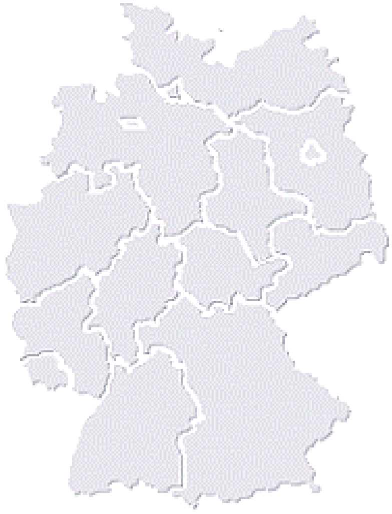
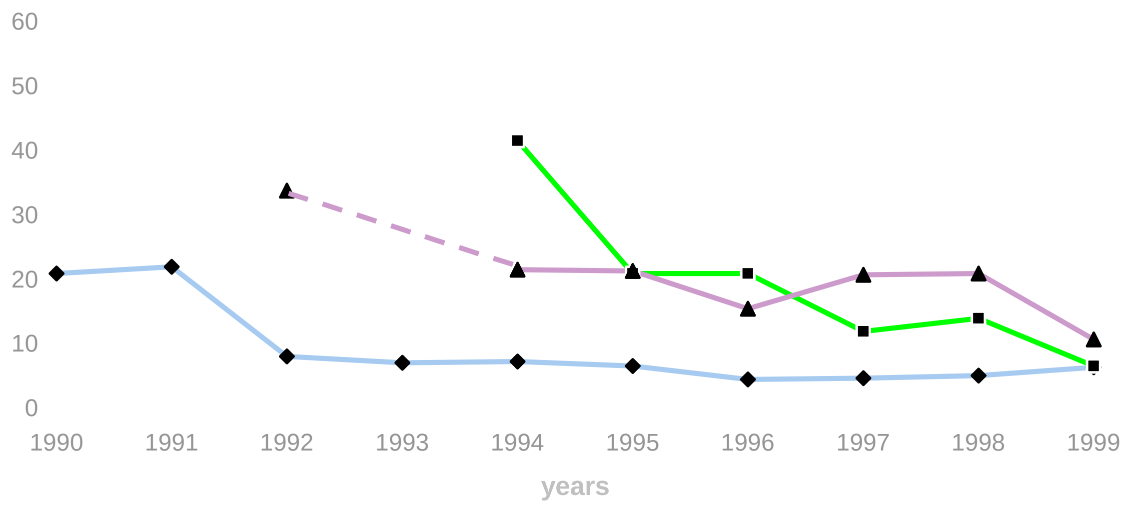
GOVERNMENT/LAENDER WORKING GROUP ON DIOXINS



DIOXINS

Data from Germany

Dioxin reference measuring program
4th Report of the Government/Laender working group on Dioxins (Germany)



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CONTENTS

List of Figures

List of Tables

1. Targets of the Dioxin Reference Measuring Program

The dioxin reference measuring program is founded on a decision taken by the 37th conference of Federal and Laender Ministers for the Environment of the Federal Republic of Germany, 21-22 November 1991. At that time, there were calls for further studies to improve the available data on dioxins in soil, water, air, foodstuffs and feedingstuffs, for the initiation and co-ordination of future government and Laender measuring programs and for a centralized documentation and evaluation of the results. To implement this it was, inter alia, agreed within the framework of the Government/Laender working group on DIOXINS to carry out a nationwide measuring program of several years duration. By observing the dioxin contamination in the environment over a longer period it should be possible to make statements on the trend. In particular, the effects of measures taken to reduce dioxin emissions on ambient air, dioxin deposition, contamination of foodstuffs and feedingstuffs and finally on human milk as the last member of the food chain were to be determined.

In the early 90-ies already numerous analytical results were available. The data available from the environmental area, however, were, for the most part, results of studies carried out as a result of cases of damage or specific emission/ambient air measurements. Therefore only a few findings on the global contamination beyond suspected sites were available. In addition, the measured data of investigations carried out in many places were scarcely comparable with each other as neither the modalities of sampling nor the measurements were harmonized before. Nethertheless the Government/Laender working group on DIOXINS tried to sum up approx. 10,000 samples so far collected in the database DIOXINS and to evaluate them to make comments on the contamination of environment by dioxins. This work included in the 3rd report, however, showed clearly the weak points of the previous data collection and documentation and were the reason why the Government/Laender working group on DIOXINS after harmonizing sampling, analyzing and documentation as well as co-ordination of reference analyzes carried through a dioxin reference measuring program.

With the dioxin reference measuring program started mid-1994 the following aims have been pursued – supplementary to the collection of the „data on environmental pollution by dioxins“ (3rd report of the Government/Laender working group on DIOXINS):

- drafting an overall picture of the contamination of humans and environment by bioxins,
- making comments on the trends of contamination of environmental media, biota and humans by dioxins and thus
- controlling and documenting the success of measures initiated to reduce the input of dioxins into environment and the exposure of humans to dioxins

2. Participation

The Laender Baden-Wuerttemberg, Bavaria, Hesse, Hamburg, North Rhine-Westphalia, Lower Saxony, Saxony and Thuringia and the Federal Environmental Agency (UBA) and the former Federal Institute for Consumer Health and Veterinary Medicine (now reorganized in the Federal Institute for Risk Assesment an the Federal Office for Consumer Protection an Food Safety) participated in the dioxin reference measuring program. North-Rhine-Westphalia took over the co-ordination of the dioxin reference measuring program.

Based on already existing programs the Laender selected individual media from the compartments to be investigated. In Table 1 Laender participating in the investigations of the respective compartments are listed:

Table 1: Federal Laender participating in the dioxin reference measuring program

Compartment		BW	BY	HE	HH	NRW	NI	SN	TH
Air	Ambient air			X				X	X
	Deposition			X	X	X			X
Soil		X	X	X	X			X	
Dairy products and raw milk		X	X	X		X	X		
Foodstuffs (except milk)		X	X				X		
Bioindicators, plant foodstuffs			X				X		
Sediment and suspended matter					X	X			
human millk[1]		X	X			X	X		
Blood lipids		X							

1) Some data from Schleswig Holstein and former BgVV are also available.

Before starting the measurements comparative studies were predominantly carried out for the individual compartments. Always one of the participating Laender and in one case the UBA took over the co-ordination and evaluation of the comparative analyses.
The comparative analyses were co-ordinated by
the Federal Environmental Agency for **soil**

- North Rhine-Westphalia for **dairy products and raw milk**
- Lower Saxony for **foodstuffs,**
- Hamburg for **sediments and suspended matter and**
- North Rhine-Westphalia for **human milk and blood lipids.**

3. Data collection

The database DIOXINS in the UBA was used as collection centre for all dioxin data including data from the foodstuff domain for data collection in the framework of the dioxin reference measuring program. Data collection for the **cow's milk** and **human** areas is carried out by the BgVV and described in Chapters 6.5 and 6.6.

For the collection of the measured data in the Laender the UBA and the former BgVV, together with the Laender, have prepared media-specific collection forms.
To fix the modalities of data transmission and data use as binding an Administrative Agreement on Data Exchange was concluded between the Government and the Laender. This agreement forms part of the General Administrative Agreement on Data Exchange.

4. Data protection and data security

For the data and information included in the central database DIOXINS the Federal Environmental Agency assumes the responsibility for data protection and security as to the storage, processing and transmission of these data to third persons.
As the transmitted data concern partly sensitive data (e.g. data of the seven-digit northing and easting values for permanent soil monitoring areas, characterizing of the emission source) respective protection mechanisms are envisaged for the data input and storage.

Appropriate recommendations were given in a legal expertise prepared by the Institut für Umweltrecht (Institute of Environmental Law) Bremen by order of the Federal Environmental Agency (s. also UBA – Textreihe 15/97). These recommendations have been implemented by the UBA in its data security conception for the client-server application.

5. Data input and data inventory

The responsibility for the data input for the compartments soil, air, bioindicators, sediments and suspended matter lies with the Federal Environmental Agency. The Federal Institute for Consumer Health and Veterinary Medicine is competent for the compartments dairy products and raw milk, foodstuffs, human milk and blood lipids.

A co-ordinating centre as described in § 3, subpara. 3 of the Administrative Agreement on Data Exchange in the Environmental Field proves to be very beneficial for the exchange of data with the Laender. By this structure the Laender participating in the dioxin reference measuring program support the co-ordinated data transfer between Government and Laender.

5.1 Survey of the data inventory relating to soil, air, sediments and bioindicators

13 monitoring programs of the Laender form the basis of evaluation for the compartments soil, air (ambient air, deposition), water (sediments/suspended matter) and bioindicators.Table 2 (comp. also Table 1) gives a survey of the respective programs.

For the study programs basically 17 individual congeners and the TEq (I-TEq, WHO-TEq) are available in the database. In addition, data relating to PCB, including coplanar PCB, may be included in the database. An extension of the substance spectrum by further analyses is possible at any time. The I-TEq according to NATO-CCMS and the WHO-TEq are automatically calculated via algorithms, with always the full and half of the limit of determination and the limit of determination equal to zero being included in the calculation. All subsequent data evaluations refer to the automatically calculated I-TEq in the database. Due to the present data inventory in evaluations emphasis was laid on the substance group of polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF) (chemical structure of selected PCDD and dibenzofurans (s. Fig. 1) (see Chapter 6). The number of data sets for the substance group of PCB does not involve a representative number of random samples and is not considered in the evaluation.

Table 2: Survey of the measuring programs soil, air, sediments and bioindicators

Data supplying authority	Name of the measuring program
Hessisches Landesamt für Umwelt und Geologie	Hesse dioxin reference measuring program „air“
	Permanent soil monitoring program of the Land of Hesse
Sächsisches Landesamt für Umwelt und Geologie	Dioxin reference measuring program „soil“
	Dioxin reference measuring program „ambient air“
Umweltbehörde Hamburg, FA für Umweltuntersuchungen	Permanent soil monitoring program Hamburg
	Deposition and sediment investigations for the dioxin reference measuring program
Landesanstalt für Umweltschutz Baden – Wuerttemberg	Permanent soil monitoring program
Landesumweltamt Nordrhein-Westfalen	Deposition investigation of the Land of NRW
	Water quality monitoring system
Thüringer Landesanstalt für Umwelt	Chloroaromate measuring program of the Free State of Thuringia
Bayerisches Geologisches Landesamt	Organ. problematic substances (PCB and PCDD/F) in soil in Bavaria
Bayerisches Landesamt für Umweltschutz	Ambient air ecological measuring system/bioindication
Staatliches Lebensmitteluntersuchungsamt Oldenburg	Program of investigation for green cabbage
Chemisches Landes- und staatl. Veterinäruntersuchungsamt NRW	Plant foodstuffs, bioindicators

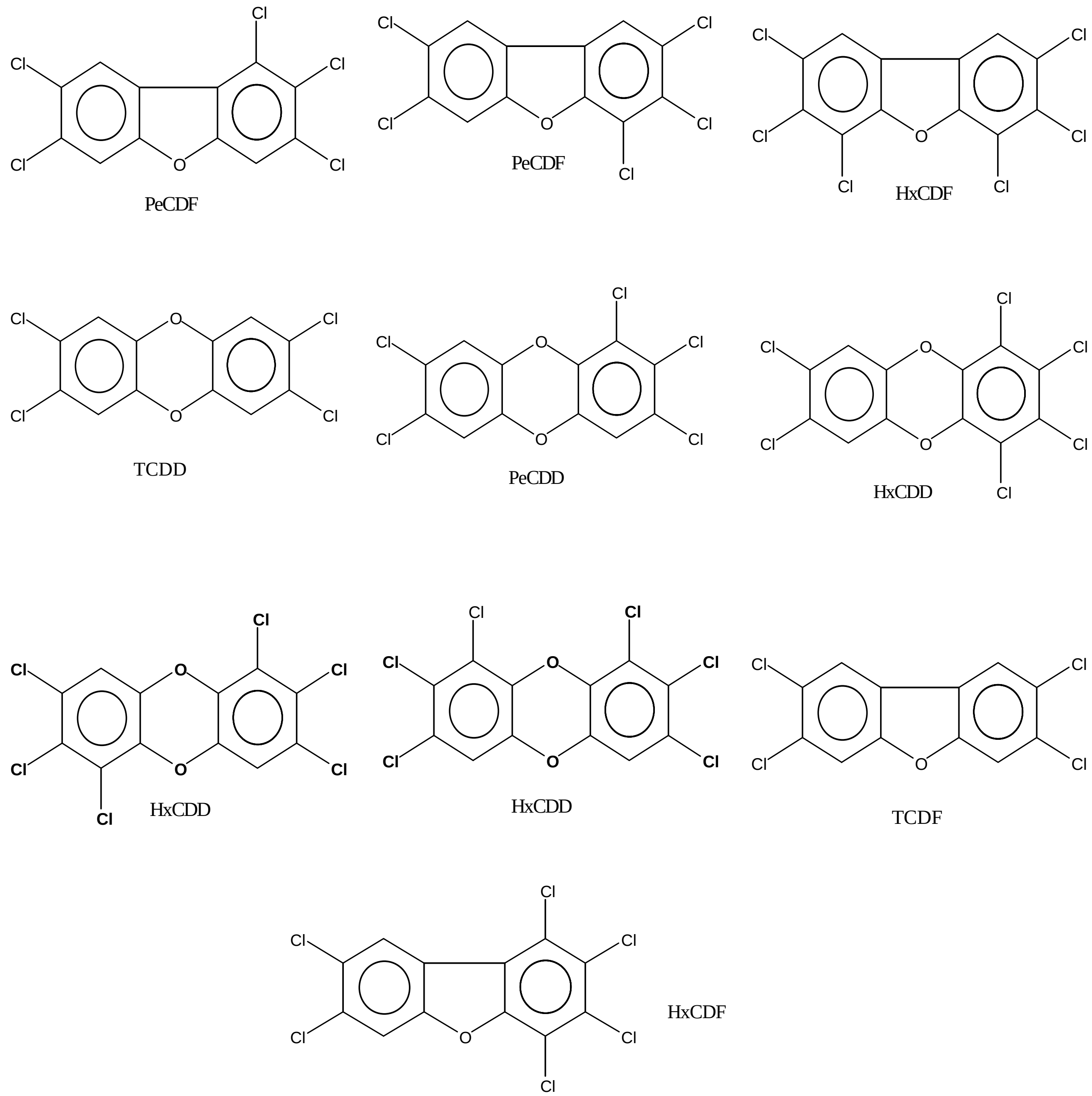


Fig. 1: Chemical structure of selected polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDF) Notably congeners chlorosubstituted in the positions 2,3,7 and 8 are toxic.

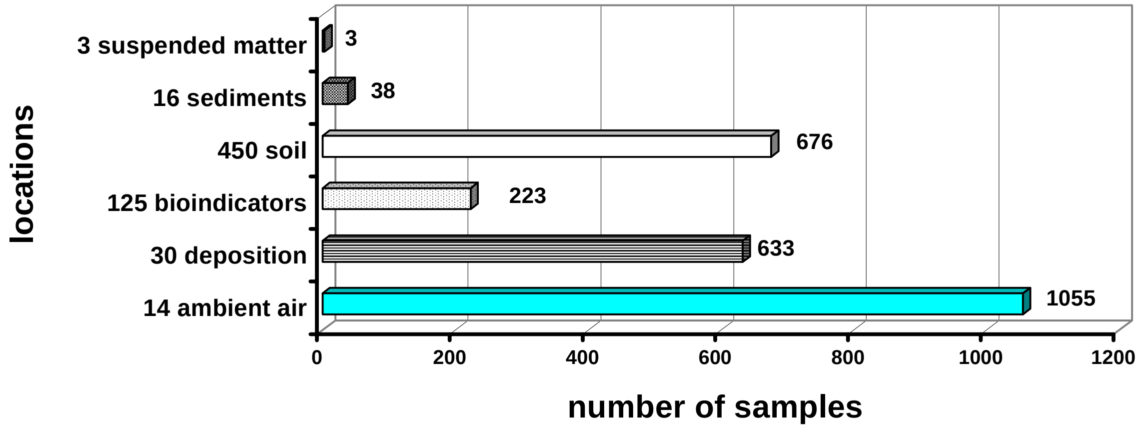


Fig 2: Survey of the number of locations and samples, classified according to compartments (except cow's milk, foodstuff samples and samples from the human area) until September 2000

5.2 Survey of the data inventory relating to foodstuffs

Data from the following measuring programs of the Laender are available for the dioxin reference measuring program:

Table 3: Survey of the foodstuff measuring programs

Data supplying authority	Name of the measuring program
Chemisches und Veterinäruntersuchungsamt Freiburg	Official foodstuff monitoring
Landesuntersuchungsamt für Gesundheitswesen Südbayern	Official foodstuff monitoring
Staatliches Lebensmitteluntersuchungsamt Oldenburg	Dioxin reference measuring program
Hessisches Landesamt für Regionalentwicklung und Landwirtschaft Kassel	Dioxin reference measuring program
Chemisches Landes- und Staatliches Veterinäruntersuchungsamt Muenster	Dioxin reference measuring program

6. Evaluation

The evaluation is carried out specifically for the compartments:

6.1 Air
6.2 Soil
6.3 Sediments and suspended matter
6.4 Bioindicators, plant foodstuffs
6.5 Dairy products and raw milk
6.6 Human data

Data on the respective properties are required for the evaluation of the data as to time, space and content from the measuring programs. The following data from the describing information documented on the basis of the collection forms for the individual compartments are considered for the subsequent evaluation:

- Characterization of the location (urban area, urban fringe, rural area)
- Method of sampling and analyzing
- Reference to time
- Number of samples

In particular, temporal and spatial components are considered in the evaluation as the target of the reference measuring program is primarily directed to trend data and the representation of the overall situation.

6.1 Air

Dioxins and furans are contained in the atmosphere basically bound to particles; only at summer temperatures a smaller share of the molecules may be present in the gaseous phase in the case of low-chlorinated dioxins and furans. The attachment of dioxins and furans to dust particles means that dioxins and furans – as dust particles – have only a retention time in the atmosphere limited to a few days (retention time of dust with a particle diameter of 1 - 10 µm: 0.4 - 4 days, retention time of fine dust smaller than 1 µm: 4 - 40 days) and are then discharged from the atmosphere by deposition. In the case of fine dust – as e.g. Sahara dust events show – a long-distance transport on a global scale is possible – in spite of the limited retention time.

Sampling during measurement of dioxin concentrations in air is carried out in a modified form my means of the measuring method tested in measuring suspended dust and is described in the VDI guideline 3498. The adsorption unit used for sampling consists of a glass fibre filter with topped polyurethane foam; this combination ensures that the particle-bound as well as the gaseous fractions of the dioxins are collected in the sampling volume which passed the adsorption unit. To achieve a sufficient detection limit, in general, sampling volumes of about 1.000 m^3 are required; only further refined analytical methods allowed, in the mean time, to work also with smaller sampling volumes.

As during dust precipitation measurement sampling is carried out during dioxin deposition measurement according to the Bergerhoff method (VDI guideline 2119, page 2 or VDI guideline 2090, page 1 of December 1999). When applying this measuring method a standardized glass vessel is exposed for more than 30 days and the dust mass sedimented in the receiver is subsequently weighed or analyzed for substances contained such as dioxins. To obtain the sampling mass required for analyzing dioxins a few sampling vessels have to be exposed simultaneously during dioxin sampling.

Proceeding on the fact that all samples of the compartment air (ambient air/depositions) available for the dioxin reference measuring program are subjected to a uniform procedure from sampling up to analyzing, the data in their entirety may be used for a statistical evaluation. With the emphasis of the dioxin reference measuring program lying on trend data this aspect is the priority of the evaluations carried out. Data with the full limit of determination were used for statistical evaluations.

Attention should be drawn to the fact that the present dioxin concentration or dioxin deposition measurements are not representative random samples for the situation of impact in the respective Federal Land but record, in general, known or suspected priorities of impact. Only in exceptional cases – e.g. to obtain reference values as evaluation aid – dioxin measurements were carried out also in emission-distant locations.

6.1.1 Ambient air

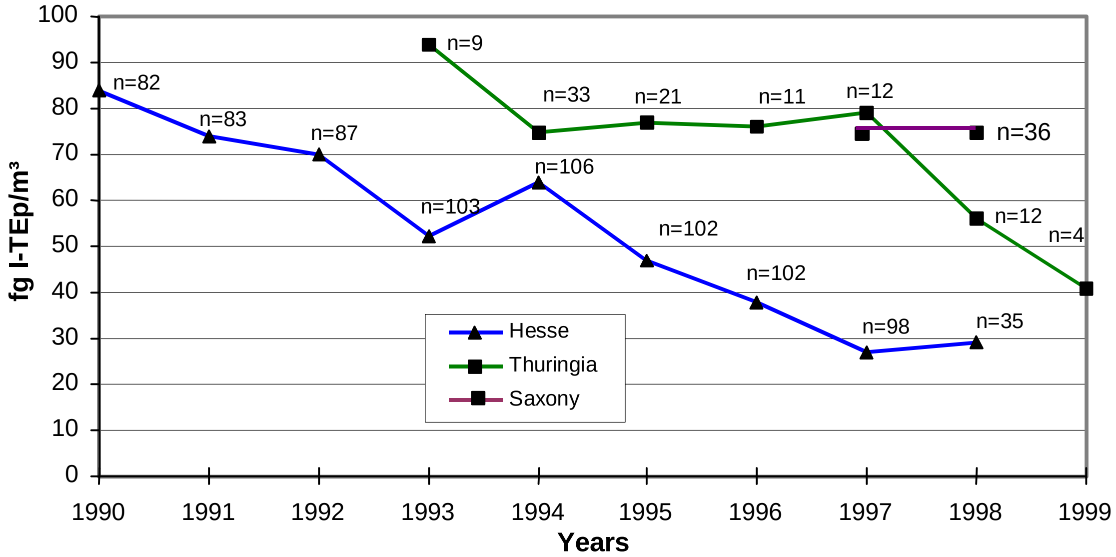
Table 4 gives a survey of the data inventory of concentration measurements, with the data being classified by Federal Land, characterization of locations, sampling period and number of samples.

To show the distribution of the data inventory onto the period of measurement the annual averages and the number of the measured values available per Federal Land are plotted as time series in Fig. 3 – classified by Federal Laender. These annual averages are in a range between 20 and 95 fg I-TEq/m^3. Apart from the distribution of the data inventory by years of measurement Fig. 3 shows a clear decline of the dioxin contamination since the series of measurements have been started in the Federal Laender Hesse and Thuringia.

Table 4: Survey of the locations of dioxin and furan concentration measurement (method VDI 3498)

Location ID	Federal Land	Characterization of location	Time series	Number of samples
328	Hesse	urban fringe, industrial area	Jan 93 - Dec 99	116
1959	Hesse	urban area with industry nearby	Jan 98 - Dec 99	19
327	Hesse	urban fringe (background station)	Oct 89 - Dec 99	191
329	Hesse	urban area, industrial area	Oct 89 - Dec 99	191
330	Hesse	urban fringe	Oct 89 - Dec 99	176
331	Hesse	urban area, industrial area	Oct 89 - Dec 99	151
486	Hesse	urban area, industrial area	Jan 96 - Dec 99	52
601	Saxony	urban area	Oct 97 - Jan 98	15
602	Saxony	urban area	Oct 97 - Jan 98	12
603	Saxony	urban area	Oct 97 - Jan 98	9
478	Thuringia	urban fringe, traffic load	Oct 93 - Dec 99	78
483	Thuringia	urban fringe, traffic load	Oct 93 - Oct 94	12
484	Thuringia	urban fringe, traffic load	Oct 93 - Oct 94	12

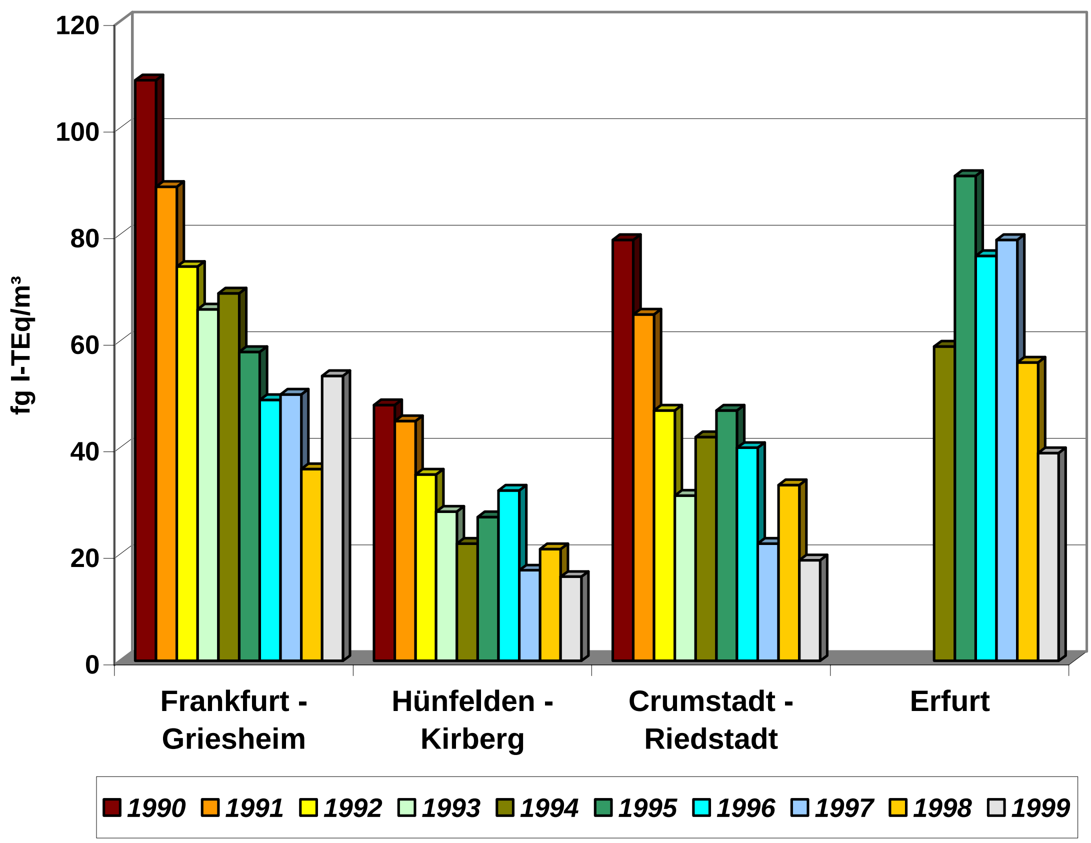
Fig. 3: Annual averages of the PCDD/F concentrations in the Federal Laender Hesse, Thuringia and Saxony in fg I-TEq/m³ air, evaluation period 1990 to 1999



To work out purposefully the trend of the dioxin contamination by means of homogeneous series of measurement the time series of the annual averages for the four measuring stations with homogeneous series of measurement covering many years is represented in Fig. 4. These four measuring stations show extremely different characterizations of the locations.

Erfurt:	city station registering the general environmental impact in the urban area characterized by road traffic and heating of buildings
Frankfurt/M.–Griesheim:	urban station in a location influenced by industry, in addition influence by a site contaminated by dioxins
Crumstadt – Riedstadt:	station in a rural area of the Rhein-Main urban area, yet with industry in the surrounding
Huenfelden – Kirberg:	station in an open territory in the low-emission posterior Taunus (location for background concentration).

