berlin	Investigation of pesticides from the GDR production for PCDD/F
Sozialministerium Baden-Württemberg	Cases of contaminated sites in Rastatt and Rheinfelden
Staatliches Amt für Umwelt Dessau-Wittenberg (Sachs.-Anh.)	Investigation of the environment of the copper mill Ilsenburg 1990 Study complex exploration Bitterfeld
Staatliches Umweltfachamt Bautzen (Sachsen)	Emission measuring program of the Free State of Saxony
Staatliches Umweltfachamt Chemnitz (Sachsen)	Emission measuring program of the Free State of Saxony
Staatliches Umweltfachamt Leipzig	Emission measuring program of the Free State of Saxony
Staatliches Umweltfachamt Plauen (Sachsen)	Emission measuring program of the Free State of Saxony
Staatliches Umweltfachamt Radebeul (Sachsen)	Emission measuring program of the Free State of Saxony
Thüringer Landesanstalt für Umwelt und Geologie	Chloroaromatics dioxin measuring program Thuringia
TÜV Hannover/Sachsen-Anhalt e.V., Niederlassung Halle	Emission measurement reports of the Land of Saxony-Anhalt
Umweltamt und Gesundheitsamt der Stadt Dortmund	Dioxin problems of the Krupp Hoesch sintering plant Westphalenhuette in Dortmund
Umweltbehörde Hamburg Fachamt für Umweltuntersuchungen	Investigation of deposition, water and sediments for the Hamburg dioxin reference measuring program Permanent soil monitoring of the Free and Hanseatic town of Hamburg
Federal Environmental Agency	Measuring points at the Elbe-river and its tributaries Nationwide analysis of sewage sludge for PCDD/PCDF Investigation of potential sources of PCDD/F in sewage sludge Investigation of potential environmental hazards in burning plastic Occurrence of PCDD/PCDF in cases of fire UBA measuring systems new Federal Laender UBA measuring systems old Federal Laender

University Bayreuth	Environmental pollution by dioxins Atmospheric input pathways and behaviour of PCDD, PCDF, PAC in a maize crop Fertilization by sewage sludge – input of PCDD, PCDF and PCB into the food chain Precipitation-dependent input of PCDD and PCDF into an urban sewerage system Investigation of emission pathways of polychlorinated dioxins and furans in chemical cleaning plants Dioxins und Furans in textiles and leather Investigation of the atmospheric input of PCDD PCDF into fodder plants Investigation of atmospheric pollution by PCDD, PCDF and coplanar PCB Environmental pollution by the atmospheric input of emissions from a waste incineration plant
University Bayreuth / Umlandverband Frankfurt	Sewage sludge surrounding area of Frankfurt/Main
University Bayreuth, chair of pedology and soil geography	PAC/PCB in urban soil of Bayreuth

12.2. Research project and recipient

The evaluation of the data collected and put into the database DIOXINS predominantly by the Laender (Tab. 19, p. 124) is carried out in the framework of the research and development projects mentioned hereinafter which were financed by the environmental research plan of the Federal Ministry for the Environment. The final reports provided the basis for the present 3rd report of the Government/Laender working group on DIOXINS.

Updating of the database DIOXINS by means of latest Laender data
Research code: 298 91 229/02

FIEDLER, ROTTLER
Verein zur Förderung der Umweltforschung, - Erziehung und Öffentlichkeitsarbeit e.V. an der Universität Bayreuth (Association for promotion of environmental research, education and public relations Reg. Association at the University Bayreuth)

WOLTER
debis Systemhaus sfi - Systemhaus für Informationsverarbeitung GmbH, Geschäftsstelle Berlin (dv-Teil Lebensmittel und Konzept Massenfluss)
(System house for information stepsing Ltd – Department Berlin (dv part foodstuffs and mass flow concept)