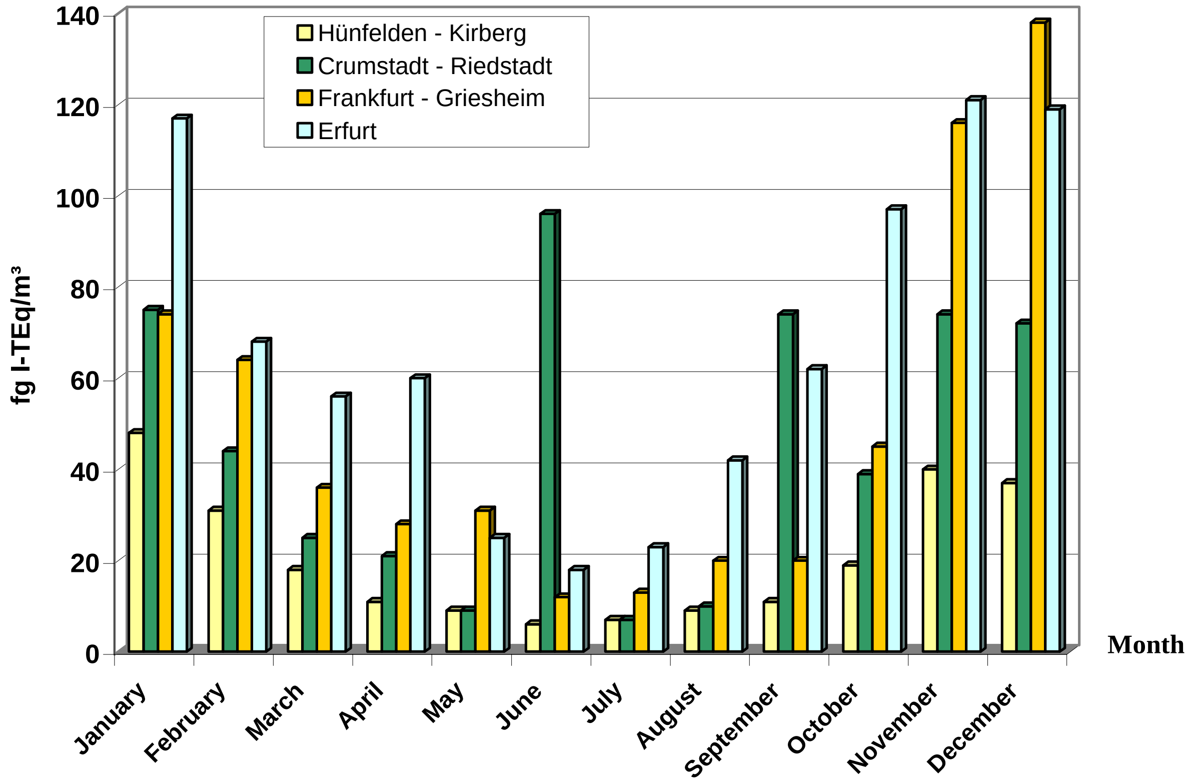
Fig. 4: Trend of the PCDD/F concentrations in selected locations of the Federal Laender Hesse and Thuringia, represented by means of the annual averages; evaluation period 1990 - 1999



In spite of this differing characterization of the locations the decline of the dioxin contamination developed similarly in all four locations reaching e.g. at the station Huenfelden-Kirberg a decline of more than 50 % between 1990 and 1998. This is an evidence of the fact that the dioxin contamination has not only dropped in the focal areas of impact – i.e. where emission-reducing measures have been taken in individual plants - but also generally in the field of background concentration. This trend of dioxin contamination should be taken into consideration when comparing the dioxin concentrations of the different years; the annual average of dioxin concentration at the background station Huenfelden-Kirberg was e. g. higher in 1990 than the annual average at the station Frankfurt/M.-Griesheim for the year1998.

The dioxin concentration of the atmosphere shows fluctuations from day to day caused by changing meteorological conditions, air mass properties (maritime, continental) or the emission conditions of the dioxin emitting plants; but apart from that a pronounced seasonal annual variation of the dioxin concentration is to be observed. Late in autumn and in winter the number of measured values exceeding the annual averages is higher than in summer (s. also 3rd Report of the Government/Laender working group on DIOXINS). Fig. 5 shows the average variation of the dioxin concentration for the four measuring stations Frankfurt/M.-Griesheim, Riedstadt, Huenfelden-Kirberg and Erfurt for the evaluation period between 1994 and 1998 based on the monthly average levels.

Fig. 5: Annual variation of the PCDD/F concentrations at measuring stations of the Federal Laender Hesse and Thuringia, calculated on the basis of monthly averages; evaluation period 1994 - 1998

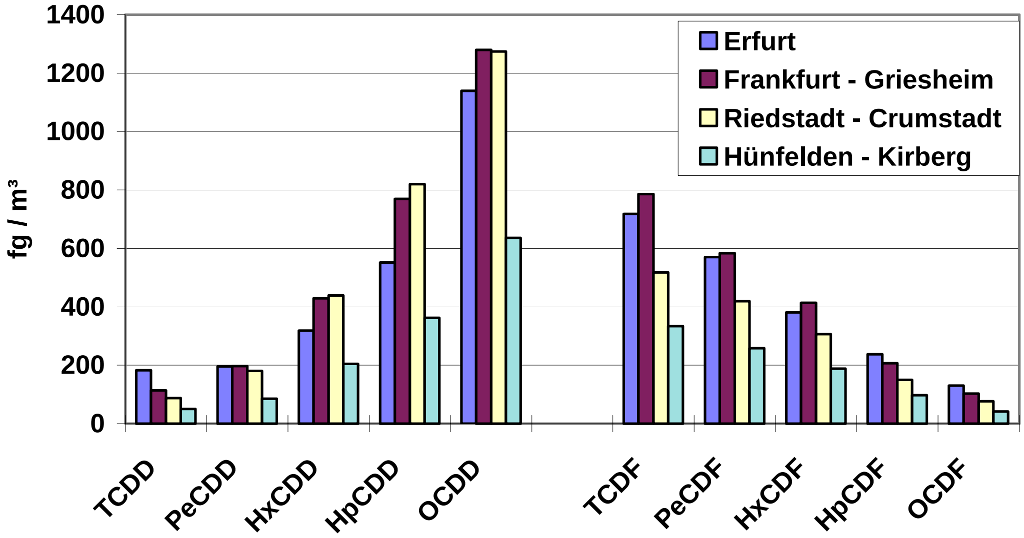


The annual variation of the dioxin concentration is very clearly shown at the stations Frankfurt/M.-Griesheim and Huenfelden-Kirberg. Also the values measured in Erfurt show this seasonal trend. At the station Riedstadt the calculated average annual variation is marked by a clearly increased concentration value in June and September 1994; when removing these two values the annual variation would be comparable with the annual variations of the two other stations. This reminds of the fact that in dioxin concentration measurements the data groups are very small, from a statistical view, so that an individual „runaway" caused by individual local disturbing effects will have still clear effects also in the case of averaging evaluations.

Since 1990, the beginning of the dioxin concentration measurements, with the dioxin contamination declining at the measuring stations the amplitude of the annual variation of the dioxin concentration was reduced, too.

The congener profiles of dioxins and furans determined by emission measurement show partly very differing structures, with either dioxins or furans dominating or individual congeners attracting attention. In the individual case congener profiles may help to detect the polluter causing an increased dioxin contamination of air. Nevertheless, - except in the direct environment of relevant dioxin emission sources – the congener profiles determined by concentration measurement in the atmosphere are surprisingly uniform. Fig. 6 shows examples of average conditions, with the congeners being combined to groups of homologues.

Fig. 6: Homology profiles of the PCDD/F concentrations in selected locations in Hesse and Thuringia, evaluation period: 1990 - 1998



It is characteristic that the dioxin concentrations grow from tetradioxins to octadioxin, whereas furan concentrations drop from tetrafurans to octafuran. By normalizing congener or homology profiles with the cumulative value for all dioxins and furans the measured values with a deviating congener profile may be clearly worked out.

The dioxin contamination of the atmosphere is the starting point of transfer of dioxins to the individual compartments. Characteristics of the dioxin concentration in the atmosphere such as the composition of the congener profile with regard to the processes of transfer between the compartments and contamination pathways should be recognizable also in other environmental compartments if the dioxin input from the atmosphere contributes essentially to an increase of the dioxin contamination of the compartment under consideration.

6.1.2 Deposition

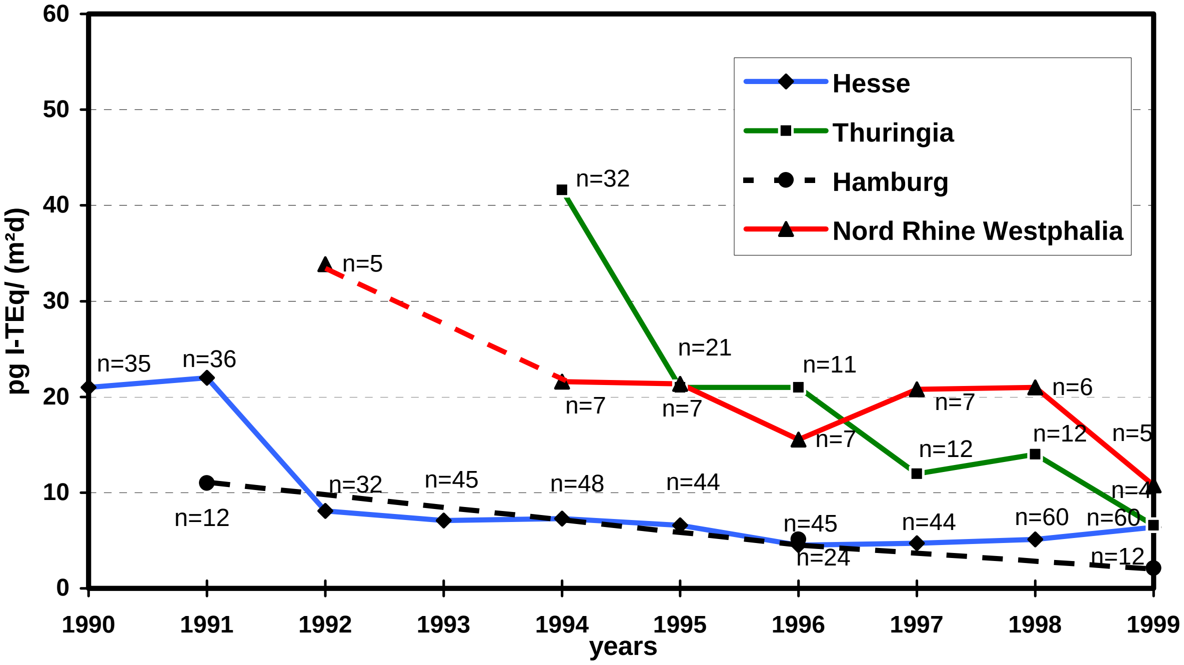
Table 5 gives a survey of the data inventory, classified by Federal Land, characterization of the location, sampling period and number of samples. In addition, the annual averages of the deposition values available for each Federal Land are compiled in Fig. 7 depending on the year of collection. The annual averages are to describe the data inventory of the respective Federal Land in a first overview. Here, we should be aware of the fact that values measured for various purposes during a year, in general, do not constitute a homogeneous data group and, in particular, no representative random sample to assess the average situation of contamination in the respective Federal Land.

Attention should be drawn to the fact that the numbers of samples taken in the various Federal Laender or in the framework of the various annual measuring programs given in Table 5 are not directly comparable with each other as the exposure periods of the samples were not equal.

Table 5: Survey of the locations of dioxin and furan deposition measurement

StID	Federal Land	Characterization of the location	Time series	Number of samples
1900	Hamburg	urban area (close to industry)	Jan 90 - May 91	6
1901	Hamburg	urban fringe (background station)	Oct 90 - Oct 91	6
1982	Hamburg	urban area (close to industry)	Jan 96 - Dec 96	12
1981	Hamburg	urban area (background station)	Jan 96 - Dec 96	12
1983	Hamburg	urban area (close to industry)	Jul 99 - May 00	6
1984	Hamburg	urban area (background station)	Jul 99 - May 00	6
357	North Rhine-Westphalia	urban area, traffic load	Nov 91 - Oct 92 Nov 93 - Dec 99	7
353	North Rhine-Westphalia	urban area	Nov 93 - Dec 94	6
354	North Rhine-Westphalia	urban area	Jan 96 - Dec 99 (annual average 96 without Aug and Sep)	4
356	North Rhine-Westphalia	urban area	Nov 91 - Oct 92 Nov 93 - Dec 97	5
358	North Rhine-Westphalia	urban area	Nov 91 - Oct 92 Nov 93 - Dec 97	5
359	North Rhine-Westphalia	urban area	Jan 97- Dec 99	3
352	North Rhine-Westphalia	urban area, traffic load	Nov 93 - Dec 99	6
362	North Rhine-Westphalia	urban area	Nov 91 - Oct 92 Nov 93 - Dec 99	7
355	North Rhine-Westphalia	urban area	Nov 91 - Okt 92 Nov 93 - Dec 99	7
487	Hesse	urban area, industrial area	Jan 96 - Jan 98	23
1960	Hesse	urban area close to industry	Jan 98 - Jan 99	12
323	Hesse	urban fringe (background station)	Sep 89 - Jan 99	104
324	Hesse	urban area, industrial area	Jan 93 - Dec 95	34
325	Hesse	urban area	Oct 89 - Jan 99	107
1961	Hesse	urban area, industrial area	Jan 98 - Jan 99	12
326	Hesse	urban area, industrial area	Okt 89 - Jan 99	106
481	Thuringia	urban fringe, traffic load	Feb 94 - Apr 99	62
482	Thuringia	urban fringe, traffic load	Oct 93 - Oct 94	12
485	Thuringia	urban fringe, traffic load	Oct 93 - Oct 94	12
489	Thuringia	urban area, traffic load	Oct 94 - Oct 95	12

Fig. 7: Annual averages of the PCDD/F deposition in the Federal Laender Hesse, Thuringia, Hamburg and North Rhine -Westphalia pg I-TEq/(m^2 d) evaluation period 1990 to 1999

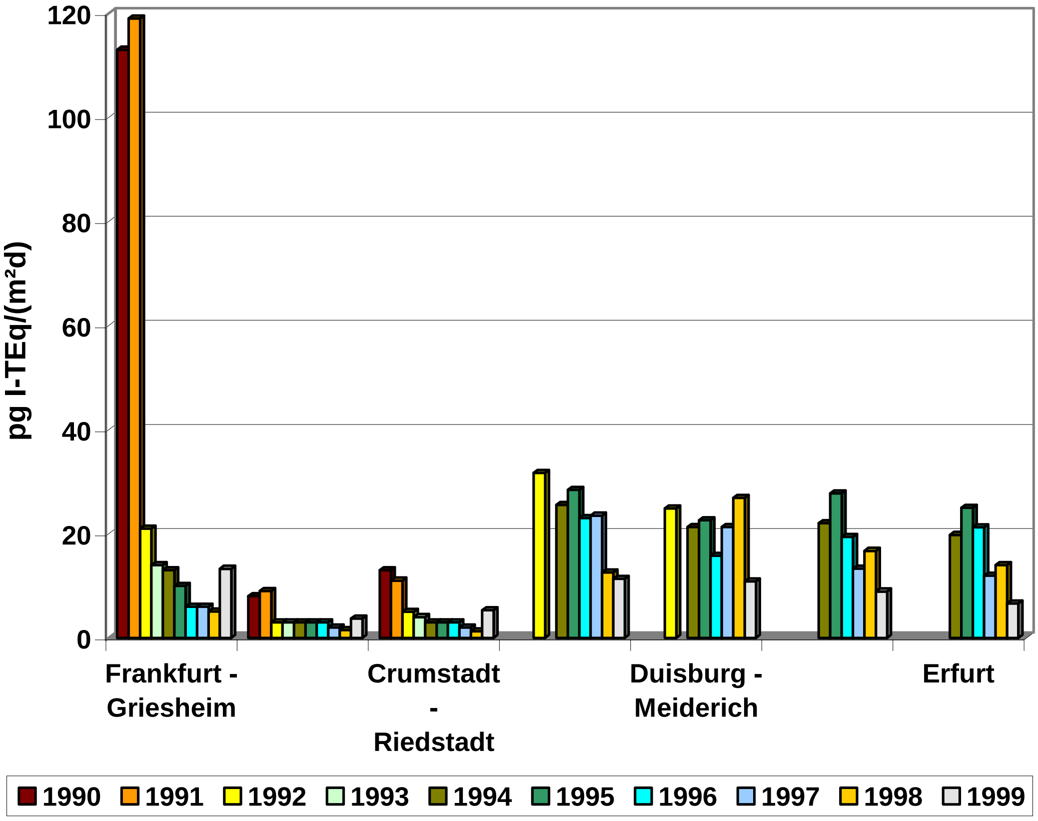


The range of these annual averages is between 4 and 41 pg I-TEq/(m^2 d).

All measuring points in North Rhine-Westphalia are determined by the industrial urban area; therefore the average may only give information on this area, however not on small towns or the rural area.

To secure the information on the trend outlined in Fig. 7 by homogeneous groups of measured values annual averages of deposition are represented as time series in Fig. 8 with series of measurement covering many years for the locations of Hesse, North Rhine-Westphalia and Thuringia.

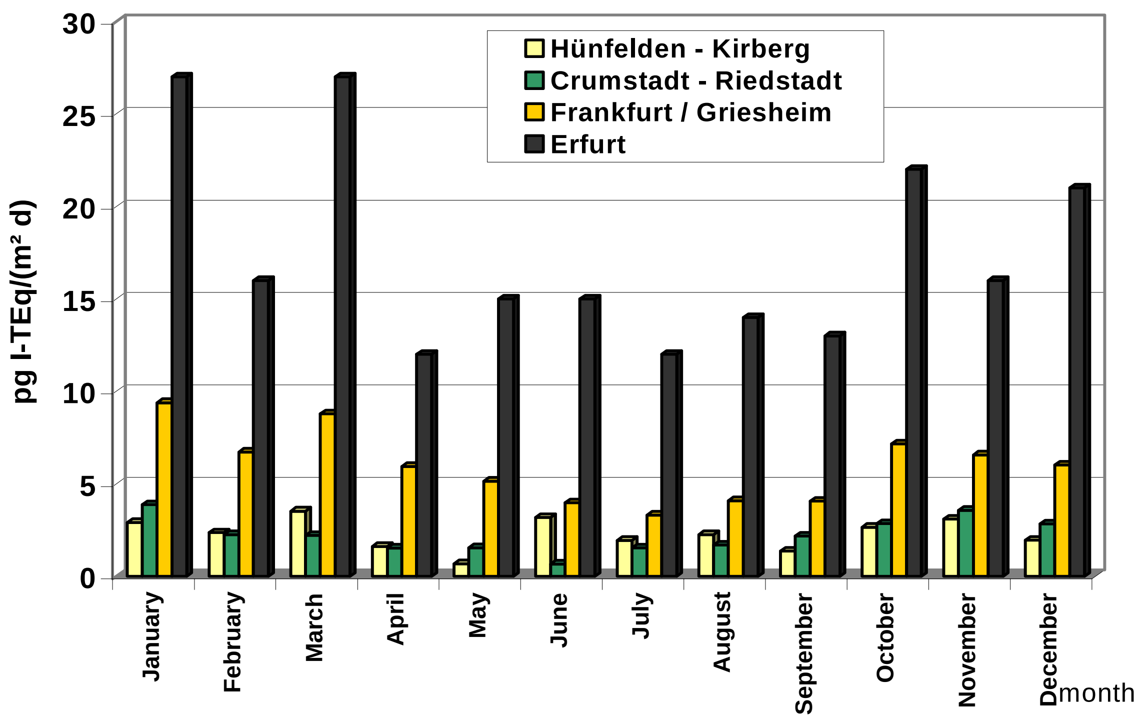
Fig. 8: Trend of the PCDD/F depositions in selected sites of the Federal Laender Hesse, North Rhine-Westphalia and Thuringia, represented by means of the annual averages, evaluation period 1990 – 1999



The clear decline of dioxin deposition in the area of measurement Frankfurt/M.-Griesheim from 1991 to 1992 shows that the measure taken at that time – careful covering of a contaminated site formerly emitting dioxin by an earth layer and grassing it – was successful. The originally high annual averages in 1993/94 in Thuringia /Erfurt were, inter alia, due to the high share of the solid fuel used at that time; yet in the mean time the annual average of the dioxin deposition in Erfurt approached the dioxin deposition rate for Frankfurt/M. The general decline of dioxin deposition is also confirmed by three selected large city locations in North Rhine-Westphalia. Even in the site Huenfelden-Kirberg situated far away from emission sources dioxin is deposited in measurable quantities; also in this location the dioxin deposition has decreased by more than 50 % since 1990.

The averaged annual variation of dioxin deposition for the evaluation period 1994 – 1998 – calculated on the basis of the monthly averages – is represented in Fig. 9 for the four locations Frankfurt/M.-Griesheim, Crumstadt-Riedstadt, Huenfelden-Kirberg and Erfurt for which the annual variation of concentration was represented in Fig. 5.

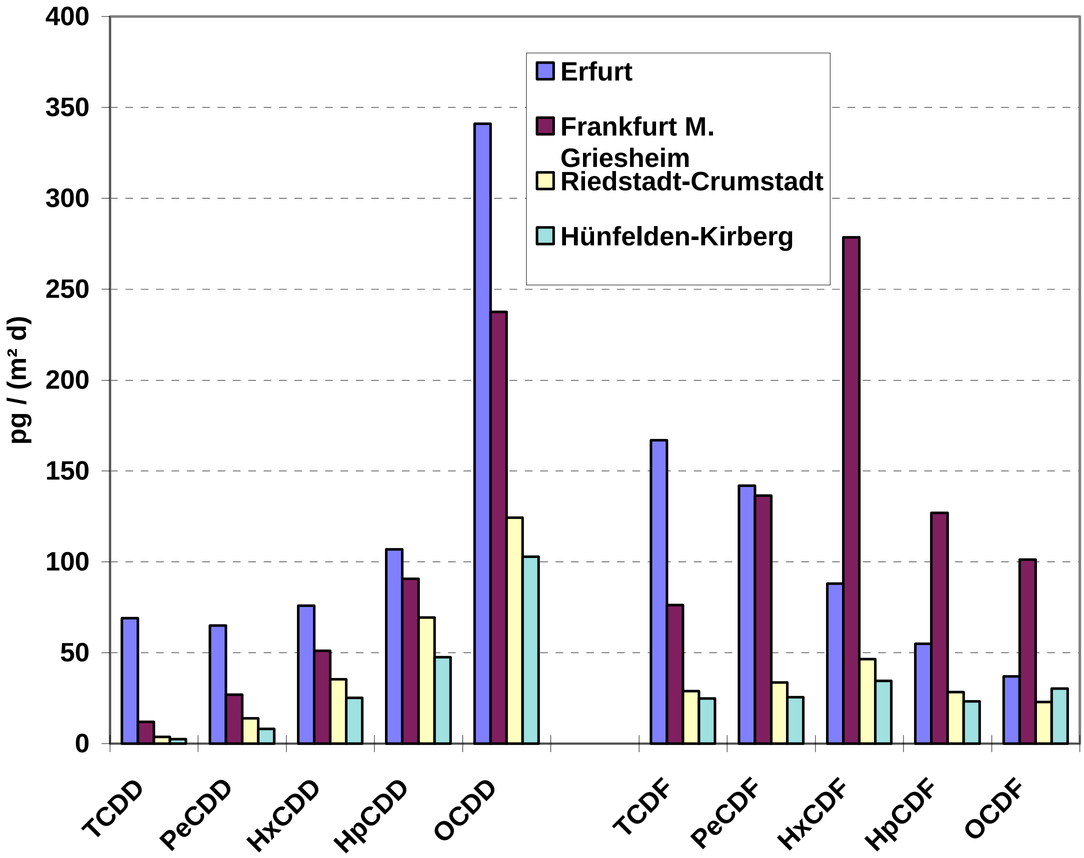
Fig. 9: Annual variation of the PCDD/F depositions in selected locations of the Federal Laender Hesse and Thuringia, calculated on the basis of monthly averages in pg I-TEq/(m² d); evaluation period 1994 – 1998



In the location Frankfurt-M.-Griesheim it is a clear seasonal trend with higher values in the winter half-year and lower values in the summer months but this is not detected in the location Huenfelden-Kirberg. The dust deposition measurements available for the two locations document that in both locations the dust deposition rate is higher in summer than in the winter months. Therefore a low dioxin concentration in the atmosphere could result in the fact that the annual variation of the dioxin deposition to be expected due to the annual variation of the dioxin concentration will be almost compensated by the seasonally varying dust deposition.

Homology profiles for the dioxin deposition measurements – calculated for the four locations Erfurt, Frankfurt/M.-Griesheim, Crumstadt and Huenfelden-Kirberg as average of the measured values available per location – are represented in Fig. 10.

Fig. 10: Homology profile of the PCDD/F concentrations of selected locations in Hesse and Thuringia, evaluation period: 1990-1998



In the homology profiles of deposition measurements location-specific effects result, as a rule, in clearly bigger differences between the locations than in measurements of the ambient air. A difference between homology profiles of ambient air and deposition measurements is to be detected for furans: whereas in ambient air the furan concentrations decline from tetrafurans to octafuran in deposition measurements the tetrafurans up to octafuran show comparable contents or a slight increase from tetrafurans to hexafurans and then again a slight decline to octafuran in the locations Huenfelden-Kirberg and Crumstadt which are far from dioxin sources.

A comparison of the dioxin ambient air with the dioxin deposition measurements shows modifications of the annual variation and for the homology profile during the transfer from air to soil for the data groups at the same location.

6.2 Soil

Soil as an essential and integral part of environment shows a very close link to all environmental compartments. Thus, the input via the air pathway plays a major part. Also various uses, associated with the input of e.g. sewage sludge, compost and pesticides, result in changes of the condition of soil. Due to the „long-term memory“ and the „integral function“ of the soil, on the one hand, and the available results of permanent soil monitoring programs of the Laender, on the other hand, the compartment soil was included in the dioxin reference measuring program.

Due to the short duration of this program (beginning in 1995) it is not yet possible to compile a temporal trend of PCDD/F concentrations in soil or horizons due to fact that recurred sampling of soil monitoring sites (BDF) is only applied after a few years.

The evaluations refer to:

- soil investigations initiated with the beginning of the dioxin reference measuring program (1995) (e.g. in Saxony setting up of soil monitoring sites (BDF),
- results of soil monitoring sites (BDF), already set up in Hesse, Hamburg, Baden-Wuerttemberg and
- results of the soil condition survey (e.g. Bavaria 1989/90).

The evaluations are aimed at determining the condition of the substance contents in soils – based on the criteria of investigation of the background contents in soils. It is not intended to derive background contents as this is not the target of the dioxin reference measuring program, on the one hand, and the data base does not meet the requirements (number of samples and quality criteria), on the other hand.

The structural trend of the area plays a major part with regard to the PCDD/F content in soils. In this connection, settlement density, distance and direction (main wind direction) to local and diffuse emission sources are influencing factors. Due to lack of ambient air or deposition types defined nationwide the factor ambient air is characterized via the type of settlement structure.

According to the requirements of the dioxin reference measuring program „soil“ sampling locations have to be situated in the following areas:

- urban area (agglomeration)
- urban fringe (urbanized area)
- rural area.

This classification is a rough structuring and follows the settlement structural area types of the Bundesamt für Bau- und Raumordnung (BBR) (Federal Agency of construction and regional planning) (formerly Bundesamt für Landeskunde und Raumordnung (Federal Agency of aerial studies and regional planning (BfLR)):

- Regional type I: (regions with high density areas) corresponds to urban area
- Regional type II: (regions with tendencies to urban areas) corresponds to urban fringe
- Regional type III (regions with a rural development) corresponds to rural region.

This classification follows the targets of the Laender working group on Soil protection (LABO) deriving the soil contents with regard to the area type (see: LABO (1998) „Hintergrundwerte für anorganische und organische Schadstoffe in Böden“ (Background values of inorganic and organic pollutants in soils), 2nd edition).

For regional considerations a higher resolution is required. Also regional influencing factors (e.g. emission source structure) have to be taken into account in generalizing the results.

6.2.1 Methodology and quality assurance

The working subgroup on the Dioxin reference measuring program of the Government/Laender working group on DIOXINS recommended to harmonize sampling and analytical methods of the programs of measurement applied or planned in the Federal Laender and to carry out comparative laboratory tests with the participating Laender. Therefore the Federal Environmental Agency in co-operation with the participating Laender prepared recommendations for data collection and documentation (data collection form „soil“). sampling strategy and analyzing. Two homogeneous reference soils were prepared and certified by interlaboratory tests.

6.2.2 Data base and structure

The survey of the data inventory in Table 7 specifies the sampling data on the soil monitoring sites (BDF) classified according to Federal Land, characterization of location, land use, sampling period and number of samples. An evaluation according to use-related, area-specific and temporal criteria was carried through for the participating Federal Laender.

Land use: arable land
forest (coniferous, mixed, deciduous forests, forest and woody plants)
grassland
parks and gardens
vine-growing regions

Area types: urban area (agglomeration - regional type I)
urban fringe (area near urban areas – regional type II)
rural area (regional type III)

Year of sampling:

Table 6 gives a survey of the data base included in the evaluations.

Table 6: Number of samples and sampling period for Laender participating in the dioxin reference measuring program „soil“

Data supplying authority	Number of samples	Sampling period
Bayerisches Geologisches Landesamt	277 [1)]	1989/1990
Hessisches Landesamt für Umwelt und Geologie	160	1992-1997
Landesanstalt für Umweltschutz Baden-Wuerttemberg	54	1985-1989, 1996 (n=2)
Sächsisches Landesamt für Umwelt und Geologie	3	1995
Umweltbehörde Hamburg, FA für Umweltuntersuchungen	10	1992, 1995

[1)] without considering suspected sites

Table 7: Data on permanent soil monitoring areas to be sampled

Federal Land	Charateriszation of location	Soil use	Period	Number of samples
Baden-Wuerttemberg	urban area	arable land	85 - 86	3
Baden-Wuerttemberg	urban area	forest	87	2
Baden-Wuerttemberg	urban fringe	arable land	86	4
Baden-Wuerttemberg	urban fringe	grassland	86	12
Baden-Wuerttemberg	urban fringe	forest	87	4
Baden-Wuerttemberg	urban area	arable land	96	1
Baden-Wuerttemberg	urban area	forest	96	1
Baden-Wuerttemberg	urban area		- - -	1
Baden-Wuerttemberg	urban area	forest	88 - 89	6
Baden-Wuerttemberg	urban fringe	forest	87 - 89	21
Bavaria	urban area	arable land	89 - 90	14
Bavaria	urban area	grassland	89 - 90	6
Bavaria	urban area	settlement area	89 - 90	9
Bavaria	urban area	special cultures	89 - 90	1
Bavaria	urban area	forest	89 - 90	22
Bavaria	rural area	arable land	89 - 90	18
Bavaria	rural area	grassland	89 - 90	46
Bavaria	rural area	settlement area	89 - 90	2
Bavaria	rural area	forest	89 - 90	39
Bavaria	urban fringe	arable land	89 - 90	37
Bavaria	urban fringe	grassland	89 - 90	22
Bavaria	urban fringe	settlement area	89 - 90	22
Bavaria	urban fringe	special cultures	89 - 90	4
Bavaria	urban fringe	forest	89 - 90	43
Hamburg	urban area	settlement area	92	4
Hamburg	urban area	forest	95	6
Hesse	urban area	arable land	92 - 96	21
Hesse	urban area	grassland	92 - 94	10
Hesse	urban area	special cultures	93	7
Hesse	urban area	forest	92 - 97	51
Hesse	rural area	grassland	95	4
Hesse	rural area	forest	95	5
Hesse	urban fringe	arable land	95	7
Hesse	urban fringe	grassland	95	22
Hesse	urban fringe	forest	95 - 97	39
Saxony	urban fringe	forest	95	1
Saxony	urban fringe	arable land	95	2

6.2.3 Evaluation

The evaluation refers to the advice and suggestions given by the Government/Laender working group on Soil protection „soil information system“ /Ad-hoc working group „permanent soil monitoring“ of LABO-AK 2 (1999), permanent soil monitoring: setting up and operation of permanent soil monitoring areas, Chap. 10/.

With regard to a harmonized data evaluation the following arrangements have been made in co-ordination with the participating Laender:

* reference of the data to dry matter (dm) or volume (provided that the apparent density data are available)
- I-TEq calculation with a detection limit equal to zero for data below the detection limit
- inclusion of all locations of soil monitoring sites (BDF) not specifically identified as reference locations (to increase the number of random samples to validate the results)

Furthermore the following variables were included in the evaluation:

- <u>soil horizons</u> – classified according to organic layers <u>(L and O horizons)</u> and topsoil <u>(all A horizons)</u>
- <u>soil use</u> – classified according to arable land, grassland and forest – with a differentiation of forest use being made between coniferous, deciduous and mixed forests
- <u>type of area</u> assigned to the sampling location – classified according to urban area, urban fringe and rural area.

The evaluation was made nationwide. Due to the different number of random samples in the Laender specific statements for the Laender were not made.

As a result of the evaluation the median (50 percentile) the 90 percentile and 10 percentile were determined. The figures with median and 90 percentile are presented. The corresponding overview is shown in Table 8.

When evaluating I-TEq values without including the detection limits there should be basically taken into account that, in particular, in older analytical results due to the detection limits having been higher at that time the dioxin contents may be underestimated compared to later investigations which finally may result in a reduction of the average contents of the total sample set. However, the present evaluation was not carried out separately by years of analyzing as this would have corresponded largely to a Laender-specific differentiation.

If the soil data of this report would be compared to those of the 3rd report of the Government/Laender working group on DIOXINS the values of the dioxin reference measuring program would be lower. This has the following reasons: In the evaluation of the soil data of the dioxin reference measuring program selected permanent monitoring programs of 5 Laender were included as documented in Table 7. This is different to the evaluation of soil data of the 3rd report of the Government/Laender working group on DIOXINS which was carried out using a more comprehensive data base. Thus, the results presented in the figures refer to an intersection of the whole soil data pool available in the database DIOXINS.

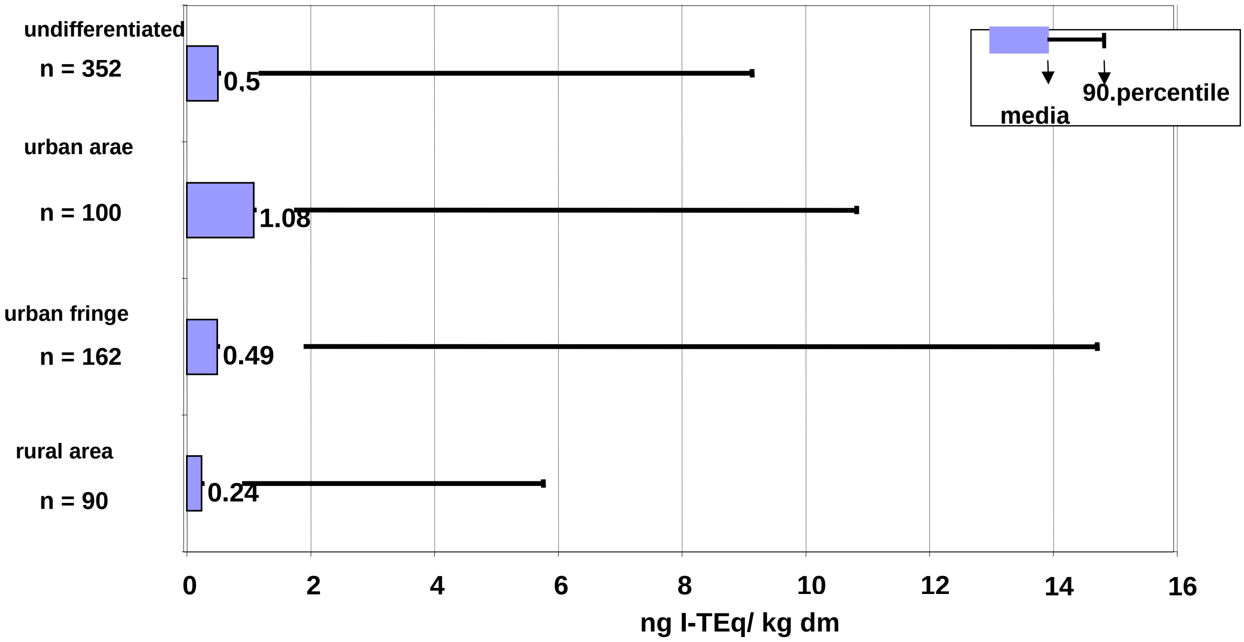
The soil data sets from areas without special impact were used for calculation in both reports. Whereas in the dioxin reference measuring program the statistical calculations were based on about 500 samples the statistical evaluation in the 3rd report of the WG on DIOXINS refers to about 750 samples as a whole. Thus, the higher number of samples affects the calculated statistical parameters (median, 90 percentile).

For the statistical evaluations in the 3rd report of the WG on DIOXINS exclusion criteria were defined which are documented in detail in Chapter 4.4 – Data quality: dioxins and calculation of I-TEq. Fig. 5 in the 3rd report of the WG on DIOXINS shows that the exclusion of data where more than 5 individual congeners meet the detection limit or are below it leads to a result higher by a factor of two. Nearly one third of the samples (in particular, samples taken before 1992) were excluded from the calculation in the 3rd report following this procedure of exclusion. Consequential these result in higher medians and 90 percentile values in this report notably in mineral topsoils and organic layers of the forest use type.

The problems associated with various analytical methods and different detection limits are explained in detail in the 3rd report, Chap. 4.3 and 4.4. The various evaluation methods show clearly how important the application of harmonized methods is in sampling and analyzing to determine dioxins in soil and to reach low detection limits for a better comparability of the results.

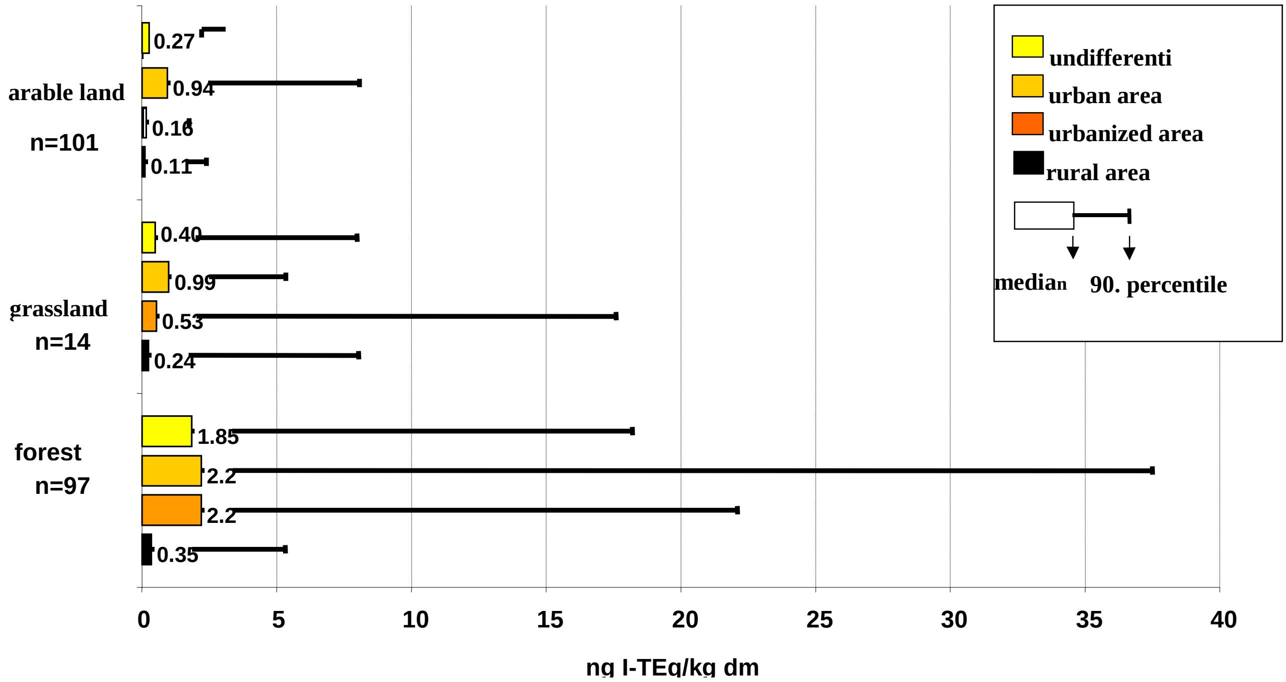
Fig. 11 shows the evaluation for topsoil considering the type of area; Fig. 12 represents the evaluation for topsoil with regard to the type of area and soil use. The data set „undifferentiated“ covers in each case all data sets of the differentiated evaluations listed under it.

Fig. 11: Evaluation for topsoil with regard to the type of area (median, 90 percentile)
Topsoil -without differentiation according to land use



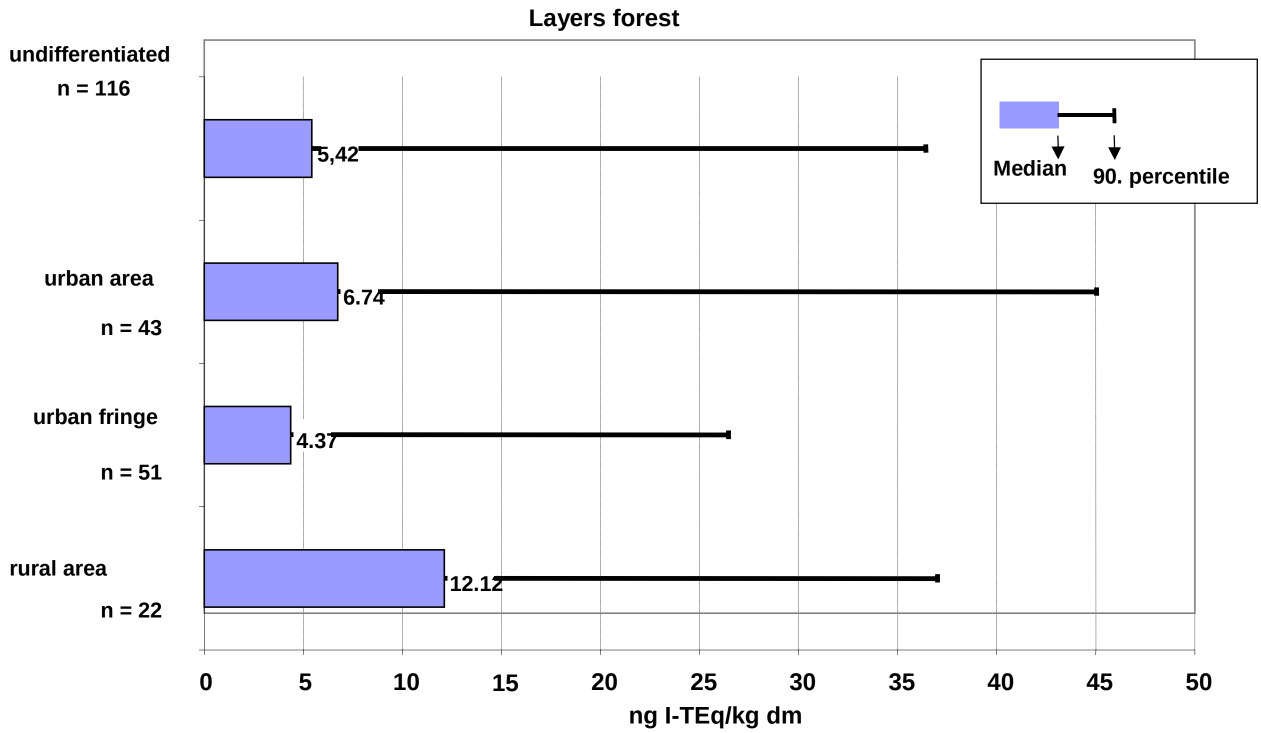
Without the respective differentiation of use the medians of the soil concentration for the urban area are below 1.1 ng I-TEqkg dm. Including the 90 percentile they go up to nearly 15 ng I-TEq/kg dm for the area type „urban fringe“.

Fig.. 12: Soil concentration (median, 90 percentile) Topsoil differentiated according to soil use and area type, dioxin reference measuring program „soil“



With regard to soil use the soil concentrations (median) in the urban areas and urban fringes for topsoil of the „forest“ use are about 2.2 ng I-TEq/kg dm, for grassland and arable land they are below 1 ng I-TEq/kg dm. The 90 percentile for the „forest“ use, in particular for the urban areas, go up to maximally 37.5 ng I-TEq/kg dm.

Fig. 13: Soil concentration (median, 90 percentile) organic layers forest, differentiated according to area type, dioxin reference measuring program „soil“



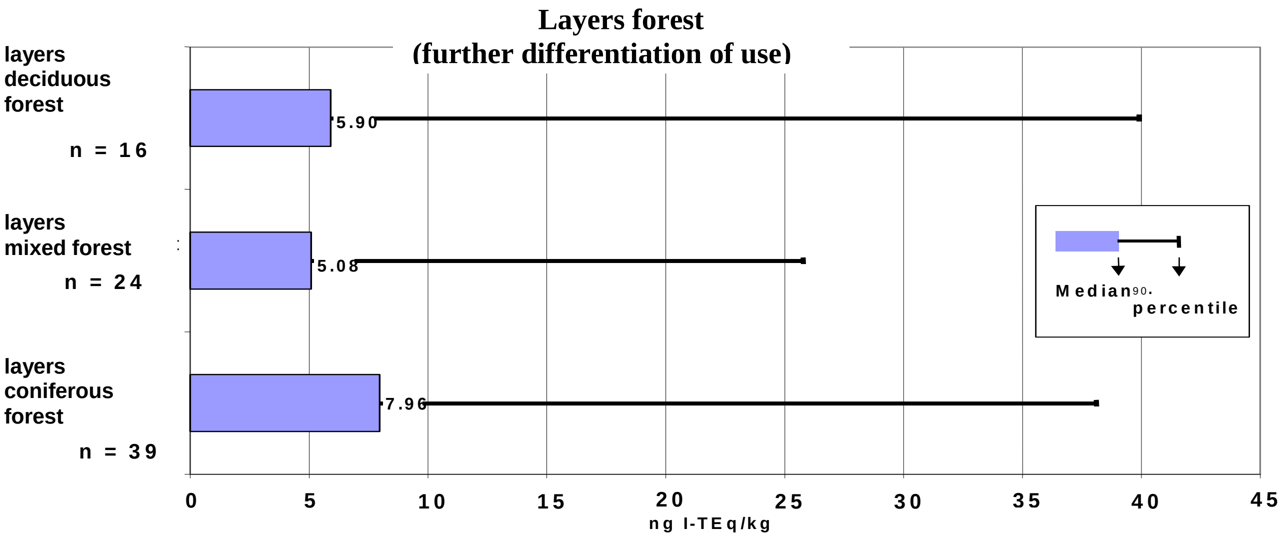
The evaluation of the organic layers from forests – according to the type of area – shows soil concentrations as medians in the range between about 4 and 12 ng I-TEq/kg dm (Fig. 13). Considerung the 90 percentile they vary between about 26 (urban fringe) and 45 ng I-TEq/kg dm (urban area).

The median value remarkably high for the rural area for the organic layers of 12.2 ng I-TEq/kg dm is probably due to the fact that the organic layers are in rural areas more undisturbed than in the urban area and urban fringe. In disturbed organic layers the PCDD/F contents are wider spread and therefore the concentration is lower.

Table 8: Survey of the evaluation of the dioxin reference measuring program „soil“ (I-TEq without including the detection limit, ng I-TEq/kg dm)

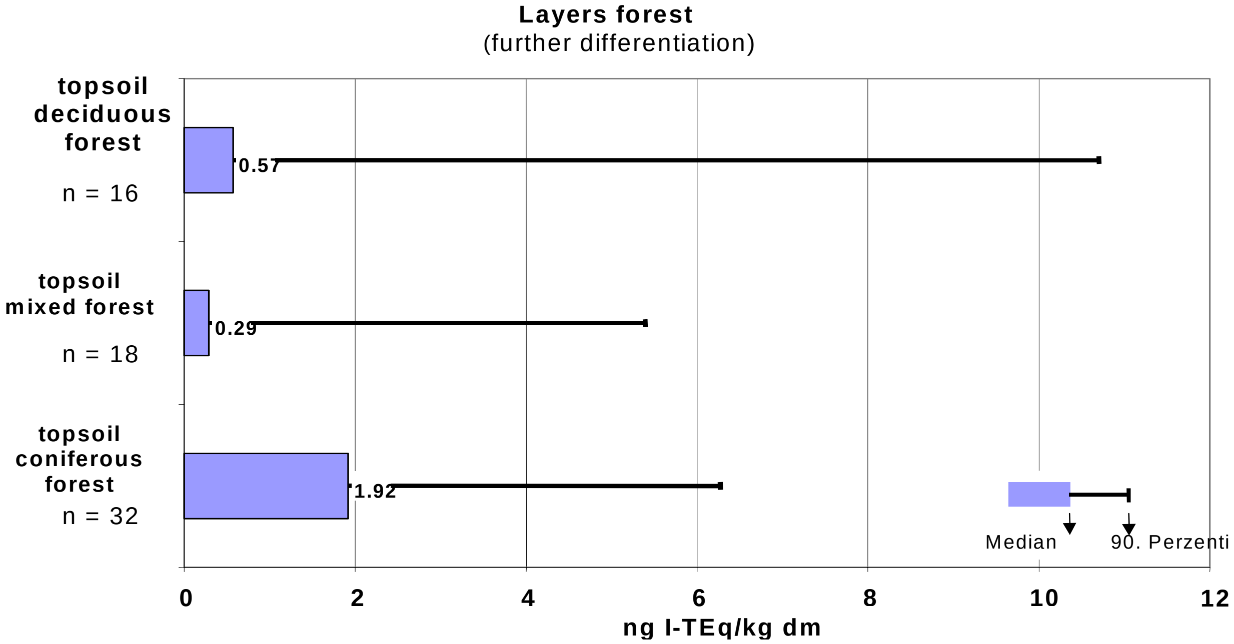
	Without specification according to the type of area				**„Rural area“ type**				**„Urban fringe“ type**				**„Urban area“ type**			
	N	**Median**	90-P	10-P	N	**Median**	90-P	10-P	n	**Median**	90-P	10-P	n	**Median**	90-P	10-P
Comprehensively for all uses																
Layers	119	**5.35**	36.4	0.690	22	**12.1**	37.0	0.976	52	**4.26**	26.5	0.655	45	**6.74**	42.9	0.963
Topsoil	352	**0.500**	9.13	0.025	90	**0.240**	5.76	0.076	162	**0.490**	14.7	0.040	100	**1.08**	10.8	0.041
Arable land																
Topsoil	101	**0.270**	2.37	0	19	**0.110**	2.39	0	61	**0.160**	1.75	0.018	33	**0.940**	8.07	0.182
Grassland																
Topsoil	149	**0.490**	7.98	0.042	50	**0.240**	8.04	0.034	72	**0.530**	17.6	0.082	27	**0.990**	5.34	0.002
Forest																
Layers	116	**5.42**	36.4	0,.675	22	**12.1**	37.0	0.976	51	**4.37**	26.5	0.650	43	**6.74**	45.1	0.942
Topsoil	97	**1.85**	18.2	0.046	21	**0.35**	5.32	0.042	41	**2.20**	22.1	0.083	35	**2.20**	37.5	0.053
thereof coniferous forest																
Layers	39	**7.96**	38.1	0.686	-	**-**	-	-	-	**-**	-	-	-	**-**	-	-
Topsoil	32	**1.92**	6.27	0.089	-	**-**	-	-	-	**-**	-	-	-	**-**	-	-
thereof mixed forest																
Layers	24	**5.08**	25.8	0.520	-	**-**	-	-	-	**-**	-	-	-	**-**	-	-
Topsoil	18	**0.290**	5.39	0	-	**-**	-	-	-	**-**	-	-	-	**-**	-	-
thereof deciduous forest																
Layers	16	**5.90**	39.9	0.497	-	**-**	-	-	-	**-**	-	-	-	**-**	-	-
Topsoil	16	**0.570**	10.7	0,046	-	**-**	-	-	-	**-**	-	-	-	**-**	-	-

Fig. 14: Soil concentration (median, 90 percentile) Layers forest, further differentiation according to soil use, dioxin reference measuring program „soil“



A further differentiation of forest soil use between coniferous forest, mixed forest and deciduous forest shows for the organic layers comparable medians in the range of about 5 ng I-TEq/kg dm for mixed and deciduous forests and about 8 ng I-TEq/kg dm for coniferous forest. The 90 percentile are in a range between 26 and 40 ng I-TEq/kg dm for this use.

Fig. 15: Soil concentration (median, 90 percentile) Topsoil forest, further differentiation



according to soil use, dioxin reference measuring program „soil“

For the topsoil of the various forest uses the background concentrations are below 0.6 ng I-TEq/kg dm in conformity with the median values for mixed and deciduous forests, for coniferous forest the median is about 2 ng I-TEq/kg dm.

6.3 Sediments and suspended matter

In the aquatic ecosystem due to their chemical and physical properties polychlorinated dibenzodioxins and furans are largely bound to the solid phase. Therefore sediments, suspended matter-bearing sediments and suspended matter were studied in the framework of the dioxin reference measuring program. The Federal Laender Hamburg and North Rhine-Westphalia took part in these investigations.
Samples of the sediments were taken by means of sediment grabs from a depth up to 20 cm. Suspended matter-bearing sediments were monthly mixed samples from sedimentation tanks flown through continuously. Samples of suspended matter at the Rhine measuring points were obtained by means of flow centrifuges.

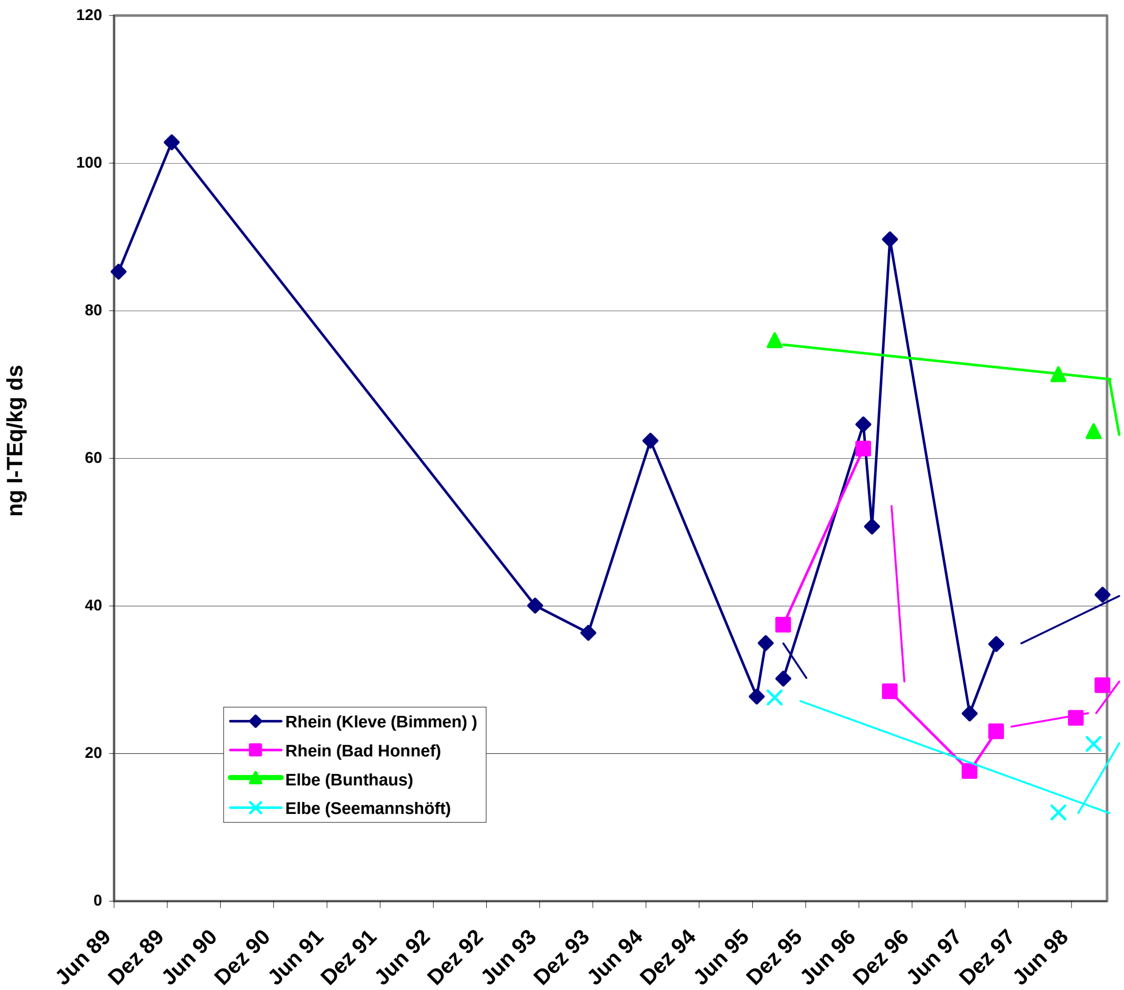
When evaluating the data the detection limits were set to zero in calculating the I-TEq.

The available data set for suspended matter is small. Only for two measuring points at the Rhine series of measurements covering a longer period are available (Kleve [Bimmen] 1989-1998, Bad Honnef 1995-1998). None of the two measuring points shows a clear trend of the dioxin contamination. The decline of the dioxin concentration to be observed due to dioxin minimizing measures taken in the past years does not yet become apparent in the Rhine. The „long-term memory“ of the sediments and resuspensions might be responsible for that. In addition, spatial differences in the contamination of the Rhine do not become obvious. (Fig. 16).

So far only 3 data sets which do not allow to make comments on a trend are available for 2 measuring points from the Hamburg section of the Elbe. Yet, it is obvious that above the Hamburg port (Bunthaus) the contamination of the suspended matter-bearing sediment is higher than downstream, near Seemannshöft; this speaks for a dioxin input from the headwaters of the Elbe, yet below the Hamburg port a certain dilution takes place due to weaker contaminated marine sediments being brought in with the tide.(Fig. 17).

Comparing the dioxin contamination of the rivers Rhine and Elbe at the measuring points mentioned shows PCDD/F contents of the same magnitude, the values varying in a range between 12 and 103 ng I-TEq/kg.

Fig. 16: Temporal trend of the PCDD/F contents in suspended matter of the Rhine and suspended matter-bearing sediments of the Elbe



For suspended matter-bearing sediments sampled at various measuring points from the Hamburg Elbe section in the same period the homology profile was presented (Fig. 17) and compared with the respective profile of selected sediments from inner-city waters of Hamburg Fig. 18). The I-TEq contents of selected sediments are between 43 and 88 ng I-TEq/kg and thus at a similar level as the values of the suspended matter-bearing sediment of the Elbe. It is obvious that the homology profiles of the Elbe measuring points are dominated by furans apart from OCDD and the contents upstream the port (Bunthaus) are higher than downstream, near Seemannshoeft and Blankenese. On the other hand, homology profiles of the sediments of inner-city waters show an OCDD-dominated pattern which corresponds to deposition samples (Chapter 6.1.2). By means of investigations carried out not in the framework of the reference measuring program and mathematical evaluations with neuron systems and multivariat-statistical methods (Hamburger Umweltberichte 57/99) it was possible to detect that the special furan contamination of the Elbe comes probably from the Bitterfeld area and was discharged through the river Mulde.

Fig. 17: PCDD/PCDF homology profiles of suspended matter-bearing sediments (28.07. – 28.08.1995) in various measuring points of the Hamburg Elbe section

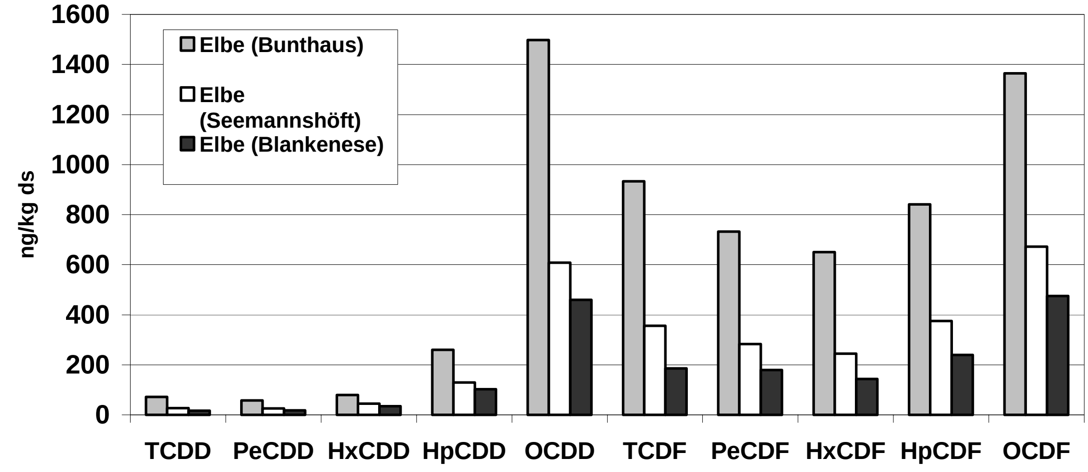
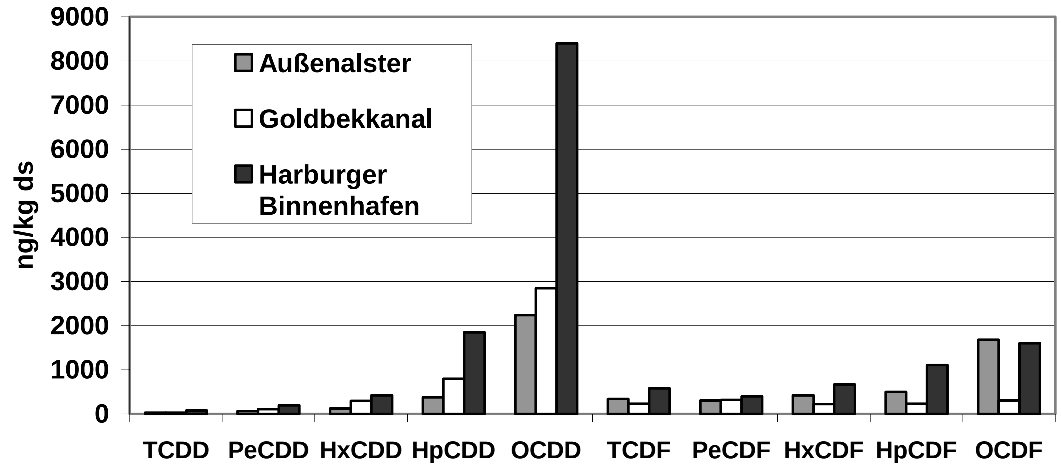


Fig.. 18: PCDD/PCDF homology profiles of selected sediments (1997 or 1998) from inner-urban waters in Hamburg



Homology profiles for the Rhine cannot be shown because respective data are not available.

6.4 Bioindicators, plant foodstuffs

Bioindication allows the qualitative and quantitative determination of the effects of anthropogenic and natural effects of airborne substances on environment using appropriate indicator organisms. It thus takes over a major role in environmental observation or monitoring. By including bioindicators the fact is taken into account that apart from the results of emission and ambient air measurement also results of measurements of effects are taken into account in preparing a purposeful strategy for air pollution control. VDI guideline 3957, page 1, of May 1999 regulates the basis and targets of these biological measuring methods to determine and assess the effects of air pollution on plants (bioindication).

With the aid of bioindicators biomonitoring allows a temporally integrating control of the condition of biotic environment. A difference is made between active and passive biomonitoring. In the case of active biomonitoring an exposure of biological material (e.g. green cabbage) takes place. But in the case of passive biomonitoring effects on bioindicators at their place of growth (e.g. spruces) are studied.

Results of bioindicator studies relating to PCCD/F contamination, always of one bioindicator of passive biomonitoring and one of active biomonitoring, are presented. In the framework of the dioxin reference measuring program the Laender Bavaria and Lower Saxony take part in these studies.

6.4.1 Spruce needles

Since 1992 the Bayerisches Landesamt für Umweltschutz (Office of Environmental Protection of the Land of Bavaria) has been carrying out studies of the PCDD/F contamination in spruce locations. These spruce locations were selected from a program existing since 1977 for the investigation of spruce needles for their sulphur content.

The spruce sampling sites are situated in intersection points of a grid with a side length of 16 km. In 1991 it was thinned out to a grid distance of 16 x 32 km in regions with especially low sulphur contamination. Since that time samples of 191 trees have been taken at a 2-year rhythm.

In 1992/93 26, in 1993/94 15, in 1995/96 20 and in 1997/98 19 spruce locations were selected for studying the PCDD/F contamination. It is the objective to obtain area-covering information on the PCDD/F contamination. Therefore the locations change for each sampling. Thus, comments on the temporal trend cannot be made. For the investigation one-year-old needles from the upper crown area of three spruces per location are combined to composite samples.

The samples are taken per period of investigation in autumn and in the following spring. The results of 80 sampling sites are described in Figs. 19 to 22.

Fig. 19: Bioindicators (spruce needles) autumn-spring 1992/1993 in Bavaria

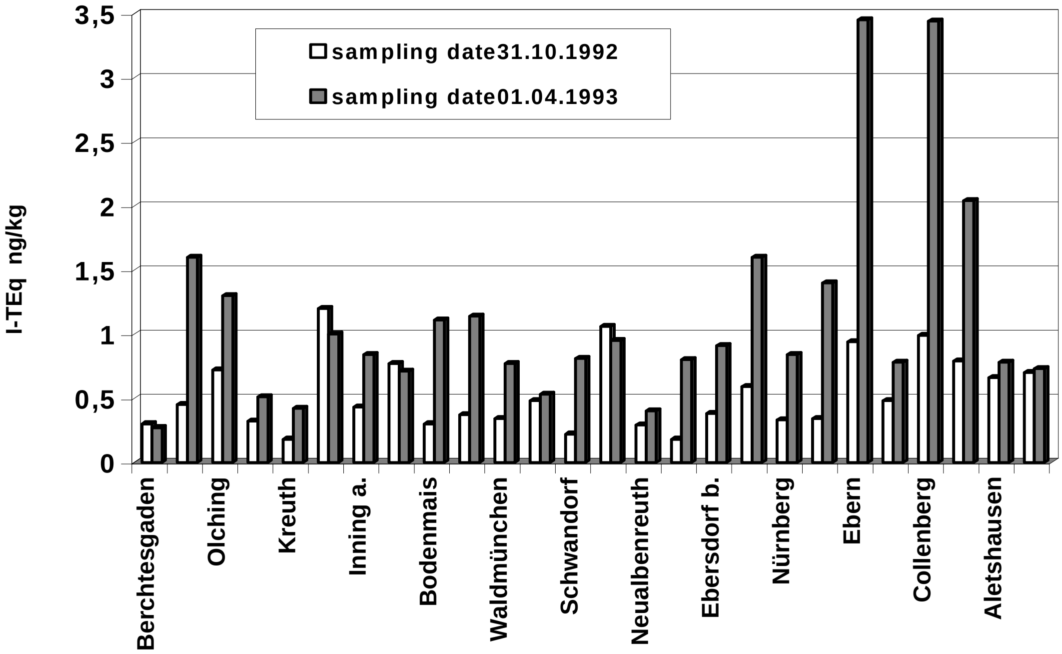


Fig. 20: Bioindicators (spruce needles) autumn-spring 1993/94 in Bavaria

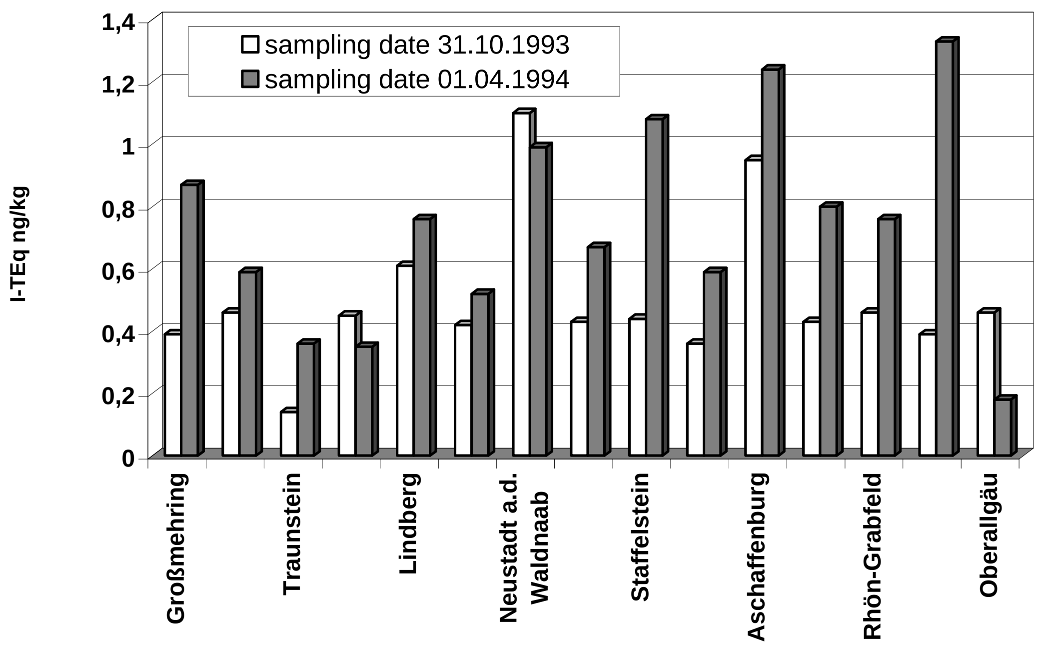


Fig. 21: Bioindicators (spruce needles) autumn-spring 1995/1996 in Bavaria

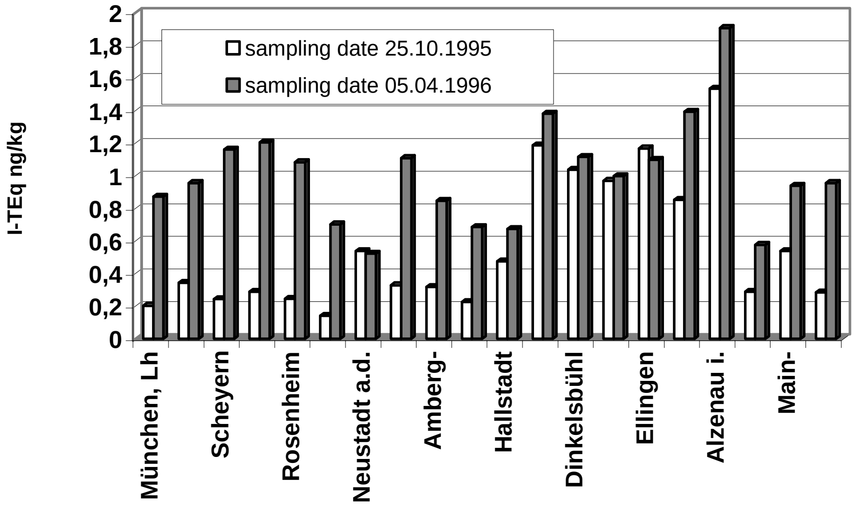
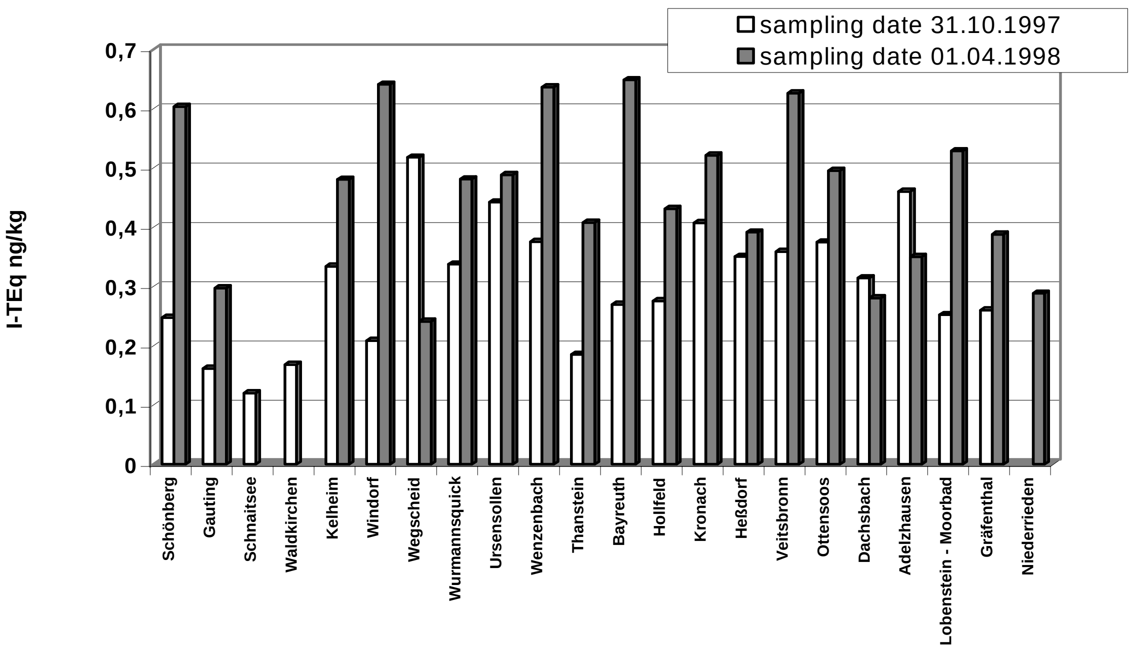


Fig. 22: Bioindicators (spruce needles) autumn-spring 1997/98 in Bavaria



During the winter period an I-TEq increase by 50 to 400 % was detected in 49 locations, in 30 locations the changes varied in the range of 50 %, one individual location attracts attention by a decline of more than 50 %. An interpretation of the I-TEq changes within 50 % seems to be scarcely appropriate due to potential inaccuracies of analyzing in the low concentration ranges and the variation limits when applying passive bioindicator approaches. However, an I-TEq increase of more than 50 % up to 400 % during winter in more than 60 % of the sampling locations can be regarded to be secure.

As causes domestic fuel and insignificant air exchange during inversion weather conditions in winter come into consideration. Direct industrial effects are out of the question due to the choice of location, however long-distance transport is not to be excluded. For the time being, a profound analysis of the connections between location conditions and I-TEq increase has not been made. First attempts to find explanations through simple parameters such as distance to the next settlement, sea level or streaming along the spruces were not successful.

6.4.2 Green cabbage

Dioxins and furans (PCDD/F) are emitted into atmosphere from various sources. The long-distance transport of these persistent compounds in the atmosphere leads to an ubiquitous distribution. Far from emission sources this causes the so-called background concentration, near emission sources an additional contamination with a shift of the congener pattern can occur.

Due to its very large leaf surface and structure and its long stay time on the cultivated area green cabbage is an appropriate indicator plant for airborne pollutants. The investigated parts of the plant do not have a direct soil contact so that pollution by adhering soil particles have only insignificant effects; furthermore the transfer of PCDD/F with the transpiration flow from soil to the green cabbage plant takes place only to a negligible extent.

Plant foodstuffs stand at the beginning of the food chain showing usually – i.e. if a special impact is not to be stated – insignificant PCDD/F contents. However, congener profiles in foodstuffs of plant origin and in animal foodstuffs differ remarkably: in plant foodstuffs we find a multitude of other congeners in addition to the 2,3,7,8 chlorosubstituted congeners.

6.4.2.1 Sampling, method, quality assurance

The samples taken in Lower Saxony come from the official foodstuff monitoring and were taken directly at the producers cultivating green cabbage for use as foodstuff. They reflect the background values without reference to certain emission sources. The samples were taken in the period between 1995 and 1999, altogether 76 samples were investigated in 39 locations. Annual sampling in representative locations of cultivation over the whole period was not possible due to the fact that the agricultural use of a location as area for cultivating green cabbage was not maintained permanently. It was not possible to exert influence on the time of sampling within a harvesting period; the interpretation of the data was thus complicated.

The green cabbage samples from Bavaria were used as indicator plants in the framework of an active biomonitoring in permanent monitoring sites which – with possibly one exception – are beyond immediate effects of individual pollution sources; these data are also background data. The 24 samples were exposed in 6 different locations in 1996 and 1997. The exposure of green cabbage took place always over 8 weeks beginning in August and October.

As has been considered to be appropriate for investigations relating to the input pathway air/plant according to the final report of the working subgroup I „Measuring programs“ of the Government/Laender working group on DIOXINS (Texts 21/92 of the Federal Environmental Agency) the green cabbage samples from Lower Saxony and Bavaria were not washed before treatment but coarse dirt particles were only mechanically removed.

6.4.2.2 Data and evaluation

The Bavarian results of the PCDD/F studies were given for ng/kg of dry mass (DS) considering half of the detection limit, the Low Saxon results for ng/kg of fresh weight considering the full detection limit. Apart from the 2,3,7,8 substituted PCDD/F congeners also the cumulative parameters of all congeners were determined.

The 76 green cabbage samples from Low Saxony studied show PCDD/F contents between 0.01 and 0.45 ng I-TEq/kg of fresh weight with a median value of 0.07 ng I-TEq/kg of fresh weight.

The 24 investigated green cabbage samples from Bavaria show PCDD/F contents between 0.07 and 1.54 ng I-TEq/kg dm with a median value of 0.37 ng I-TEq/kg dm.

A comparison of the data from Lower Saxony (reference: fresh weight) and Bavaria (reference: dry mass) shows similar dioxin contents in the green cabbage samples investigated – when considering a medium water content of green cabbage of 83.6 %.

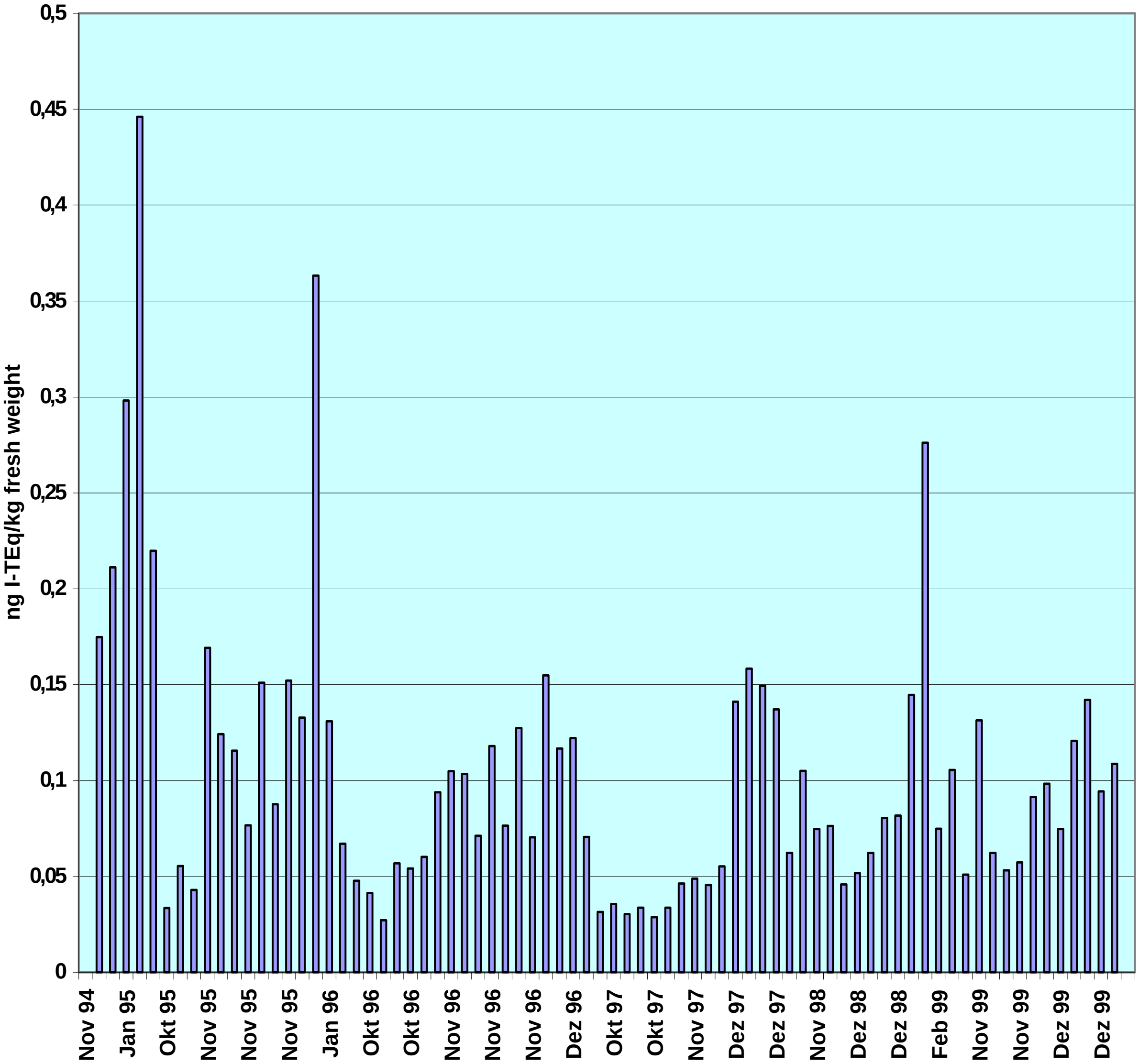


Fig. 23: Trend of the PCDD/F contents in green cabbage from Lower Saxony in the years between 1995 and 1999

In Fig. 23 the I-TEq contents of all samples investigated in the period between 1995 and 1999 in Lower Saxony are represented. By means of this representation a difference between a constant or slightly declining PCDD/F contamination in the period under investigation may not be reliably made.

For seven selected locations in Lower Saxony where sampling was possible at least three times the dioxin contamination of green cabbage was represented as a time series in Fig. 24. As green cabbage shows a distinct annual variation with an increase in the PCDD/F contents within a harvesting period samples with the same month of sampling are to be preferred to make comments on the trend. Using this criterion you can see in the figure that in altogether five locations a decline of the PCDD/F contents in green cabbage from Lower Saxony is to be detected during the period of study. In two other places (Loeningen and Papenburg) we can rather proceed from a nearly constant contamination.

As the sampling locations are spread over Lower Saxony and the decline predominates in the indvidual locations we can, in general, proceed from a decline of the dioxin contamination via air input as individual indications of an increase are scarcely available.

Fig. 24: Time series of the PCDD/F contents in selected locations of Lower Saxony

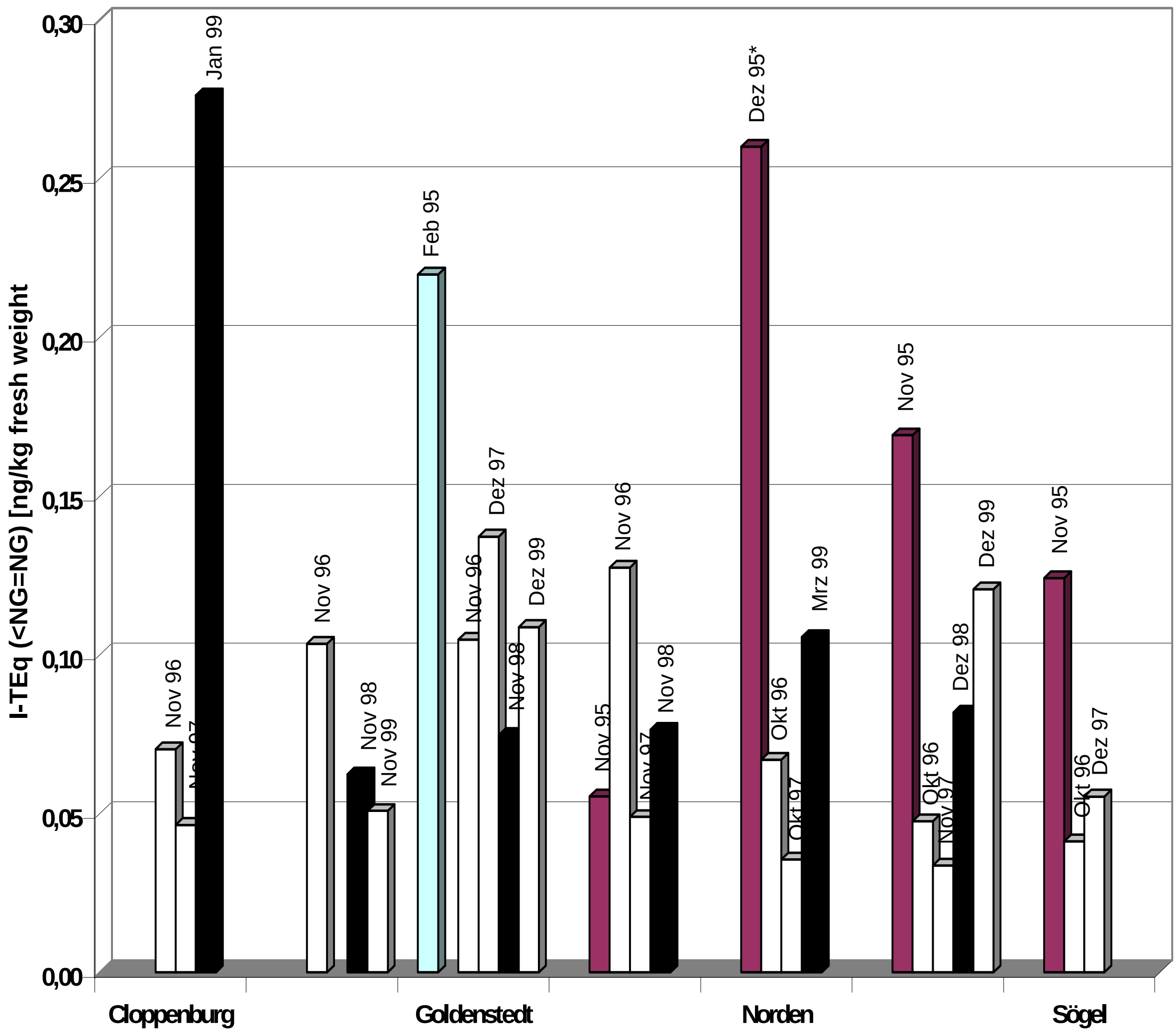


Fig. 25 shows the annual variation of the average monthly PCDD/F contents in green cabbage from Lower Saxony for the months October to March for the period between 1995 and 1999. An increase in the PCDD/F contamination with the vegetation period may be detected. This may be, on the one hand, due to the longer stay time, yet also to an increased dioxin input in the winter months.

Also the values from Bavaria graphically represented in Fig. 26 show that the dioxin contents in green cabbage are higher in the months October/November than in the months August/September (by a factor of 2-6 depending on the location). Probably the effects of intensified domestic fuel and inversion weather conditions with a reduced air exchange in autumn/winter are responsible for the seasonal increase. The increased PCDD/F contents of the location Weissenstadt situated in the Bavarian/Czech border area point to a large-area ambient air from distant emission sources.

As the dioxin contamination of green cabbage is affected by many parameters well-founded comments on the temporal trend of the dioxin contents in the Bavarian green cabbage samples cannot be given due to the short period of monitoring.

Fig. 25: Annual variaton of the average monthly PCDD/F contents in green cabbage for the period between 1994 and 1999 in Lower Saxony

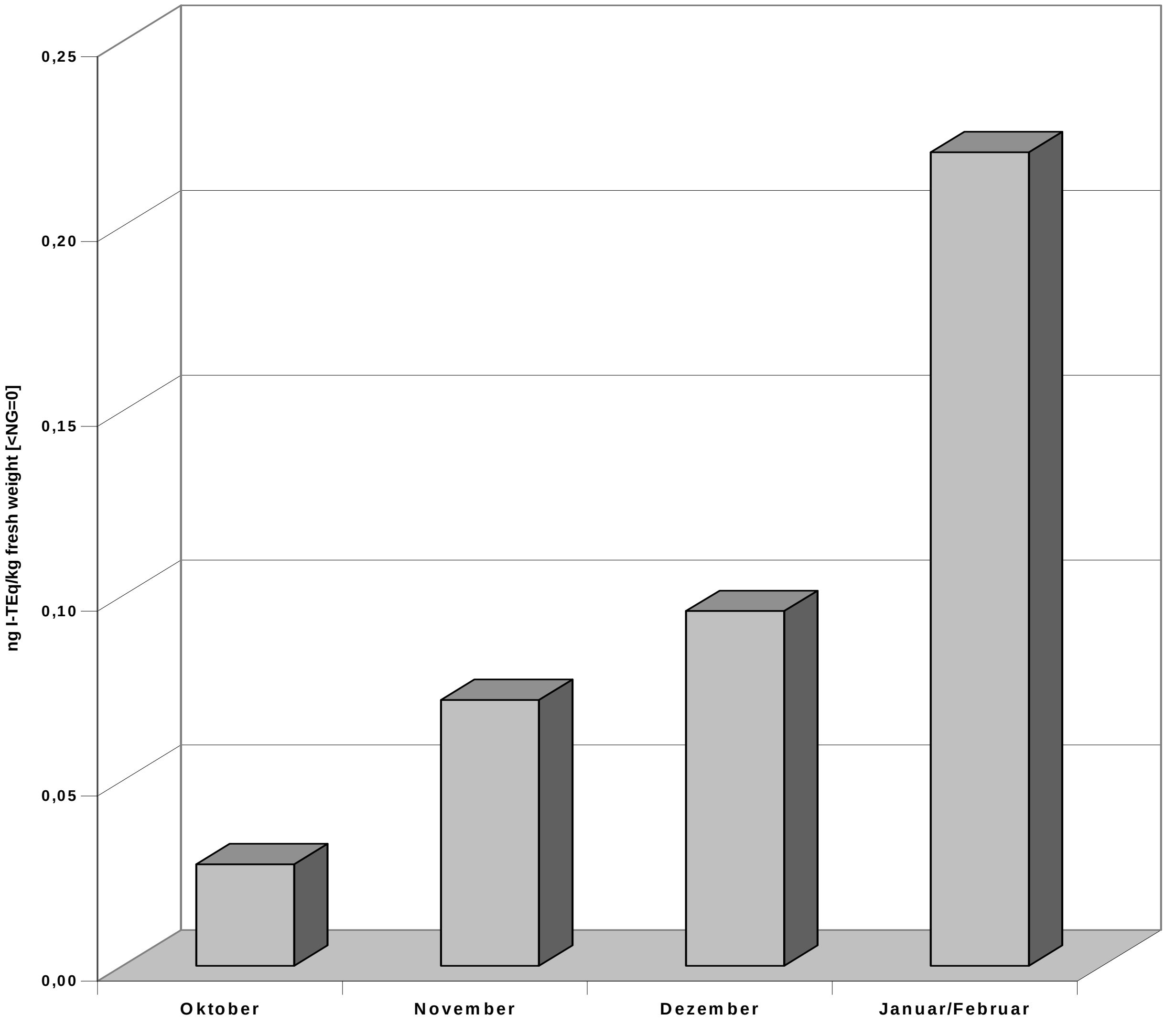
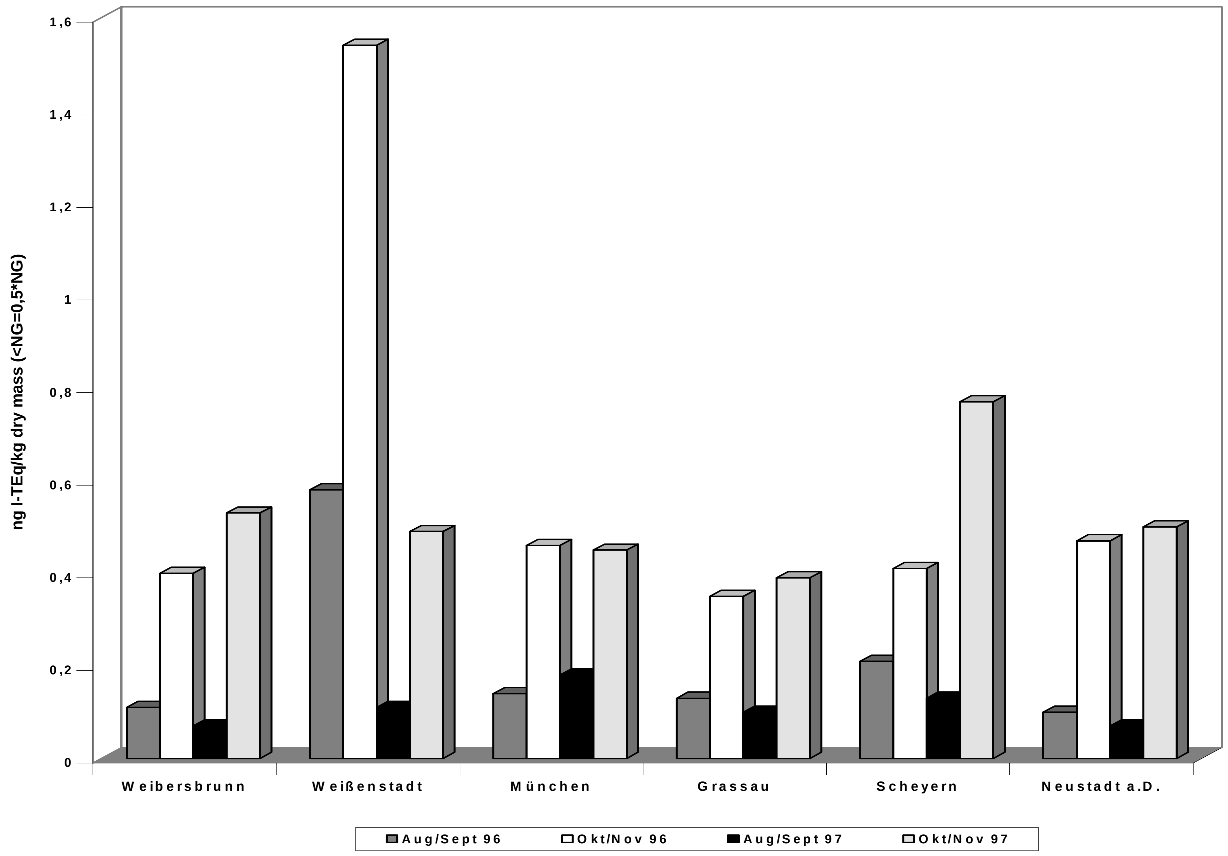


Fig. 26: PCDD/F contents of Bavarian green cabbage samples from exposure tests



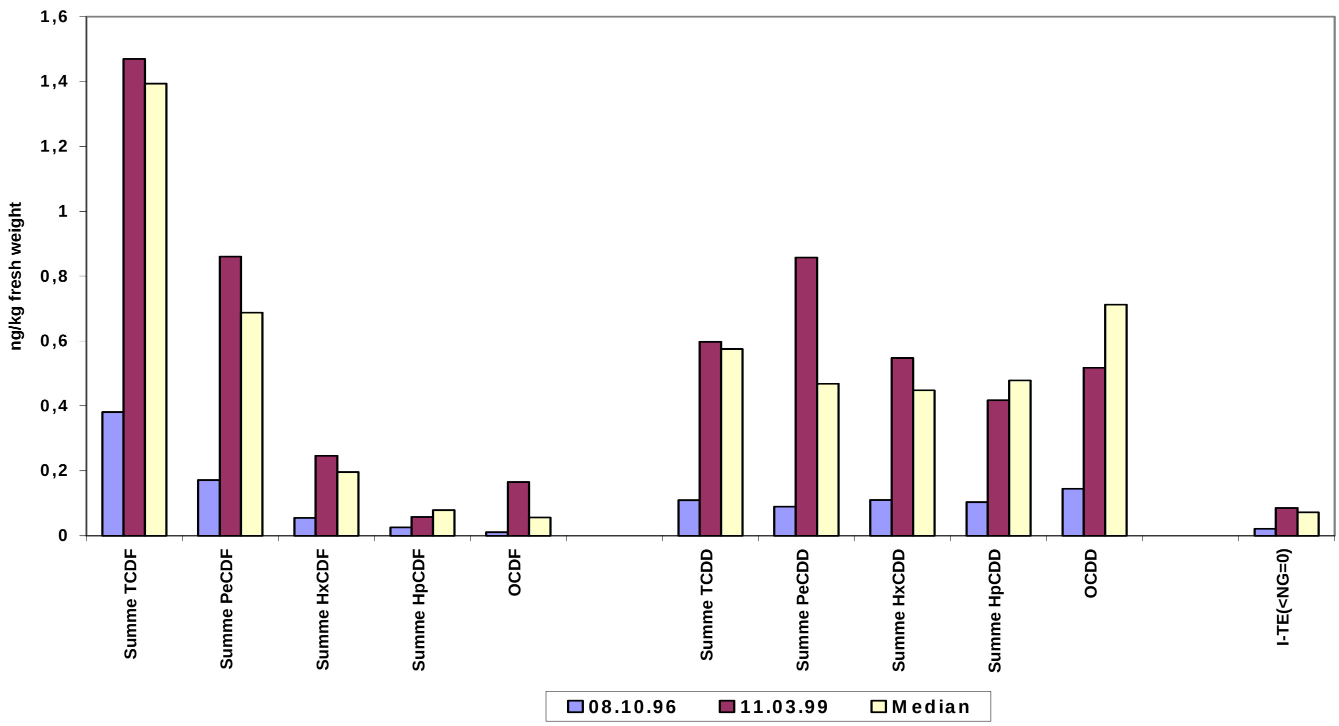


Fig. 27: Homology profiles of the median values of all green cabbage samples and of two individual samples from Lower Saxony

In Fig. 27 the homology profiles of the median values of all green cabbage samples from Lower Saxony and of 2 green cabbage samples taken at the beginning or the end of the harvesting period are compared. The homology profiles are identical in all three cases; disturbing effects by local sources are not to be detected. For green cabbage there is to be noticed that in furans the chlorohomologue contents of tetrachlorinated to octaclorinated compounds decline strongly whereas the chlorohomologue contents of dioxins from tetra to octachlorinated compounds are equally high. Basically the homology profiles correspond to the profile of the burning pattern. The pattern typical of the deposition with increasing contents of low to higher chlorinated dioxins is not to be detected.

The objective of the dioxin reference measuring program to determine a temporal trend of the dioxin contamination of the bioindicator „green cabbage“ may not yet be considered to be fully reached for the time being proceeding from the data material available for the compartment.
This is primarily connected with the fact that the PCDD/F input from the atmosphere into the compartment green cabbage depends on many varying and not controllable factors such as e.g. duration of vegetation, seasonal differences of impact and changing climatic influences (wind direction, wind speed, rain frequency, temperature, inversion weather conditions etc.). In addition, the series of measurement available are too short. To make reliable comments the monitoring period has to be extended. However, the data available at present point altogether to a slight decline of the situation of PCDD/F contamination in green cabbage for the years 1995 to 1999.

6.4.3 Lettuce and green cabbage: biomonitoring of foodstuffs

In Baden-Wuerttemberg lettuce and green cabbage samples were investigated which were not planted purposefully as indicator plants but come from enterprises producing foodstuffs. Hereby, the samples „lettuce 1“ and „green cabbage 1“ come from a rural region reflecting the usual background concentration. The samples lettuce 2 and 3 and green cabbage 2 come from a rural region where in 1997 a waste incineration plant was to be put into operation. Therefore in three agricultural enterprises three plant foodstuffs were investigated in the framework of a reference measuring program to detect potential effects. All samples show dioxin contents with a variation of background concentration usual for plant foodstuffs. Clear uniform temporal trends are not to be recognized in the monitoring period between 1996 and 2000; the contents are altogether kept in the same order of magnitude. It may be excluded that the opening of the waste incineration plant will result in significant changes of the dioxin contents of the plant foodstuffs produced in the surrounding.

6.4.4 Lettuce

Table 9: Dioxin contents in lettuce

	Background	Background	MVA	MVA	MVA
Year	Lettuce 1	Green cabbage 1	Lettuce 2	Green cabbage 3	Green cabbage 2
	pg I-TEq/kg of fresh weight				
1996			1.9	7.3	
1997	13.0	15.5	4.5	5.5	23.8
1998	7.8	23.3	6.0	11.4	42.5
1999	6.2	24.5	4.8	8.4	64.6
2000	9.5	23.6	5.4	7.5	38.4
	pg I-TEq/kg of dry mass				
1996			32	141	
1997	228	72	88	84	111
1998	191	120	97	173	225
1999	162	84	103	93	265
2000	194	125	102	108	201

6.4.5 Grass samples

In 5 locations in Baden-Wuerttemberg always grass, hay and grass silage samples were taken each year in the period between 1996 and 2000. Two locations are situated in a rural region, one location shows beginnings of an urban area and two locations are characterized as „large urban areas“. Table 10 summarizes the results of the study.

The range < 100 to < 300 pg TEq/kg LTS (air-dried dry matter, equivalent to 88% dry matter) may be regarded as usual background concentration of grass, hay and grass silage samples where the detected variations are usual. Especially striking was the extremely increased content of the grass sample „rural area II“ from 1998. Comprehensive additional investigations have shown that the grass comes from a special area where the competent Office of Agriculture carried out fertilization tests and applied higher contaminated Thomas meal fertilizers in 1988.

Table 10: Dioxin contents in grass, hay and silage (I-TEq/kg LTS)

Year	Grass	Hay	Silage	Grass	Hay	Silage
	Rural area I			Rural area II		
1995	283			221	204	62
1996	239	87	108	114	entf	entf
1997	87	111	68	52	97	86
1998	70	145	84	5198	161	105
1999	84	73	289	287	129	144
	Urban fringe					
1995	263					
1996	174	112	258			
1997	89	318	254			
1998	77	135	109			
1999	88	345	282			
	Urban area high density area I			high density area II		
1995	415			entf.	entf	entf
1996	105	207	158	122	149	entf
1997	65	145	244	74	429	134
1998	500	378	306	112	365	197
1999	143	80	277	98	80	128

6.5. Dairy products and raw milk

The data on dioxin concentrations in cow’s milk presented by Baden-Wuerttemberg, Bavaria, Hesse, Lower Saxony and North Rhine-Westphalia in the framework of the reference measuring program comprise dairy, tank, collecting milk, butter, cream and cheese. The dioxin concentration of the dairy products butter, cream, cheese and dairy milk are to be considered as being on an equal level as the concentrations refer to the fat content.

Collecting farm milk represents milk of a farm. Whereas tank milk characterizes milk of a few farms and thus smaller areas, bigger catchment areas are covered by dairy milk, butter and cream. This refers, on an intensified scale, to mixed samples of milk from various dairies. Therefore in the case of area-covering sampling by means of dairy products representative data on larger areas may be derived. On the other hand, collecting farm and tank milk samples allow to make comments on local disturbing effects and feedingstuff contamination (see Chapter 6.5.8 Citrus pulp).

In spite of differing samples and sampling well-founded comments may be made on the basis of the altogether 1397 results of study presented on:

- the concentration level
- the variation of the dioxin concentrations
- the distribution of congeners
- the ratio between WHO-PCDD/F-TEq (without PCB similar to dioxin) and I-TEq,
- seasonal variations,
- the effects of regions (rural area, urban area, urban fringe) and
- the contamination by citrus pulp.

Due to seasonal variations data on the trend of concentration over a period of a few years have to be derived, preferently by means of samples taken in the same season and in the same location. By means of linear regression the changes of concentration within the monitoring period may be estimated (ascent of the equalization lines). Samples additionally contaminated by citrus pulp have not been considered. This applies also to calculations of the seasonal variations.

To calculate the TEq concentrations half of the detection limits were applied to the congeners not detected. In general, all laboratories reached low detection limits. This becomes also obvious by the fact that the calculation of the TEq showed only neglectable differences depending on the consideration of congeners detected with including the full or no detection limit (upper or lower bound).

6.5.1 Survey of samples

Results of investigation of 706 dairy products (milk, cream, butter, cheese), 235 tank milk samples and 456 collecting farm milk samples are available. A survey of these samples is given in Table 11.

Table 11: Survey of samples**:** type and number of samples, number of areas (dairies, tank tours, farms) and period of investigation

type of sample	n samples	n areas	year
Baden-Wuerttemberg			
butter/cream (rural area)	56	5	1996-2000
tank milk (rural area)	88	9	1996-2000
tank milk (rural area in the vicinity of industry)	10	1	1996-1999
farm milk (rural area)	38	3	1994-2000
Bavaria			
dairy milk	201	65	1992-1999
diary milk (pool samples)	39	26	1996, 1999
butter	11	11	1998
farm milk (rural area in the vicinity of emission sources)	129	20	1989-2000
farm milk (rural area)	27	6	1989-2000
Hesse			
tank milk (rural area)	5	1	1995-2000
tank milk (urban area)	15	3	1995-2000
farm milk (rural area)	5	1	1995-2000
farm milk (urban area)	15	3	1995-2000
Lower Saxony			
tank milk (rural area)	117	10	1994-2000
farm milk (special program)	110	6	1992-2000
farm milk (milk hygiene program)	42	6	1995-2000
North Rhine-Westphalia			
dairy products	399	43	1990-1998
farm milk (rural area)	36	2	1996-2000
farm milk (urban area)	38	2	1996-2000
farm milk (urban fringe)	16	1	1996-1999

6.5.2 Trend of dioxin concentrations

6.5.2.1 Dairy products

In Baden-Wuerttemberg, Bavaria and North Rhine-Westphalia the 706 dairy product samples (dairy milk, butter, cream, cheese) summarized in Table 11 were investigated between 1990 and 2000.

Baden-Wuerttemberg

The samples originate from rural areas without special dioxin emission sources, thus representing the background concentration. Sampling was concentrated on the months February to September.

The trend of the dioxin concentrations for all dairy samples (butter and cream) may be seen in Fig. 29. The concentrations drop in summer by 96 from, on average, 0.53 pg I-TEq/g of fat (minimum-maximum: 0.44-0.71) to, on average, 0.30 pg T-TEq/g fat (minimum-maximum: 0.25-0.33) in summer 2000. The short and clear increase in between is remarkable due to the additional contamination by citrus pulp.

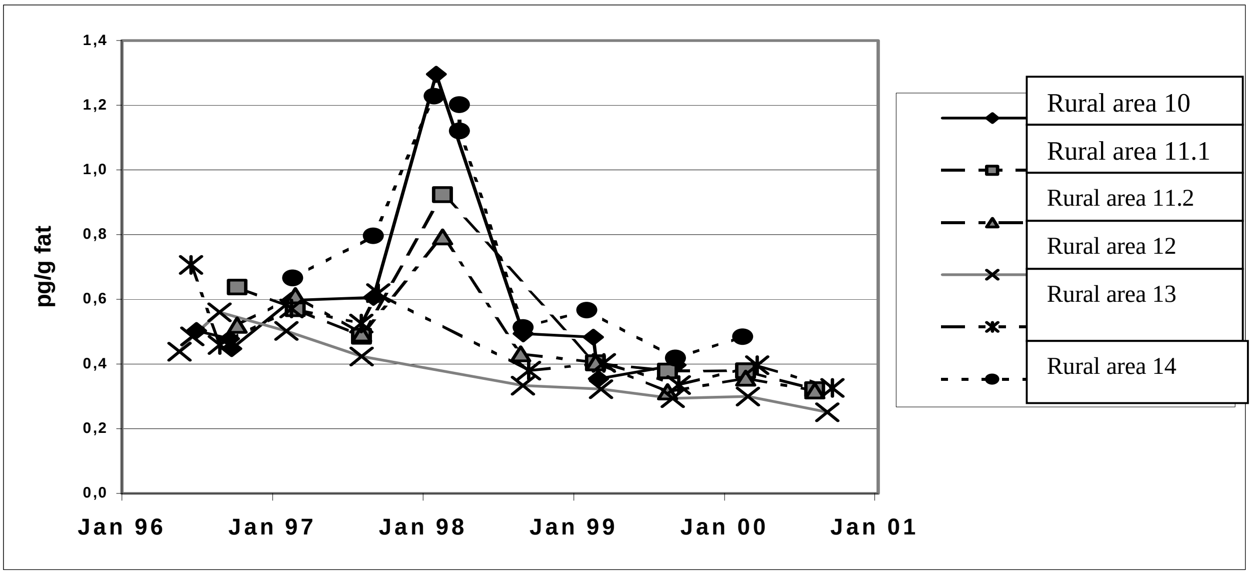


Fig. 28: Trend of the dioxin concentration (I-TEq) in dairy products from Baden-Wuerttemberg

Bavaria

The samples originate also from rural areas without special dioxin emission sources, thus representing – with possibly one exception – the background concentration. The samples were predominantly taken in the 4th quarter.

The results of dioxin concentrations in diary milk (dm), pooled dairy milk (dm-pool) and butter represented in Fig. 29 show a decline of the dioxin contamination (except butter samples contaminated by citrus pulp) in the monitoring period: in autumn 1992 dairy milk with average dioxin contents of 0.88 pg I-TEq/g fat (minimum-maximum: 0.69-1.12) shows higher contents than in autumn 1999 (average: 0.37-0.40 pg I-TEq/g fat, minimum-maximum: 0.27-0.48).

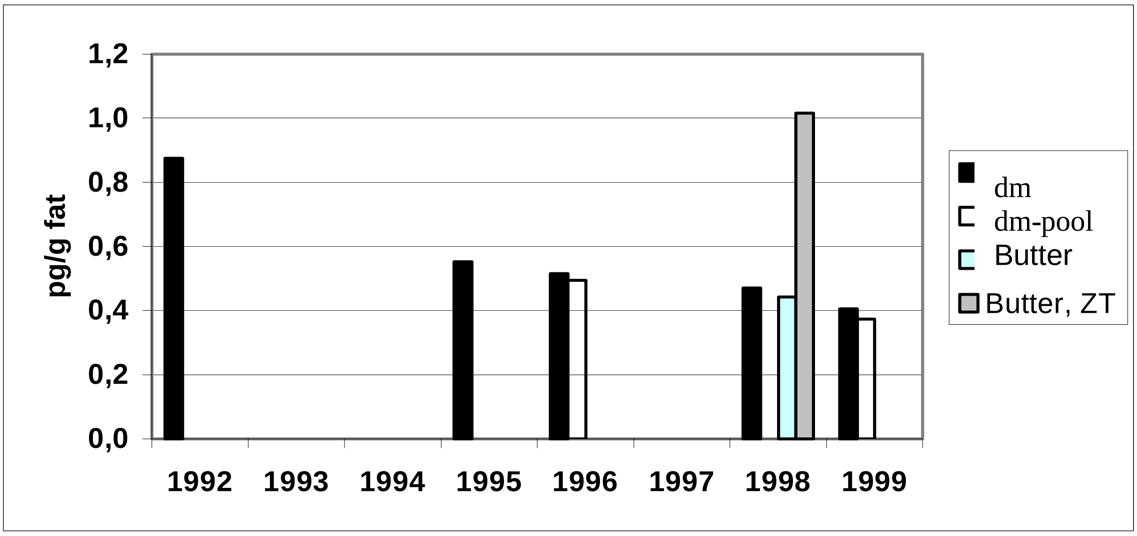


Fig. 29: Averages of dioxin concentrations in dairy products from Bavaria (Mm= dairy milk, ZT= citrus pulp)

North Rhine-Westphalia

Four times a year samples were taken in all dairies in 1990, 1994 and 1998 area-covering, i.e. in March/April (end of winter feeding), May (first cut), July (second cut) and September (before livestock housing). Due to the structure of the Land we may proceed on the fact that in this Federal Land a comparatively higher share of samples from urban areas or urban fringes is contained.

The results are compiled in Table 12, also here the decline of the dioxin concentration may be clearly recognized.

Table 12: Dioxin concentrations (pg I-TEq/g fat) in dairy products from NRW
* median without results of samples contaminated additionally by citrus pulp
** maximum caused by citrus pulp

Year	Median	Minimum	Maximum
1990	1.35	0,76	2.62
1994	1.02	0.61	1.75
1998	0.66*	0.47	1.78**

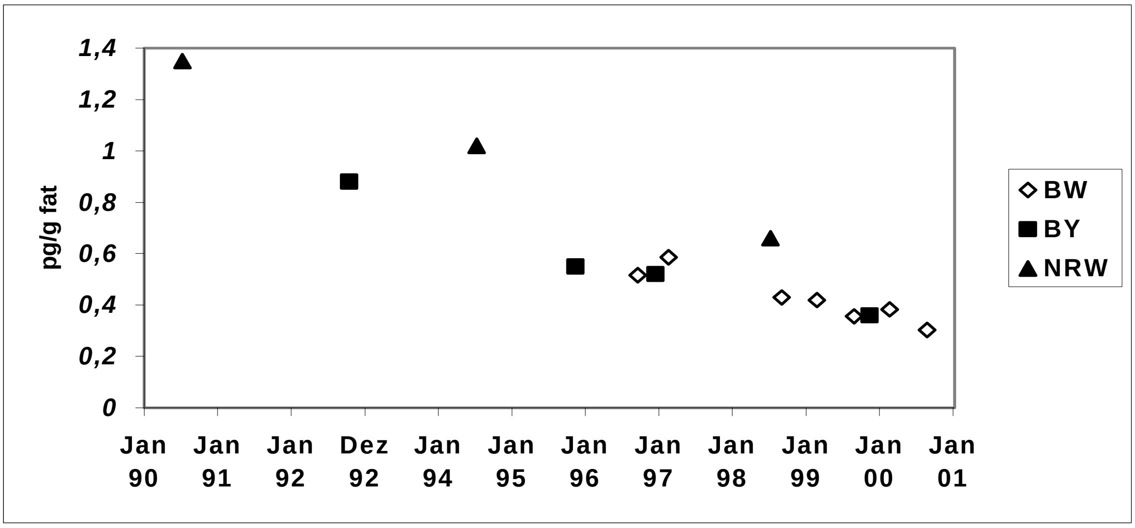
Summary

Summing up the results the dioxin contamination in Baden-Wuerttemberg declined by approx. 40 % or approx. 0.2 pg I-TEq between 1996 and 2000 (4 years), in Bavaria by approx. 60 % or approx. 0.5 pg I-TEq between 1992 and 1999 (7 years) and in North Rhine-Westphalia by approx. 50 % or approx. 0.7 I-TEq between 1990 and 1998 (8 years). These data prove a clear decline of the dioxin contamination in dairy products for each Land with its appropriate basic conditions differing due to sampling.

Calculation of the decline of concentration within the monitoring period

The decrease in concentration in the dairy products of the three Federal Laender is to be clearly recognized in Fig. 30. Here only data are contained which are collected at the same time within a year. The (negative) slope of the equalization lines (linear regression) is identical with the decline of the dioxin concentration between 0.06 and 0.09 pg I-TEq/g fat a year. In view of the various sampling conceptions and monitoring periods and the complicated analyses these are coinciding results.

Fig. 30: Trend of the average dioxin concentrations (I-Teq) in dairy products in three Laender



6.5.2.2 Bulk tank milk

In Baden-Wuerttemberg, Hesse and Lower Saxony 235 samples of bulk tank milk (raw milk) compiled in Table 11 were investigated between 1994 and 2000.

Baden-Wuerttemberg

Also these samples originate predominantly from rural areas without special dioxin emission sources, thus representing the background concentration. A small part of the samples was collected in a region with the establishment of industries (see Table 11). Sampling was carried out predominantly in the months of February and September.

The trend of the dioxin concentrations for all milk samples can be seen in Figure 31. As has been already detected in dairy products the clear increase in concentration in 1997/98 caused by citrus pulp is striking here, too. The dioxin concentration of samples from rural areas declines from 0.45 pg I-TEq/g fat, on average, in June 1996 to 0.36 pg I-TEq/g fat, on average, in September 2000.

The samples from areas with industries near by are slightly stronger contaminated by dioxins. Also in these samples the dioxin concentrations decreased in the monitoring period.

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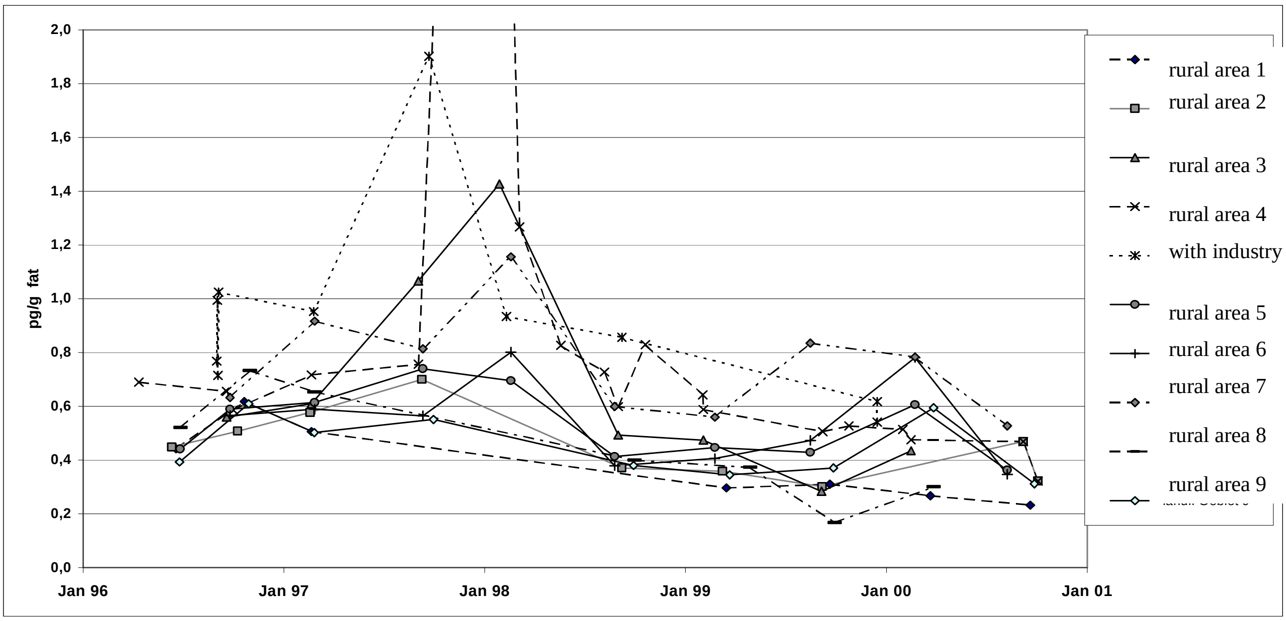
Fig. 31:Trend of dioxin concentrations (I-TEq) in tank milk from Baden-Wuerttemberg



For tank milk from 9 rural areas where samples were taken predominantly in September and February between 1996 and 2000 the concentration decline is calculated, on average, with 0.06 pg I-TEq/g fat a year (ascent of the equalization lines). Thus, a good agreement with the data derived for butter and cream has been reached. The decline is between 0.017 and 0.149 pg I-TEq/g fat a year (minimum-maximum) for the individual tours. Compared with this the respective variations in dairy products between 0.044 and 0.073 pg I-TEq/g fat a year were clearly smaller.

Due to the differently composed tank tours in the region with the establishment of industries the decline is not calculated for this area. However, we may proceed from a decline of the concentrations.

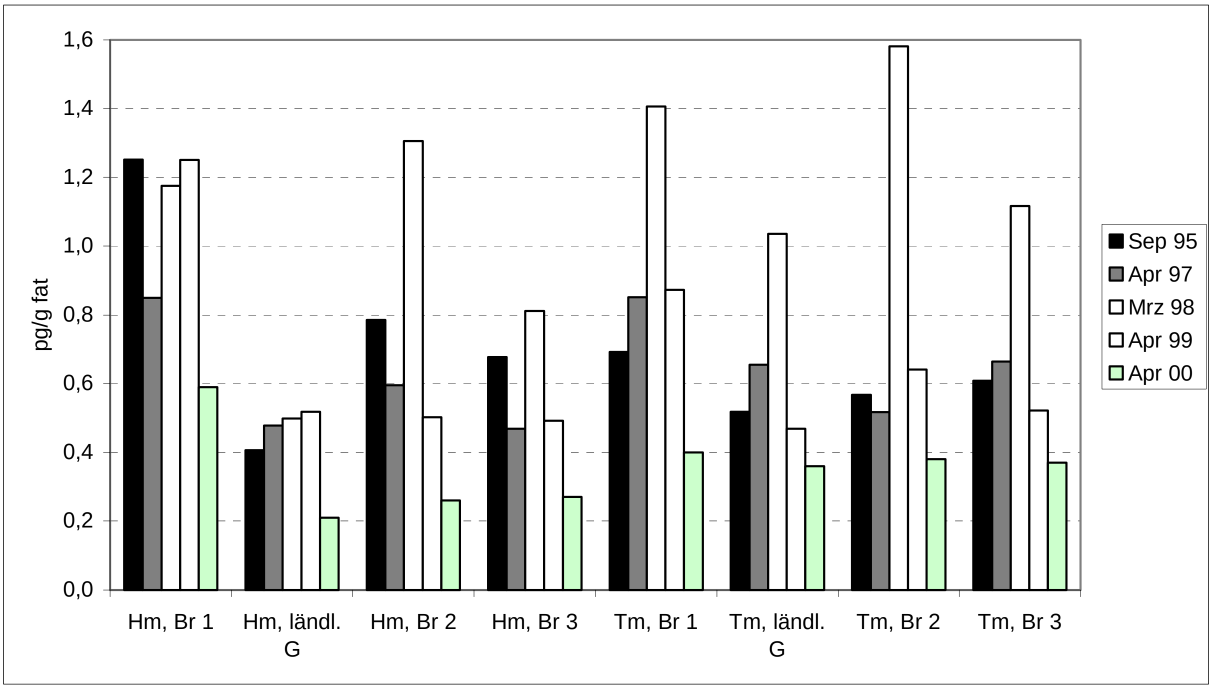
Hesse

The samples come from 3 regions of the South Hessian urban area and from a rural area (see Table 11). In 1995 sampling was carried out in September and between 1997 and 2000 at the end of March/beginning of April.

The trend of the dioxin concentration in Hessian tank milk between September 1995 and April 2000 is represented in Fig. 32 (together with farm milk). In this period the concentrations drop from 0.52-0.69 to 0.36-0.40 pg I-TEq/g fat (minimum-maximum). In all four locations remarkable contaminations by citrus pulp were to be observed in March 1998.

The contents in milk from 4 areas go down by 0.09 pg I-TEq/g fat a year, on average, between 1997 and 2000 (ascent of the equalization lines: maximum: 0.127, minimum: 0.030). The data of September 1995 were not used due to the deviating sampling month.

Fig. 32: Dioxin contents (I-TEq) in farm milk (Hm) and tank milk (Tm) in 3 areas of an urban area (Br1-3) and a rural area (G) in Hesse (each column corresponds to a sample)



Lower Saxony

In Lower Saxony sampling for tank milk from 4 rural regions covered the whole year. The temporal trend of the dioxin contents is represented in Fig. 33. During the various monitoring periods in the second half of the 90-ies the concentrations decrease from 0.4 – 0.6 pg I-TEq/g fat at the beginning to 0.2 – 0.5 pg I-TEq/g fat. In all four regions contamination by citrus pulp was to be observed in 1997/98 which was partly remarkable. Fig. 33 shows that the trend of concentrations is only difficult to detect due to the seasonal variations of the dioxin contents if sampling took place at various times within a year. Thus, a direct comparison of data is only possible if they were taken in the same season. Furthermore, at least a period of three years was to be covered. Results of samples keeping these preliminary conditions in the four tank tours are represented in Fig. 34. By means of this figure a decline of the contaminations is to be recognized at any time (March, June, December) of the respective monitoring periods, however with the trend of concentration being not always uniform. This is possibly connected also with changes of the tank tours resulting from shut down of dairies. A decline of the dioxin concentrations in the tank milk samples by 0.04 pg I-TEq/g fat a year (minimum-maximum: 0.02-0.06) is calculated through the ascent of the equalization lines as average for the 4 regions.

Fig. 33: Trend of the dioxin concentrations (I-TEq) in tank milk from four rural areas in Lower Saxony (ZT= additional contamination by citrus pulp)

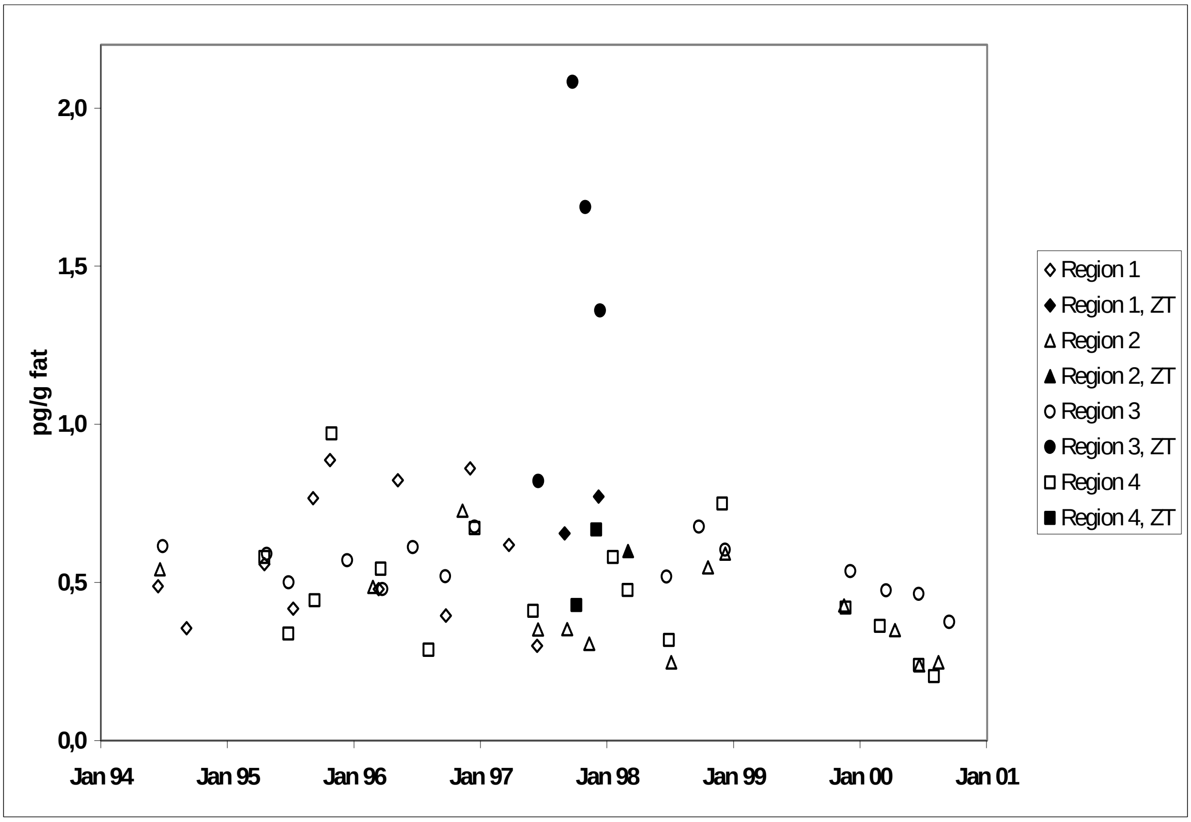
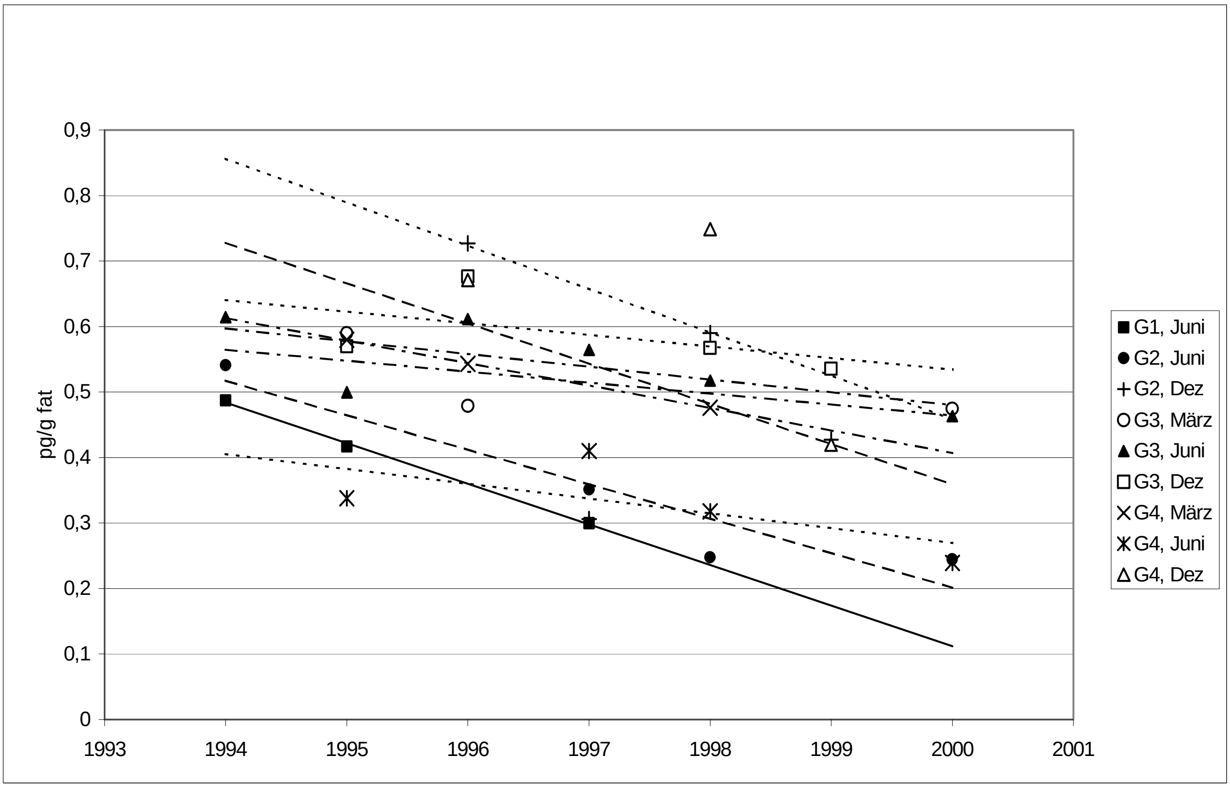


Fig. 34 Dioxin concentrations (I-TEq) in tank milk from four rural areas (G) in Lower Saxony in specific seasons



Summary

The decline of the dioxin concentration in tank milk from the three Federal Laender is between 0.04 and 0.09 pg I-TEq/g fat a year, thus being largely identical with the decline calculated for dairy products. Basically we can proceed on the fact that for an area-covering evaluation of the trend of concentration results on dairy products are more informative than those on tank milk. The bigger spreading of the results for tank milk supports this assumption. In view of the various monitoring periods and sampling conceptions and the difficult analyzing differences between the individual Federal Laender may not be derived.

6.5.2.3 Farm milk

Between 1989 and 2000 five Federal Laender investigated 456 samples of farm milk (raw milk). The details are shown in Table 11.

Baden-Wuerttemberg

Farm milk samples were taken in a rural area without special dioxin emissions in three farms, predominantly in the months February and September. The trend of the dioxin concentration is represented in Fig. 35. The dioxin concentration decreases from 1.05 (2 samples in November 1994) or 0.66 (October 1996) to 0.40-0.45 pg I-TEq/g fat (February 2000). In 1997/98 contamination by citrus pulp was to be observed in 2 farms. The otherwise practically congruent trend of concentration in 3 farms should be mentioned which, in addition, is within the respective range for tank milk and butter/cream from Baden-Wuerttemberg (see Figures 28 ad 31).

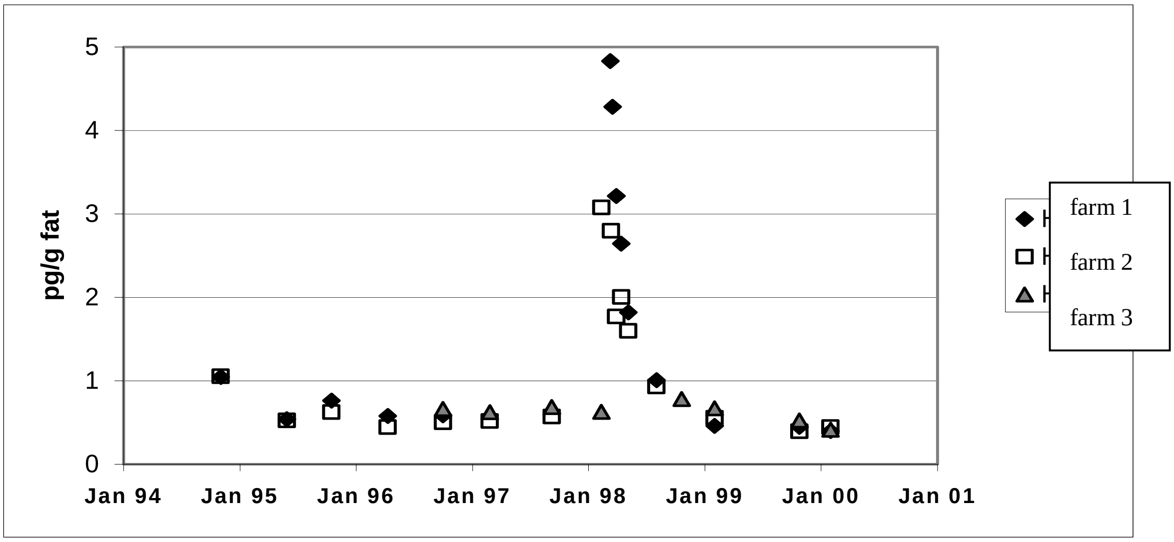


Fig. 35: Trend of the dioxin concentration (I-Teq) in milk from three farms in Baden-Württemberg

For the two farms where sampling has been started in 1994 the decline of dioxin concentration totals 0.107 and 0.109 pg I-TEq/g fat a year. For the third farm the decline is calculated as 0.050 pg I-TEq/g fat a year between 1996/97 and 1999/2000. Thus, the calculation of the results relating to a decline of the dioxin concentration for dairy products (cream, butter), collecting farm and tank milk in Baden-Wuerttemberg lead practically to the same results.

Bavaria

In Bavaria 156 samples were collected in 26 farms predominantly in the 4th quarters between 1989 and 1999. In the focal point of the investigations were 129 samples from rural areas from which it was not possible to exclude an additional contamination due to the vicinity of emission sources. Further details on the origin of these samples are compiled in Table 13.

Table 13: Origin of farm milk samples in Bavaria

Number of places	Type of contamination*)
6	background, thereof 1 unknown
3	Metal foundry (Alu), copper recycling
9	8 waste incineration plants (2 thereof with a refinery or chem. ind.), 1 waste oil incineration
5	4 chemical industry (1 thereof with waste incineration plant), 1 refinery with waste incineration plant
3	Cement works (1 thereof situated at a motorway)
2	Motorway, thereof 1 with cement works
1	Dumping site

*) Locations with 2 contamination sources were indicated twice

The trend of the dioxin concentration for all farm milk samples is shown in Fig. 36. Samples with a background concentration have average concentrations of 1.35 pg I-TEq/g fat in 1989. However, farm milk from areas with emission sources shows comparatively high average dioxin contents in this period (average 1989: 3.98 pg I-TEq/g fat, minimum-maximum: 2.36-5.62, average 1990: 2.58 pg I-TEq/g fat, minimum-maximum: 0.90-3.89, see Fig. 36). Yet, there are also samples among them the concentration of which is in the range of the background concentration.

In 1998 and 1999 the concentrations decreased clearly: samples with a suspected additional contamination have an average dioxin concentration of 0.53 pg I-TEq/g fat (minimum-maximum: 0.29-1.33 see Fig. 36) in these two years and samples with a background concentration 0.40 pg I-TEq/g fat (minimum-maximum: 0.28-0.61, see Fig. 36).

It is not possible to carry through an investigation considering the type of contamination as a too small number of samples is available and partly overlapping types of contamination are to be stated.

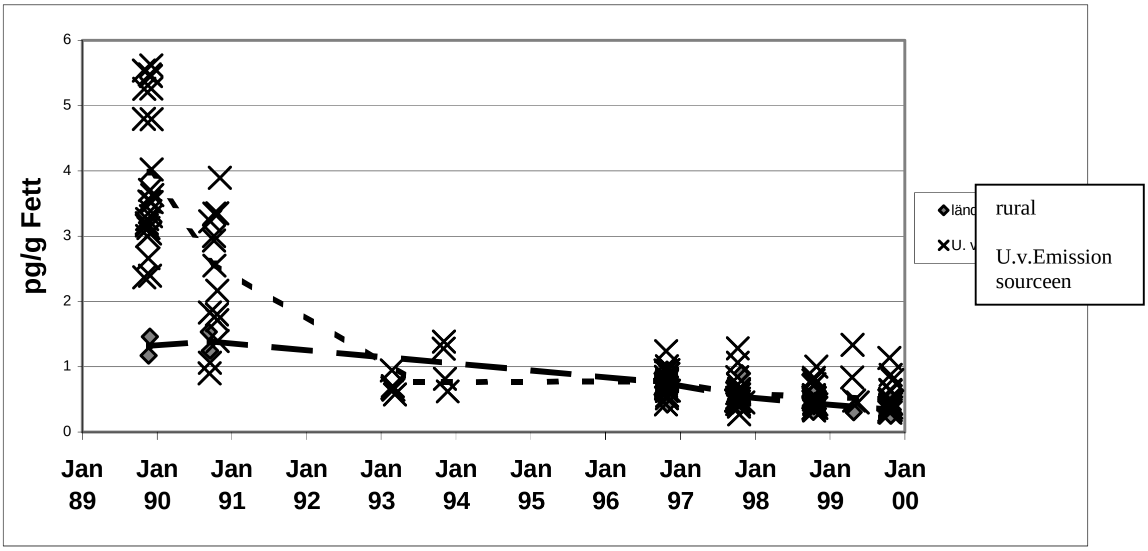


Fig. 36: Trend of the dioxin concentration (I-TEq) in farm milk from rural areas and the sourrounding of emission sources in Bavaria

Before calculating the decline of the dioxin contamination the samples from rural areas (background concentration) have to be separated from those from areas in the vicinity of emission sources. Between 1989/90 and 1999 the concentrations in rural areas decreased by 0.107 pg I-TEq/g fat a year, on average, (minimum: 0.087, maximum: 0.118) and in the areas in the vicinity of emission sources 0.323 pg I-TEq/g fat a year, on average, (minimum: 0.092, maximum: 0.482). The decline in heavily contaminated areas is thus especially high. In farm milk representing the background concentration the concentration declines stronger than in dairy products. This is presumably due to the slightly longer monitoring period in the case of farm milk samples as the decline of the concentration in the periods longer ago was bigger.

Hesse

In Hesse milk samples have been taken in 4 farms which, at the same time, form part of the tank milk samples described in the Tank Milk Chapter. The same sampling method was applied to the two types of samples.

In Fig. 32 the trend of dioxin concentration between September 1995 and April 2000 is represented (together with tank milk). The concentration declined from 0.41-1.26 to 0.21-0.59 pg I-TEq/g fat (minimum-maximum).

Two of the 4 farm milk samples do not show indications to a higher contamination by citrus pulp though all 4 collecting tank milk samples from March 1998 were remarkable. Therefore there should be assumed that an essential part of the milk in areas where samples were taken was additionally contaminated by citrus pulp in spring 1998. On the other hand, not all farms were affected by it. In farms which did not show remarkable contents in the milk in March 1998 mixtures of feedingstuffs were given the components of which did not contain citrus pulp.

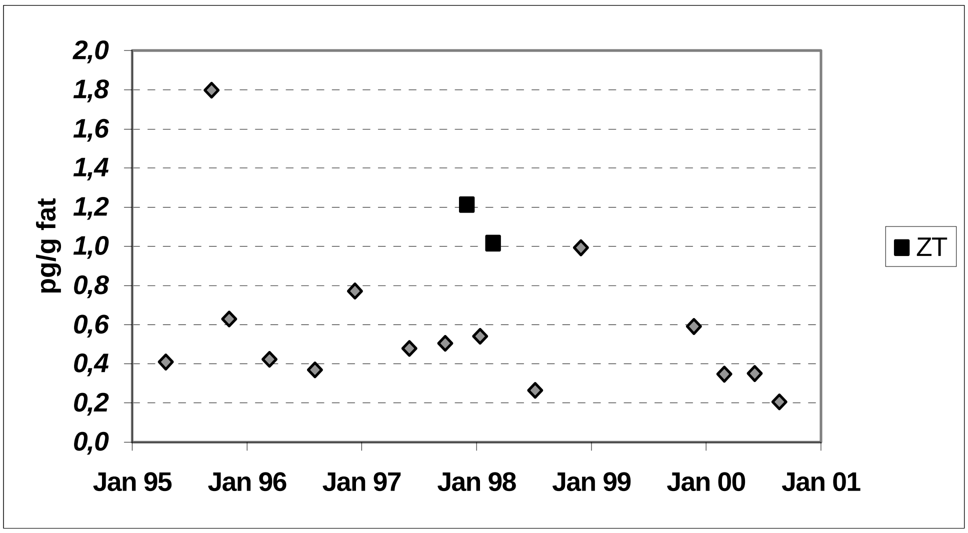
The contents in the milk from the 4 farms where samples were taken always in March/April declined between 1997 and 2000 (3 years) (without data of 1998 due to the contamination by citrus pulp and of 1995 due to the deviating sampling month) by 0.08 pg I-TEq/g fat a year, on average, (maximum: 0.103, minimum: 0.055). Therefore practically the same average decline of the dioxin concentration was stated in the farm and tank milk samples investigated here. The decline of concentrations calculated for farm and tank milk of an area, however, deviate clearly from each other.

Lower Saxony

Milk hygiene program

The trend of the dioxin concentration in farm milk of a region in Lower Saxony (background concentration) is represented in Fig. 37. The dioxin concentrations decline from 0.4-1.8 (1995) to 0.2-0.4 pg I-TEq/g fat (2000). In 1997/98 contamination by citrus pulp was observed. The trend of the concentration of these farm milk samples is within the respective range for tank milk from Lower Saxony.

Fig. 37: Trend of the concentration (I-TEq) in farm milk (milk hygiene program) of a region in Lower Saxony (ZT = additional contamination by citrus pulp)

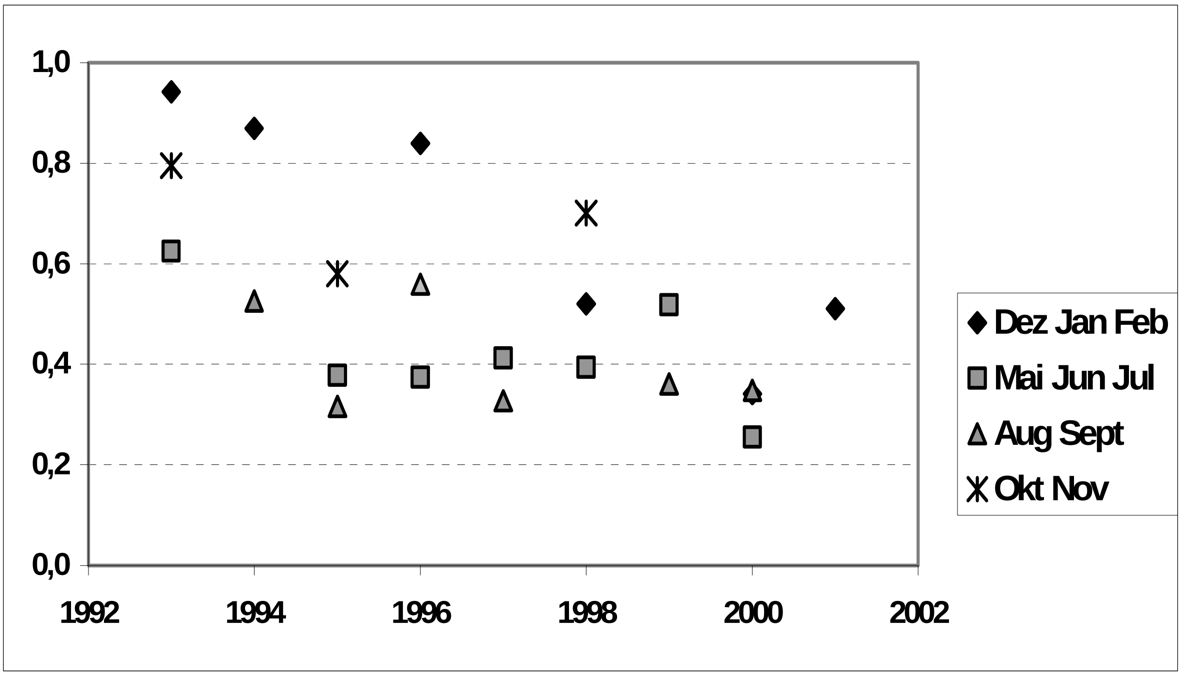


By means of the data available the decline in the concentration may be calculated for three periods (June, August/September, November/December) for intervals of 3 to 5 years. The characteristic decline for farm milk of this region is calculated roughly as average from the decline in concentration in the 3 periods totalling 0.09 pg I-TEq/g fat a year. It thus agrees well with the values from other Federal Laender. However, there is evident that the declines in the various periods differ clearly. As has been already stated for tank milk from Lower Saxony the trend of concentration is not uniform here, either.

Special farm milk program

In the framework of the special farm milk program 110 samples were investigated in 6 farms between 1992 and 2000. If the data of the periods December-February, May-July, August/September and October/November are compiled for each year from the data for judging the trend of the concentration a decline of contamination (0.03, 0.06 and 0.12 pg I-TEq/g fat a year) is detected in 3 farms in the course of 6 to 8 years. The decline calculated for the respective farms for the various comparative periods partly differ strongly from each other. In the case of a non-uniform trend of concentration an average decline by 0.07 pg I-TEq/g fat a year is calculated for the period between 1992 and 2000. In Fig. 38 this is shown by the example of farm 1. For a location consisting of three adjacent farms the monitoring periods in the individual farms are too short to make clear decisions.

Fig. 38: Trend of the concentration (I-TEq) in milk (special program) of a farm from Lower Saxony

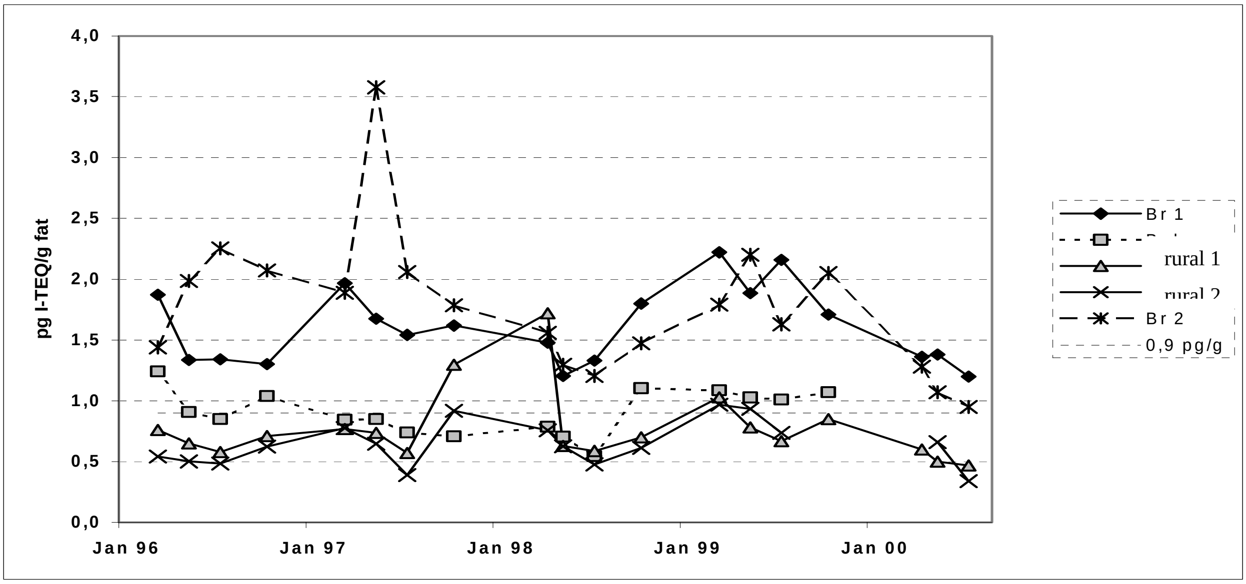


North-Rhine-Westphalia

In North Rhine-Westphalia 90 collected milk samples were investigated in five farms always in March/April, May, July and October between 1996 and 2000. Always two farms are situated in urban areas or rural areas. One farm is situated in an urban fringe.

A survey of the dioxin contents of all samples is given in Fig. 39. In the year 2000 in boths farms the average concentrations total 0.51 pg I-TEq/g fat in rural areas and 1.21 pg I-TEq/g fat in urban areas. In 1996 the respective concentrations amounted to 0.61 or 1.70 pg I-TEq fat and were thus higher. As has been expected, the dioxin contents of the samples from the urban fringe are between those of the two other areas, however closer to the samples from rural areas. The calculated average decline in the concentrations of 0.03 pg I-TEq/g fat a year between 1996 and 1999. This is to be regarded only as an estimate covering only the five farms chosen as an example.

Fig. 39: Trend of the dioxin concentrations (I-TEq) in milk from 5 farms in 2 urban areas (Br), 2 rural areas and in one urban fringe (Brd) from NRW



Summary

In Table 14 the results of the decline in the dioxin concentrations of farm milk from five Federal Laender are compiled. The declines are between 0.03 and 0.11 pg I-TEq/g fat a year (background concentration) and thus in a similar, yet slightly higher range than those detected for dairy products or tank milk. Due to the differing sampling conditions, the spreading of the results and a worse representiveness of the farm milk samples as compared with dairy products additional interpretations are not possible. It is, however, probable that in samples from the

surrounding of emission sources higher declines of the dioxin concentrations have to be stated than in samples from rural areas.

Table 14: Decline of the dioxin concentrations (pg I-TEq/g fat a year) in farm milk samples from five Federal Laender

Time	Difference	n farms	BW	BY	HE	NI	NRW
95-00	5	1				-0.09	
92-00	8	3				-0.07	
97-00	3	4			-0.08		
89/90-99	9/10	4		-0.11			
89/90-99	9/10	19		-0.32*			
96-00	4	5					-0.03
94-99	5	2	-0.11				
96/97-99/00	3	1	-0.05				

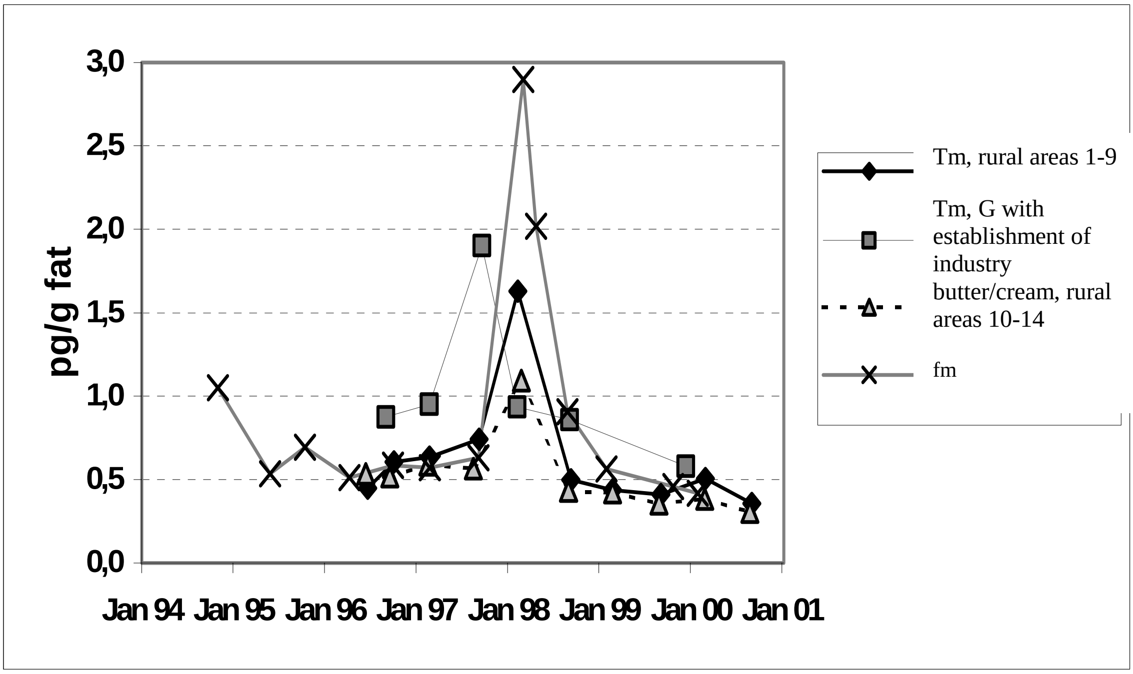
* Samples from areas near emission sources

6.5.2.4 Overall consideration of the results of dioxin concentrations in milk from the rural area

In Baden-Wuerttemberg all 3 types of samples (tank, farm milk and dairy products) from rural areas were investigated. Fig. 40 shows that the average concentrations of these samples were largely parallel in the course of time and thus nearly identical. In 2000 the dioxin concentrations were between 0.3 and 0.5 pg I-TEq/g fat with higher values – depending on the season – coming from the 1st quarter.

In Bavaria dioxins in dairy milk as well as farm milk were determined. In 1999 in both types of samples the same average dioxin concentrations of 0.4 pg I-TEq/g fat were determined for rural areas.

Fig. 40: Trend of the average dioxin concentrations (I-TEq) in tank milk (Tm), farm milk (fm) and dairy products in Baden-Wuerttemberg (rural areas (G) and areas with industries near by)



In four bulk tank milk samples from Hesse (rural area and urban area) the dioxin concentrations were between 0.36 and 0.40 pg I-TEq/g fat in April 2000. The collected farm milk samples from the same areas investigated at the same time showed concentrations between 0.2 and 0.6 pg I-TEq/g fat.

The dioxin concentrations in bulk tank and farm milk samples from Lower Saxony (rural areas) investigated in the year 2000 amounted to 0.2-0.5 or 0.2-0.7 pg I-TEq/g fat. Also here the samples from the 1st quarter were in the upper part of the indicated range.

The average concentrations in the two farm milk samples from rural areas in North Rhine-Westphalia amounted to 0.5 pg I-TEq/g fat in the year 2000.

To sum up there can be stated that the background concentration of the dairy products last measured in 1999 and 2000 in the two Federal Laender was between 0.3 and 0.5 pg I-TEq/g fat. The dioxin concentrations in bulk tank and farm milk samples measured in the other three Federal Laender at the same time allow to draw the conclusion that the background concentration must be similar there. Thus, the dioxin contamination has clearly declined as compared with the preceding years.

The decline of the dioxin concentrations in the dairy products studied here is between 0.06 and 0.09 pg I-TEq/g fat a year or 6-10 % a year in various monitoring periods. The respective ranges are slightly higher for farm and tank milk. This is plausible because dairy products are made of farm and tank milk.

Because of various types of samples, monitoring periods and sampling conceptions, however, comparisons between the individual Laender are only possible on a limited scale. Altogether a decline of contamination by more than 60 % may be derived from the data presented during the past decade.

6.5.3 Samples from urban areas, an urban fringe and rural areas with emission sources in the vicinity

Baden-Wuerttemberg

The TEq contents in the tank milk from rural regions influenced by industries are with 0.82 pg I-TEq/g fat (minimum-maximum: 0.54-1.03, without citrus pulp) by 55 % higher than the respective contents in the samples with background concentrations (0.53; minimum-maximum: 0.17-0.92 pg I-TEq/g fat). It looks as if the differences diminished in 1999 (see Figs. 31 and 40).

In dairy products from the more densely populated rural area the concentrations are slightly higher (no. 14, see Fig. 28).

Bavaria

The dioxin contents of collecting farm milk samples from rural areas with emission sources in the vicinity were more than twice as high (factor 2.5)) in 1989/90 (average: 3.49 pg I-TEq/g fat) than those from rural areas (average: 1.35 pg I-TEq/g fat). In the second half of the 90-ies the difference diminished to approx. 20 % (factor 1.2). If the background concentration of dairy milk (see Fig. 36) is used for comparison the difference between rural area without and with emission sources in the vicinity totaled approx. 35 % for the second half of the 90-ies.

Hesse

The tank samples from the urban area are with their dioxin contents by 20 % above those from the emission-distant area. In the case of collecting farm milk the respective difference is with 69 % essentially higher (see Fig. 32).

North Rhine-Westphalia

The milk from the two selected farms from the urban areas is clearly higher contaminated than that from rural areas (factor 2.5). As has been expected the dioxin contents of samples from the urban fringe are between those of the two other areas, however closer to the samples from rural areas. The differences of contamination were the smallest in the year 2000 (see Fig. 39).

Summary:

Samples from urban areas, an urban fringe and rural areas in the vicinity of emission sources are comparatively higher contaminated by dioxins than samples with background concentration. This refers notably to collecting farm milk due to the purposeful sampling in view of the vicinity of dioxin sources. The differences between the samples quantified for the individual Laender are exemplary, yet not representative.

The differences of the concentrations in samples from contaminated and rural areas have obviously diminished in the last few years.

Chapter 6.5.4 deals with the differences of the congener distribution in these samples.

6.5.4 Congener distribution

The evaluations are restricted to the congeners important for TEq with TEF $\geq$ 0.1 (except 2,3,7,8-TCDF), as congeners with TEF < 0.1 and 2,3,7,8-TCDF are frequently in very small concentrations, thus being not reliably detectable. When looking through the data samples remarkable <u>only</u> due to contents of congeners with TEF < 0.1 and 2,3,7,8-TCDF were not found either.

In all samples with an increasing degree of chlorination the PCDD concentrations increase whereas in PDCF 2,3,4,7,8-PeCDF and HxCDFs dominate. Classified according to the various types of contamination (background, citrus pulp, urban area including rural areas in the vicinity of emission sources) the distribution of individual congeners is indicated in Table 15 in the form of their respective share in the TEq.

It may not be recognized whether the shares of individual congeners in the TEq in samples with background concentration change between the years or in the course of the years. Therefore we should proceed from a largely constant pattern. 2,3,4,7,8-PeCDF, 1,2,3,7,8-PeCDD, 2,3,7,8-

TCDD, HxCDFs and HxCDDs (in a decreasing order) supply shares of 10 % or more to the I-TEq.

As in the WHO-TEq the share of 1,2,3,7,8-PeCDD increases clearly as against that of I-TEq due to its higher factor the shares of other congeners go down (with the order remaining equal). Thereby, the shares of HxCDDs are reduced to slightly below 10 %. 2,3,4,7,8-PeCDF and 1,2,3,7,8-PeCDD with approx. 61 % contribute together to I-TEq and with approx. 67 % to WHO-TEq.

In samples from urban areas, urban fringes and areas with industry and contaminated areas the congener pattern changes insignificantly. Compared with samples from rural areas an increase in the shares of PCDFs, HxCDFs and 2,3,4,7,8-PeCDF for these samples at the costs of shares of PCDDs, HxCDDs and 1,2,3,7,8-PeCDD may be detected altogether for these samples.

Table 15: Shares (%) of the dioxin congeners in the TEq and PCDD/F-WHO-TEq in milk products from rural areas (background concentration), from urban areas (including rural areas in the vicinity of emission sources) and with additional contamination by citrus pulp

	TCDD	**PeCDD**	**HxCDD**	**23478-PeCDF**	**HxCDF**	**other**	**PCDD**	**PCDF**
I-Teq								
rural areas	13	16	10	45	12	4	41	59
urban areas	13	14	9	48	13	3	36	64
citrus pulp	33	17	6	31	11	2	56	44
PCDD/F-WHO-Teq								
rural areas	11	28	9	39	10	3	49	51
urban areas	11	25	7	42	12	3	44	56
citrus pulp	28	29	5	27	9	2	62	38

6.5.5 WHO/I-TEq ratio

The PCDD/F-WHO-TEq (without PCB) in milk fat are by 14 – 18 % higher than the I-TEq – largely independent from the year, type of sample and Federal Land.

In spite of a different distribution of the congeners the samples measured in connection with citrus pulp do not attract attention by a different ratio between the two TEq.

6.5.6 Seasonal changes of dioxin concentrations in the course of a year

For comments on the seasonal trend the results from the period between July 1997 and the end of 1998 will not be considered due to the special contamination by citrus pulp.

Baden-Wuerttemberg: If the results for bulk tank milk and butter/cream are summed up you can calculate from the data sets dioxin concentrations by 4-9 % higher at the end of the winter (February/March) compared to autumn and by 18 % higher than in summer (August).

Bavaria: Results relating to dairy milk (Mm) or pooled dairy milk (Mm-P) from 1996 and 1999 are especially suited for the seasonal comparison of the dioxin concentrations. The evaluations show that the dioxin concentrations in spring (April) are by 15 % higher than in autumn (November) and are nearly equal in spring (March) and summer (July) (in summer by 5 % higher).

Lower Saxony: Comments on the seasonal trend may be derived from the data collected in the framework of the special program. The dioxin contents in farm milk go up in the years 1993-2000, on average, from the months May/June/July to August/September by 20 % (4 farms), to October/November by 48 % (4 farms) and to December/January/February by more than 60 % (2 farms). As here a few farms of a closely limited area are concerned the calculated data are not to be regarded as representative. Fig. 38 shows the remarkable seasonal fluctuations by the example of farm 1. Obviously the same seasonal trend exists also for tank milk (Fig. 33).

North Rhine-Westphalia: By means of a few locations for collecting farm milk comparisons of the dioxin concentrations in the course of a year are not appropriate as two locations have to be assigned to urban areas and contamination by citrus pulp is detected. On the other hand, in Fig. 39 can be seen that – similar as in Baden-Wuerttemberg and Lower Saxony – the concentrations are on the lowest level in summer and are clearly higher in spring and in autumn. This is in accordance with a comprehensive series of investigation of dairy milk in North Rhine-Westphalia in 1990, 1994 and 1998.

Summary

The dioxin concentrations vary in the course of a year by approx. 20 %. This result underlines convincingly that only samples covering the same period should be used as a prerequisite to making comments on the trend.

In Baden-Wuerttemberg, Lower Saxony and North Rhine-Westphalia a concurrent seasonal trend of concentration was detected. At the end of the winter it shows the highest concentrations which subsequently decline clearly during grazing (spring and summer) increasing again towards autumn. As against this a different trend of concentration was detected in Bavaria where the highest contents occurred in summer. Whether the two different ways of trend of the concentration in the course of a year depend predominantly on feeding and how far calving or other influences have to be considered may not be clarified for the time being.

6.5.7 Exceeding of the target value of the Government/Laender working group on DIOXINS

In its 2nd report of 1993 the Government/Laender working group on DIOXINS suggested a target value (0.9 pg I-TEq/g fat) of dioxin concentrations in milk which in the long term should be achieved by reducing the dioxin inputs into environment or falling below them. This value was derived from consumption data in connection with the precautionary value for the daily intake of dioxins of 1 pg I-TEq/kg KG per day fixed at that time.

For dairy products from Baden-Wuerttemberg all samples are below the target value of the Government/Laender working group since 1996. However, the time of contamination by citrus pulp in the years 1997/98 (Fig. 28) is excluded. In 1992 the dioxin concentrations of 67 % of the dairy milk samples from Bavaria remained below the target value. Subsequently all samples – except a few samples with contamination by citrus pulp and another sample – fulfill the requirements of the target value.

In table 16 the share (%) of dairies in North Rhine-Westphalia is indicated where samples which remained below the target value were taken area-wide. Cases remaining below the target value increase clearly. Excluding the samples additionally contaminated by citrus pulp in 1998 nearly 90 % of the samples in this year might fall below the target value.

Table 16: Shares (%) of dairies in NRW the dairy products of which remain below the target value (0.9 pg I-TEq/g fat) recommended by the Government/Laender working group on DIOXINS

Year	Number of dairies	% < target value
1990	43	5
1994	30	31
1998	29	76*

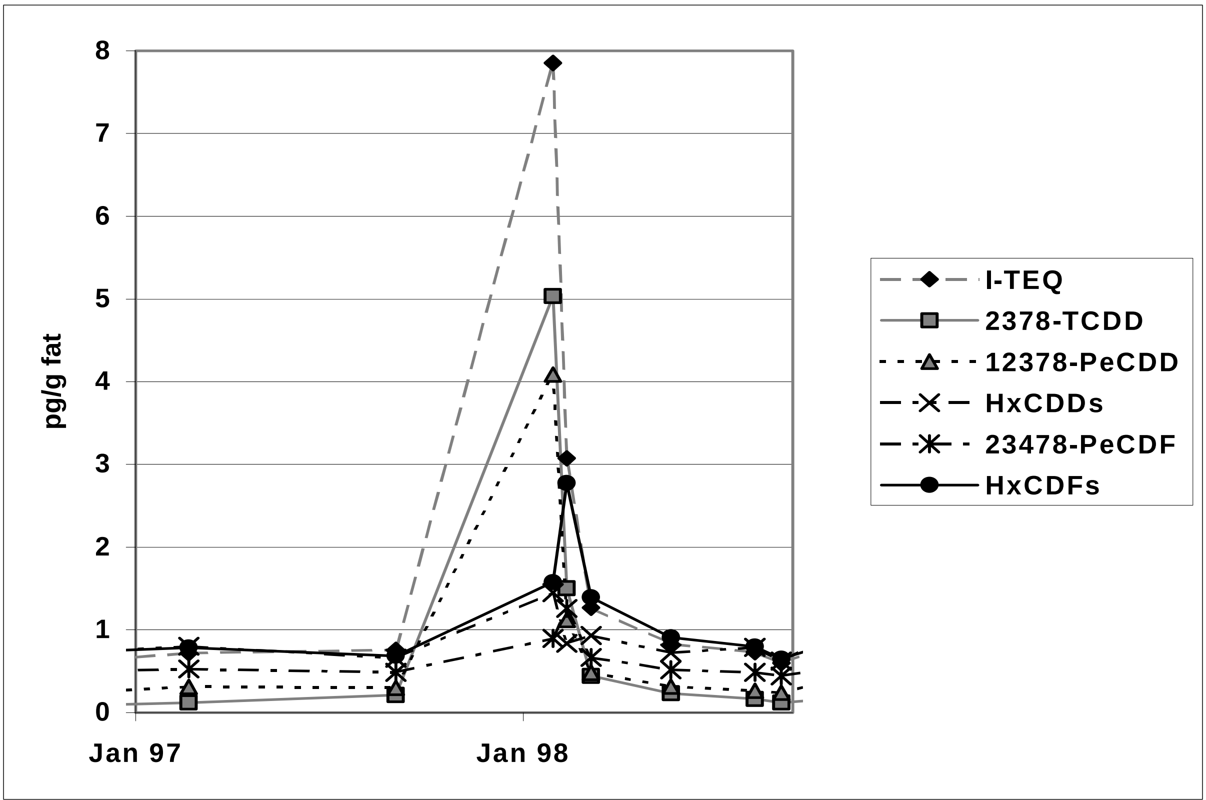
* contain samples contaminated additionally by citrus pulp.

To sum up there can be stated that due to the general decline of the dioxin contaminations the target value in dairy milk samples was exceeded only in individual cases in the last few years. However, in 1997/98 due to the contamination by polluted citrus pulp preliminarily higher rates of exceeding (see Figs. 28 and 29) were detected. In the early 90-ies this rate was by 95 %. The same comments may be derived for tank and farm milk.

6.5.8 Citrus pulp

In 1997 there was noticed in Baden-Wuerttemberg that the trend of a gradual decline of the dioxin contents in milk samples observed since the early 90-ies was reversed: the samples collected until August showed dioxin contents of 0.62 pg I-TEq/g fat, on average, whereas the samples collected as of September showed 0.89 pg I--TEq/g fat, on average. Hereby, this increase was always caused by the same congener pattern. The rising trend continued in the samples collected early in 1998 increasing in an individual sample (tank milk) up to exceeding the recommended guide value for the marketability of milk. Apart from 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD also HxCDFs, 2,3,4,7,8-PeCDF and HxCDDs increased in these samples (in a declining order). In Fig. 41 the trend of the concentration of these congeners in milk of this especially remarkable tank tour (rural area no. 4, see also Fig. 31) is represented.

Fig. 41: Increase in the dioxin concentration in tank milk of a rural area in Baden-Wuerttemberg by contaminated citrus pulp



The determination of the causes was difficult as dioxins appear always only as pollution or by-products and many different products or processes had to be checked. Apart from that, there was clear that this was not a local problem but a source of a transregional significance had to be detected: Two butter samples taken late in January in 1998 in Schleswig Holstein with 1.41 or 1.37 pg I-TEq/g fat showed similar high contents as butter samples produced in Baden-Wuerttemberg which were collected at the same time (s. Fig. 28). Also commercial milk samples from other Federal Laender collected in the framework of the official food control in Baden-Wuerttemberg showed the same trend (increasing dioxin contents caused by the same dioxin pattern). Equally in all five Laender participating in the dioxin reference measuring program where milk was investigated a clear increase in the dioxin concentration was detected in this period (see Figs. 28, 29, 31, 32, 33, 35, 37, 39, 40 of Chapter 6.5.2).
Alarming dioxin results of February 1998 in the tank milk tour from the rural area no. 4 and in the respective farm milk samples were the key to clarifying the cause.

This region in the Upper Rhine plain has been subject to regular investigations since 1994 as on the opposite Rhine side in the Alsace chemical industry had been established from which dioxin emissions were feared. However, the investigations of plant foodstuffs and milk did not show an increase in the dioxin content. Therefore the highest alert phase was proclaimed when samples taken from the tank milk from this region in February 1998 showed 7.86 pg I-TEq/g fat (rural area no. 4, Figs. 41 and 31). This was a content by 10 times higher than it had been detected in this collecting milk still early in September 1997. Immediately all larger milk supplying farms of this tour were checked. In this connection, one farm with 4.83 pg I-TEq/g fat was detected which in the past had also always inconspicuous dioxin contents (farm 1, rural area 4, Fig. 35). As a modification of the production conditions was not to be detected in this farm all potential causes were checked: use of disinfectants, cleansing agents, pesticides, animal pharmaceutics, construction materials and feedingstuffs. By means of various samples it was then possible for the Chemische Landesuntersuchungsanstalt Freiburg (Regional Institute of Chemical Investigation) to find the cause in March 1988. Citrus pulp imported from Brazil contained in the milk producing feedingstuffs up to 25 % was highly contaminated by dioxins.

Comprehensive additional investigations ensured the study of causes: it was possible to detect in 10 various farms checked immediately a clear correlation between feeding with citrus pulp and dioxin content of the milk. A further important member in the chain of evidence proving that the correct cause has been found was the decline of the dioxin content after stopping feeding with the contaminated feedingstuff in the two farms where the highest dioxin contents had been detected. The peak contamination declined in the course of a few weeks.

About one year later the dioxin content of all other samples contaminated by citrus pulp were again in the range of the formerly usual background concentration (see. Figs 28,31,35, 40 and 41). In the other Federal Laender a similar trend of the contamination was to be observed (see Figs. 29, 32, 33, 37 and 39 of Chapter 6.5.2).

The economic significance was essential: The world market of citrus pulp production comprises approx. 1.5 million tons a year of a value of approx. US $ 100 to 150 million. About 60 % of this production come from Brazil, with the production period covering there about the period from September to February. Thereupon follow primarily US products. In 12 member countries of the European Union still 92 000 t of citrus pulp of Brazilian origin were met in March 1998 which had to be removed from the market and to be disposed.

On 24/07/1998 the Standing Committee for Feedingstuffs of the EU fixed a preliminary maximum quantity of 500 pg I-TEq/kg for citrus pulp which by all member states had to be immediately included in their national law.

In January 1999 a mission of an EU delegation to Brazil was organized to learn the state of the study of causes and to check the measures taken by Brazil to avoid the incident to happen again. Thereby, it turned out that contaminated lime had caused the contamination of citrus pulp by dioxins: lime is used for neutralizing the peels after pressing out the juice. In Brazil there exist two types of lime:

- lime extracted from lime deposits, thereupon burned and
- lime coming from processes of chemical industry.

In the present case lime from a chemical production process had been sold to the citrus pulp industry. The lime causing the contamination of the citrus pulp contained up to 2500 pg I-TEq/g of dioxins.

Brazil fixed the same dioxin tolerance for citrus pulp which had been fixed by the EU stipulating in addition, that this limit applies also to lime processed by the citrus pulp industry. Apart from that, comprehensive control measures were fixed to be carried through on all levels of production, storage and transport for the lime supplying industry and the citrus pulp production. In 1999 two EU missions were organized to Brazil the aim of which was to check the efficiency of the measures taken.

The effects of the contamination of milk by citrus pulp are explained by means of 10 tank milk samples in Baden Wurttemberg contaminated most heavily by citrus pulp: The average I-TEq contents of these samples are essentially higher than those of the samples with background concentration (difference: factor 3.7). 2,3,7,8-TCDD and 1,2,3,7,8-PeCDD are remarkable in congeners with average contents by nearly 13 or 5 times higher whereas the contents of HxCDFs, 2,3,4,7,8-PeCDF and HxCDDs go up by 82, 53 or 22 % (Fig. 42). In the case of the contamination by citrus pulp being lower the increase for the last 3 congeners may scarcely or not be detected.

The changes of the congener concentrations in milk samples contaminated by citrus pulp have effects on the shares of individual congeners in the TEq: In these samples the shares (%) of 2,3,7,8-TCDD - and thus the sum of the PCDD - increase in the TEq essentially at the costs of the sum of the PCDF as compared with the samples with background concentration whereas all other shares in the remaining congeners – with the exception of the 1,2,3,7,8-PeCDD – are reduced (see Table 15).

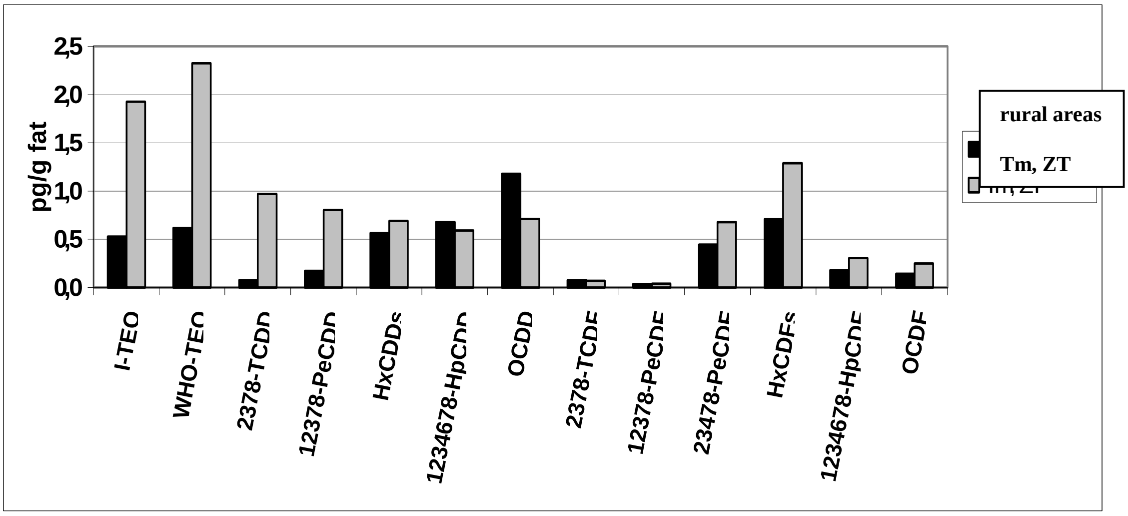


Fig. 42: Average dioxin concentrations in tank milk (Tm) from rural areas (Ge, 1996-2000) and with a clearly additional contamination by citrus pulp (ZT, 1997/98)

To estimate the extent of the additional contamination by citrus pulp the number of all samples investigated in addition to the number of samples contaminated by citrus pulp in the period known for the contamination (1997/98) for each Federal Land are indicated in Table 17. Thereof the conclusion can be drawn that a high share of samples 1997-98 was contaminated by citrus pulp.

In Bavaria the extent was presumably the lowest. In Hesse and Lower Saxony no area-covering comments can be made due to the investigations having been restricted to collecting tank and farm milk.

As the Brazilian citrus pulp was supplied to Rotterdam the convenient „Rhine route“ is to be regarded as an important way for its further transport to south. Therefore the high share of samples contaminated by citrus pulp from Baden-Wuerttemberg and North Rhine-Westphalia seems to be plausible.

Table 17: Number of milk samples contaminated additionally by citrus pulp (ZT) as compared to all samples investigated in this period

Land	Dairy products		Tank milk		Farm milk	
	all	ZT	all	ZT	all	ZT
BW	4	4	8	8	3	2
BY	18	3			26	0
HE			4	4	4	2
NI			4	4	6	2
NRW	29	17			5	2

The dioxin concentrations in all samples contaminated by citrus pulp and measured in the framework of the reference measuring program are, on average, by a factor of 2.5 higher than the background concentration. The daily dioxin intake of humans through the whole food would thus go up by approx. 50 % - due to the high share of milk fat in the food fats. Other foodstuffs (meat) also contaminated by citrus pulp remain unconsidered. Therefore there cannot be excluded that in Laender with a widespread feeding of citrus pulp in foodstuff production this could have effects on the internal dioxin impact on the population if the duration of this additional impact would be estimated at approx. half a year. The results of dioxin concentrations in human samples from Baden-Wuerttemberg and North Rhine-Westphalia seem to allow such an interpretation. Further subjects of discussion relating to this field will be dealt with in detail in Chapter 6.6.6.

6.6 Human data

6.6.1 Preliminary remarks

Investigations of human milk for dioxins were mainly carried through by the Federal Laender Baden-Wuerttemberg, Lower Saxony and North Rhine-Westphalia, to a smaller extent, among others, by Bavaria, Schleswig-Holstein and by the former BgVV. The vast majority of the samples was analyzed upon request of mothers. Sampling or selection of samples was not carried out according to defined criteria but was purely accidental. The target of the dioxin reference measuring program to repeatedly investigate samples from representative and identical locations may not be applied to the investigation of human milk due to the specificity of sampling.

From the comprehensive data material available reliable comments on the trend of the background concentration of human milk by dioxins may be derived. Therefore the decision was made to include also investigations of human milk in the report of the dioxin reference measuring program supplementary to the purposeful investigations in the environmental and foodstuff media. This is to enable a comparison of the temporal trends of the various environmental media up to the final member of the food chain, the man, thus describing as comprehensively as possible the overall situation. At the same time, it is possible to check by means of the temporal variation of the dioxin contamination of human milk which may be regarded as bioindicator for the „internal“ impact on humans to what extent technological and regulatory measures aimed at minimizing the dioxin emission and the exposure of humans are successful.

Apart from the investigations of human milk Baden-Wuerttemberg carries out interdisciplinary investigations, among others, for dioxins in the blood of 9- to 11-year old school children in the framework of the „monitoring health departments“ project. These investigations show a defined design of investigation and take place in 4 selected locations at intervals of 1 to 2 years. Thereby the probands have to meet specific criteria of selection.

The data collected in this way draw a representative picture of the internal impact on school children by dioxins and its trend conforming with the requirements defined by the dioxin reference measuring program. The Land Baden-Wuerttemberg itself took over the evaluation and compilation of these data.

6.6.2 Data collection in the human milk and dioxin human data base

The data on dioxins in human milk and dioxins in blood were transmitted by the Laender to the human milk and the dioxin human data base of the Government and the Laender set up by the former BgVV since 1991 and developed gradually further. Nationwide investigations of persistent and lipophile residues in human milk (organochloropesticides, PCB, dioxins, synthetic musk scents) and of dioxins in further human samples are included in this data base. About 10 % of the stored data sets concern investigations of dioxins. Hereinafter there shall be reported on these facts.
In the first stage of setting up this data base - referring to data of 1991-94 – the Laender transmitted to the former BgVV basically analytical results of 17 2,3,7,8 substituted congeners investigated in human milk (except Baden-Wuerttemberg 1992) which as individual data were included in the human milk data base.

Further information about the mother or the sample background is available only for a small part of the samples for this period.

Supplementary to the analytical data the Laender supplied data on the age, number of breast-feeding periods and the origin of the mother (old or new Federal Laender, foreign origin) for the subsequent period between 1995 and 1999 as these factors may affect the dioxin contamination of the mother.

In the third stage the interlink age of the human milk data base at the former BgVV with the dioxin data base at the UBA and a more comprehensive documentation of numerous exposure factors are implemented. To this end, together with the Laender a collection form for residues in human milk and dioxins in further human matrices was developed which served for the uniform documentation of relevant influencing parameters, description of methods and analytical data. This collection form was harmonized with the requirements of the dioxin data base as far as this was possible and necessary. It was technologically implemented by a data processing system jointly used with the dioxin data base. Thus, the prerequisites as to the content and technology were provided for a joint inter-media evaluation of dioxin data in the environmental, foodstuff and human sectors.

In the human milk and dioxin human data base in the former BgVV relevant person-related data are stored. The former BgVV is responsible for observing the provisions of the Federal Data Protection Act. The former BgVV reported to the Federal commissioner for data protection on the provisions envisaged for ensuring data protection (coding, anonymizing, access rights). After checking the documentation he states summarizingly that there are no objections to the method and the envisaged data processing from the viewpoint of data protection.

6.6.3 Data inventory of the human milk and dioxin human data base for dioxin investigations

As of 31.1.2000 results of 920 dioxin analyses including 3057 samples have been transmitted to the data base. The data are classified as follows:

human milk:	1037 samples with 888 analyses
blood:	2020 samples with 32 analyses

The Laender Lower Saxony, Baden-Wuerttemberg, Bavaria and Schleswig-Holstein made available the measured values of 17 2,3,7,8-substituted dioxin and furan congeners for each analyzed human milk sample (exception BW 1992). Measured values from North Rhine-Westphalia for the years 1991-94 are available as aggregate data (average, median, minimum, maximum for the congeners), for the years 1995-98 also as individual data.

The data from the Laender are transmitted in the structures described for the first or second stages of setting up the data base. A subsequent collection and transmission of the supplementing parameters inquired by the collection form for the investigations of human samples for dioxins carried out since 1991 was neither possible for reasons of data protection law nor implementable by the Laender. Comments as to the analytical methods and the location or exposure-related evaluations as they have been carried out for environmental media are therefore, as a rule, not possible for data from the Laender. The data of the former BgVV have been already documented on the basis of the collection form.

In Tab 18 a compilation of the data transmitted and stored is represented.

Tabe 18 Data inventory of the human milk and dioxin human data base (as per 31.1.2000)

Federal Land	Year	Matrix (FM=human, B= blood	Number of collected samples	Number of analyzed samples	Pool samples	Measued values of each analysis	aggregated measued values	Data of 17 congeners	I-TEq	Age of	Number	Origin	Further	Remarks
NI	1991	FM	77	77		X		X	X	X	X	X	X	Personal data partly
	1992	FM	109	109		X		X	X	X	X	X	X	Personal data partly
	1993	FM	45	45		X		X	X	X	X	X	X	Personal data partly
	1994	FM	34	34		X		X	X	X	X	X	X	Personal data partly
	1995	FM	83	83		X		X	X	X	X	X	X	Personal data partly
	1996	FM	18	18		X		X	X	X	X	X	X	
	1997	FM	15	15		X		X	X	X	X	X	X	
	1998	FM	25	25		X		X	X	X	X	X	X	
NRW	1991	FM	111	111			X	X	X					
	1992	FM	56	56			X	X	X					
	1993	FM	78	78			X	X	X					
	1994	FM	50	50			X	X	X					
	1995	FM	38	38		X		X	X	X	X	X		
	1996	FM	22	22		X		X	X	X	X	X		
	1997	FM	11	11		X		X	X	X	X	X		
	1998	FM	15	15		X		X	X	X	X	X		
BW	1992	FM	6	6		X			X					
	1993	FM	5	5		X		X	X					
	1994	FM	6	6		X		X	X					
	1995	FM	14	14		X		X	X					
	1996	FM	41	9	X	X		X	X					Personal data due to pool
	1997	FM	82	11	X	X		X	X					samples not possible
	1998	FM	23	6	X	X		X	X					dito
SH	1991	FM	3	3		X		X	X	X	X	X		
	1992	FM	4	4		X		X	X	X	X	X		
	1993	FM	3	3		X		X	X	X	X	X		
BY	1992	FM	2	2		X		X	X					
	1997	FM	18	3	X	X		X	X	X	X		X	
	1998	FM	6	1	X	X		X	X	X	X		X	
	1999	FM	2	2		X		X	X					
BE (BgVV)	1993	FM	10	1	X	X		X	X	X	X	X		WHO field study
ST (BgVV)	1990/91	FM	9	9		X		X	X	X	X	X	X	contaminated area
	1997	FM	10	10		X		X	X	X	X	X	X	contaminated area
BW	1993/94	B	158	3	X	X		X	X	X		X		Project monitoring health departments
	1994/95	B	452	5	X	X		X	X	X		X	X	Project monitoring health departments
	1995/96	B	205	2	X	X		X	X	X		X	X	Project monitoring health departments
	1996/97	B	407	6	X	X		X	X	X		X	X	Project monitoring health departments
	1998/99	B	798	16	X	X		X	X	X		X	X	Project monitoring health departments

6.6.4 Evaluation of the data on dioxins in human milk

In view of their evidence the data available on dioxins in human milk may be classified by 2 categories:

- The investigations of human milk for dioxins about which the Federal Laender reported were carried out predominantly in the framework of breast-feeding consultations. The level of the dioxin content characterizes the general background concentration in general occurring in Germany. Data on exposure factors are not available for these investigations.
- In a research project of the former BgVV the specific effects of a known contamination source on dioxin contents in human milk were repeatedly investigated in a locally limited area.

These two categories are separately evaluated.
Evaluation procedure:

1. The total dioxin contents of the 17 2,3,7,8-substituted congeners were calculated in a uniform way using the I-TEF (NATO/CCMS, 1988) and the WHO-TEF (WHO, 1997) by including concentrations below the detection limit with half of the value of the detection limit indicated.
2. The averages and the medians were weighted according to the numbers of samples used as basis as far as data from pool samples or aggregate data are available for the individual Federal Laender as well as for the whole of Germany.
3. The 95 percentile were determined without weighting. As partly aggregate values were included for the period 1991 – 1994 and the results of pool samples for 1996 – 1998 the data of the 95 percentile are to be regarded only as estimates.

In the following explanations the I-TEq are used – if not mentioned otherwise – to ensure the comparability with data reported for the other compartments.

6.6.4.1 Data for characterizing the background concentration

Representativeness of the data

The data transmitted by the Federal Laender have so far not been based on representative samples at random. The investigations were rather made by the Federal Laender upon request of interested mothers, e.g. in the framework of breast-feeding consultations.
The intake of dioxins is effected through food by 90 to 95 %. Due to the transregional supply with foodstuffs we can basically proceed from a comparable basic contamination of the various

regions. Thus, a summarizing evaluation of the data of the individual Federal Laender possibly serves the characterization of the situation in the Federal Republic of Germany. As the dioxin contents in milk fat, blood lipids and body fat are basically comparable the contamination of the total population of the respective age may be determined from the dioxin contents in human milk.

Data inventory and temporal trend of the background concentration of human milk

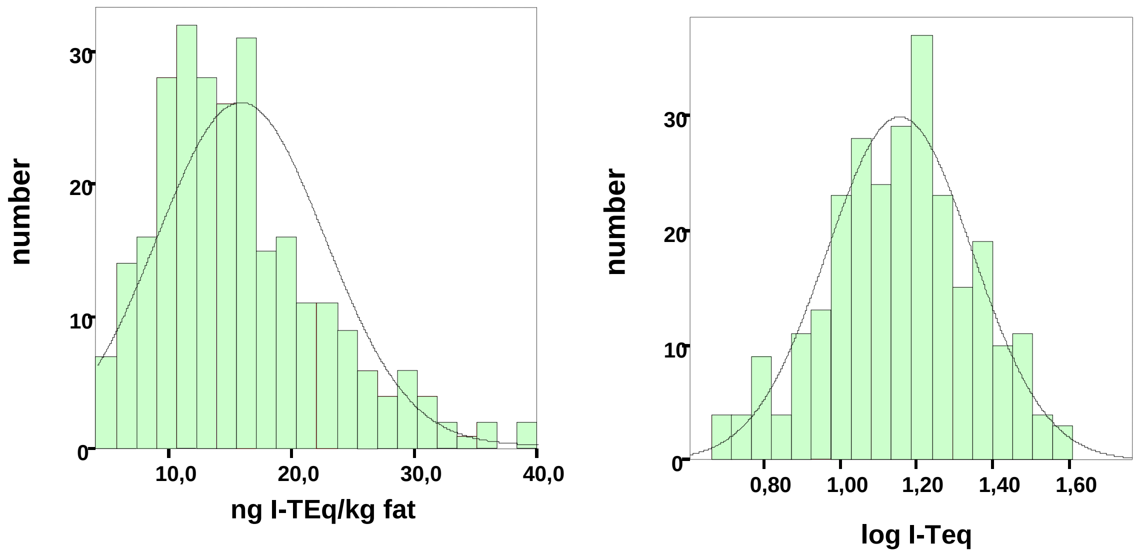
The data on which the characterization of the background concentration has been based so far are compiled in Tab. 19.

Table 19: Dioxin contents of the background concentration of human milk, classified according to Federal Land and sampling period, data in ng I-TEq/kg fat

Year	Land	n samples	n analyses	Minimum	Average	Median	Maximum
1992	BW	6	6	16.9	33.0		47.8
1993	BW	5	5	13.6	18.4	18.3	22.3
1994	BW	6	6	11.4	23.9	22.5	43.9
1995	BW	14	14	9.4	17.5	16,.	25.0
1996	BW	41	9	7.7	12.8	14.3	28.2
1997	BW	82	11	6.4	11.0	12.4	28.7
1998	BW	23	6	7.8	13.5	11.3	16.5
1991	NRW	111	111	6.4	23.3	22.4	51.1
1992	NRW	56	56	3.5	20.5	19.2	39.0
1993	NRW	78	78	5.3	20.9	21.1	37.6
1994	NRW	50	50	4.9	17.2	16.2	30.3
1995	NRW	38	38	6.0	16.1	15.7	30.3
1996	NRW	22	22	4.9	14.1	14.4	30.5
1997	NRW	11	11	9.7	11.9	10.5	16.9
1998	NRW	15	15	4.7	13.8	13.4	23.5
1991	NI	77	77	7.3	24.8	22.8	58.1
1992	NI	109	109	4.9	20.6	19.5	46.0
1993	NI	45	45	4.1	16.0	16.7	29.8
1994	NI	34	34	6.1	16.8	15.2	37.5
1995*	NI	83	83	5.4	18.7	16.5	39.0
1996	NI	18	18	5.1	16.6	16,6	30.1
1997	NI	15	15	6.0	12.7	12,5	23.5
1998	NI	25	25	5..3	12.3	11.1	28.9
1991	SH	3	3	19.2	32.9	23.7	55.7
1992	SH	4	4	19.3	22.9	23.0	26.5
1993	SH	3	3	7.9	18.5	19.1	28.4
1992	BY	2	2	16.2	16.7		17.1
1997	BY	18	3	9.7	12.8	10.6	16.9
1998	BY	6	1		10.5		
1993	BE/BgVV/WHO	10	1		16.6		

To give information on the trend the comparability of the test groups is of decisive importance as regards the influencing factors. Therefore information on this should be placed in front. Data on the influencing factors such as e.g. habits of nutrition, BMI (body measure index) or length of the breast-feeding period have so far not been collected. Data relating to age and number of breast-feeding periods are available for a part of the data sets (see Table 18). In the period 1995-1998 the annual averages of the age values are between 31.6 and 33.1 years (minimum = 24, maximum = 43), the averages for the number of breast-feeding periods vary between 1.3 and 1.6 in the same period. As regards these two influencing parameters the group composition of the individual years is nearly comparable.

Fig. 43 Frequency distribution of the dioxin contents in human milk (data of 1995-98, 271 measurements); normal distribution and log-normal distribution



The individual measured values available were, first of all, checked as to their frequency distribution. Due to the declining number of samples a presentation for separate years was not appropriate. Therefore the data for 1995 – 98 were summed up. The frequency distribution followed the expected log-normal distribution (Fig. 43). Due to the data transmitted partly in an aggregate form the frequency distributions for the period 1991-1994 were renounced.

The individual data from the Federal Laender were statistically evaluated. The descriptive parameters (minimum, maximum, average and median) are summed up in Table 19. The indication of the 95 percentile on Federal Land level was renounced as frequently the test groups were too small for that or pool samples were investigated.

Taking the variabilities of the analytical methods into account the dioxin contents of the human milk samples from various Federal Laender are on a similar level in the course of a sampling year, confirming thus the transregionally characterized background concentration. In the participating Federal Laender a synchronous, distinctly falling trend for averages and medians is to be detected in the period under investigation 1991-1997 (exception 1998, see Chapter 6.6.6) (Fig. 44).

Fig 44: Temporal trend (1991-98) of the average dioxin contents in human milk from the three Federal Laender BW, NI and NRW and of the data summed up nationwide

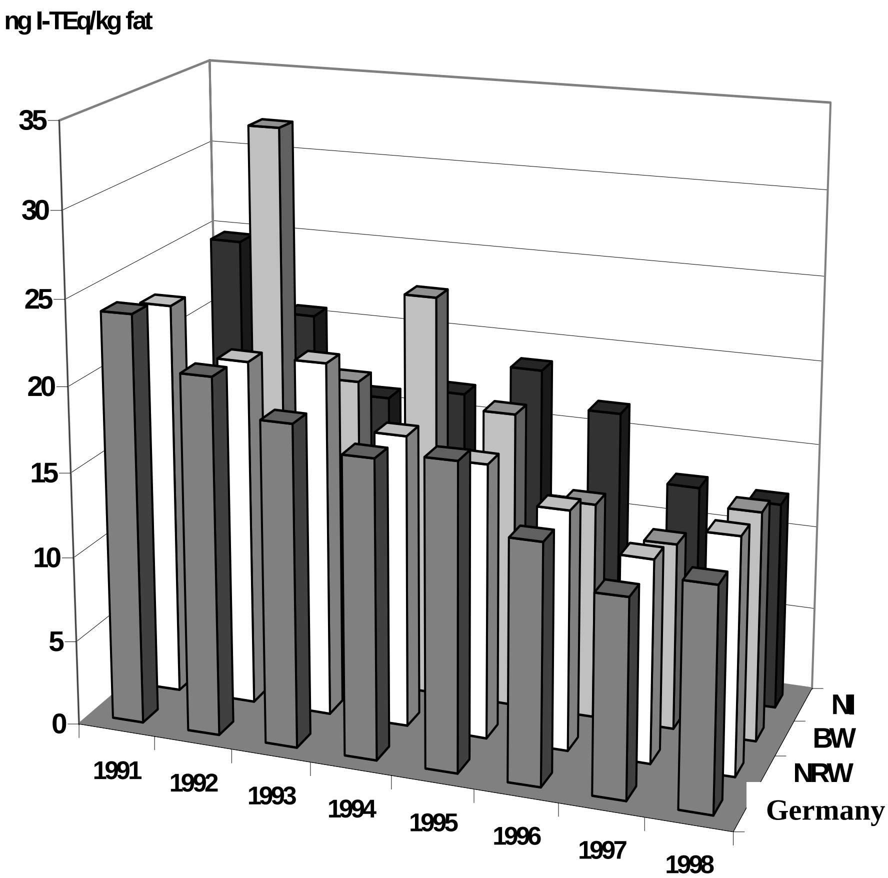
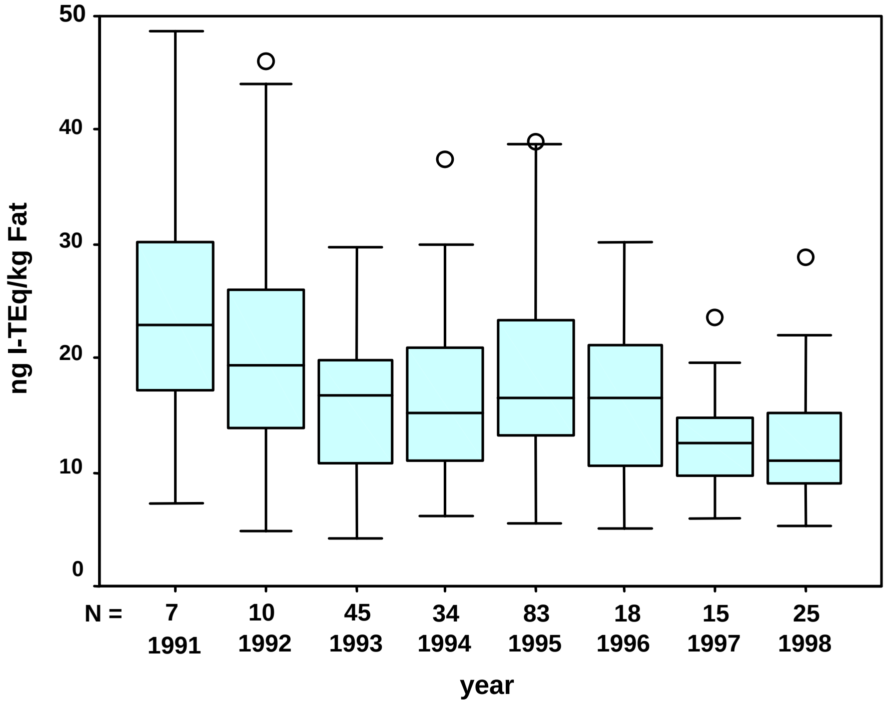


Fig. 45: Box plot on the temporal trend of the dioxin contents in human milk from Lower Saxony (0=runaway)



The longest time series are available for North Rhine-Westphalia, Lower Saxony and Baden-Wuerttemberg. As already since 1991 individual data have been available the described trend for the data from Lower Saxony is represented as box plot by means of this example. (The box plot characterizes the interquartile distance, i.e. the value range of the 25 to 75 percentiles, the median and the spreading range of the values which are no runaways; 0 = runaway).

Hereby there is obvious that the spreading ranges of the data are reduced (Fig. 45). This is due to the declining peak impacts, where, on a limited scale, the declining number of the investigated samples and, in addition to the nationwide data, the investigation of mixed samples from Bavaria, Baden-Wuerttemberg and Berlin have to be considered (see also Figs. 46 and 47).

Fig. 46: Box plots on the dioxin contents in human milk from Baden-Wuerttemberg (BW), Lower Saxony (NI) and North Rhine-Westphalia (NRW) for the period 1995-98 (o = runaway)

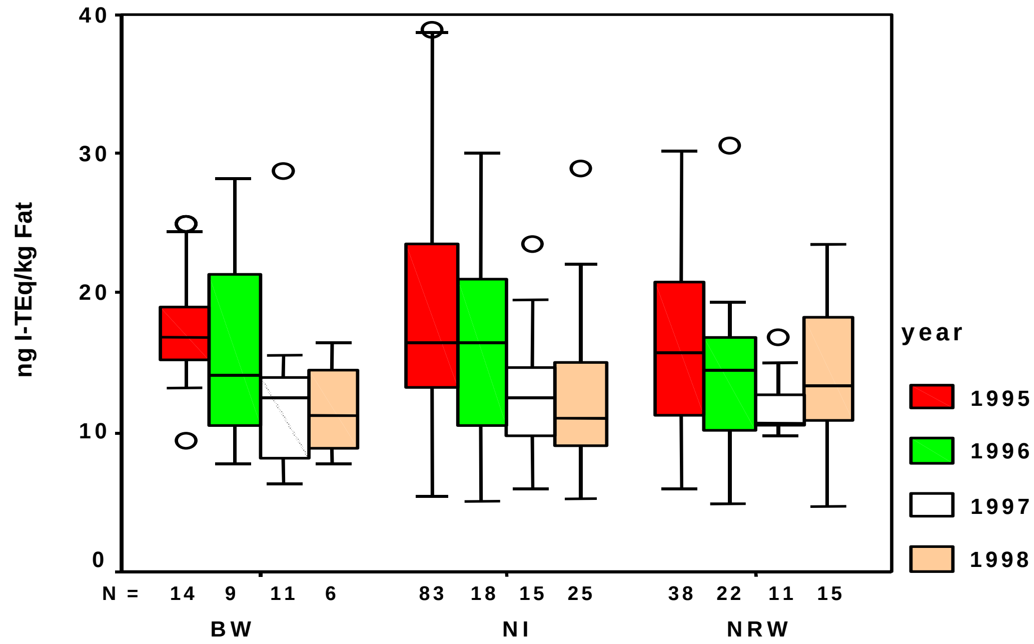
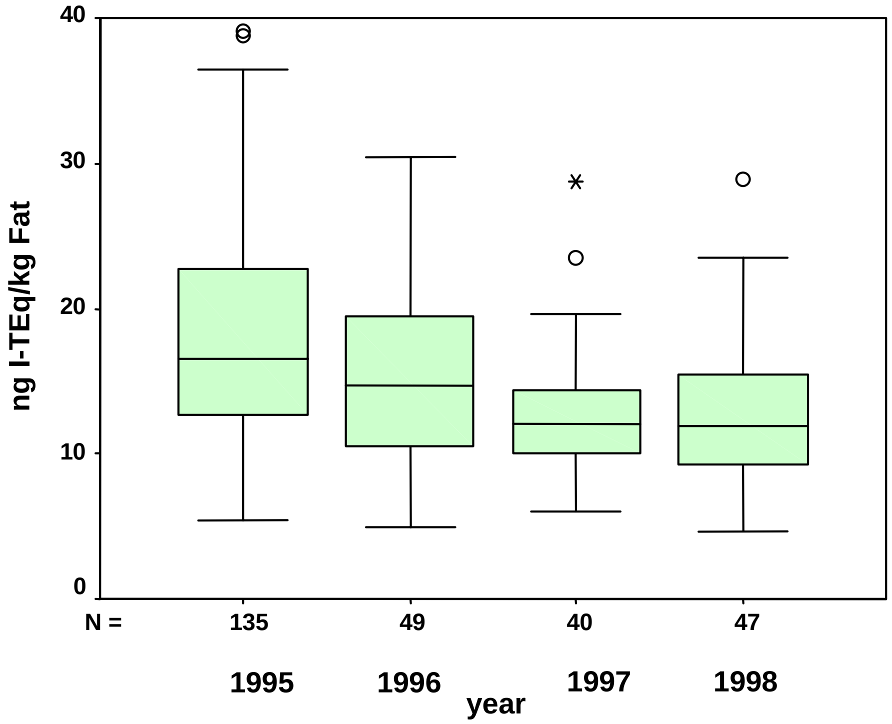


Fig. 47: Box plot on the dioxin contents in human milk ; nationwide trend 1995-98 (0 = runaway; * = extreme value)



These declining dioxin contents to be observed in the Federal Laender are also reflected in the data summed up nationwide. The descriptive statistical parameters minimum, average, median and maximum are summed up by years in Tab. 20. They were extended by the 95 percentile. To complete the temporal trend published data from Lower Saxony, North Rhine-Westphalia and from the former BgVV were supplemented for the period 1986-90 (1). The average and maximum dioxin contents calculated by means of the WHO-TEF were included for comparison. The WHO-TEq are numerically by 16 % higher, on average, than the data based on the I-TEF.

Table 20: Average PCDD/PCDF contents in human milk (calculated from the data collected in the Federal Laender; averages and medians weighted)

Year	n	ng I-TEq/kg fat					ng WHO-TEq/kg fat	
		Minimum	**Average**	**Median**	**95 perc.**	**Maximum**	**Average**	**Maximum**
1986-90	728	5.6	30.6	29.2		87.1	35,7	
1991	191	6.4	24.1	23.4	48.4	58.1	27.8	64.9
1992	171	3.5	21.0	20.6	39.2	47.8	24.0	53.8
1993	141	4.1	18.9	20.9		37.6	22.0	
1994	90	4.9	17.5	17.2	36.7	43.9	20.4	50.3
1995	135	5.4	17.9	16.5	32.3	39.0	20.9	45.8
1996	81	4.9	14.0	13.7	29.6	30.5	16.2	35.4
1997	126	6.0	11.6	12.4	23.3	28.7	13.5	32.8
1998	69	4.7	12.9	12.0	23.0	28.9	15.0	31.9

The average dioxin contents of approx. 30 ng I-TEq/kg fat in human milk from Germany observed in the second half of the 80-ies are comparable to those from other West European countries. (2). Since the early 90-ies a continuously falling trend of the average dioxin contamination is to be stated. Until 1997 the average content went down by about 60 %. It is remarkable that in 1998 the average dioxin contents are by approx. 10 % higher as compared with 1997. This is, in particular, due to data from Baden-Wuerttemberg (values of 1998, on average, by 22 % higher than in 1997) und from North Rhine-Westphalia (values of 1998, on average, by 16 % higher than in 1997) whereas in Lower Saxony and Bavaria insignificantly declining average dioxin contents have been detected also in this period (see Tab. 19). It remains to be seen in the years to follow how this trend will develop.

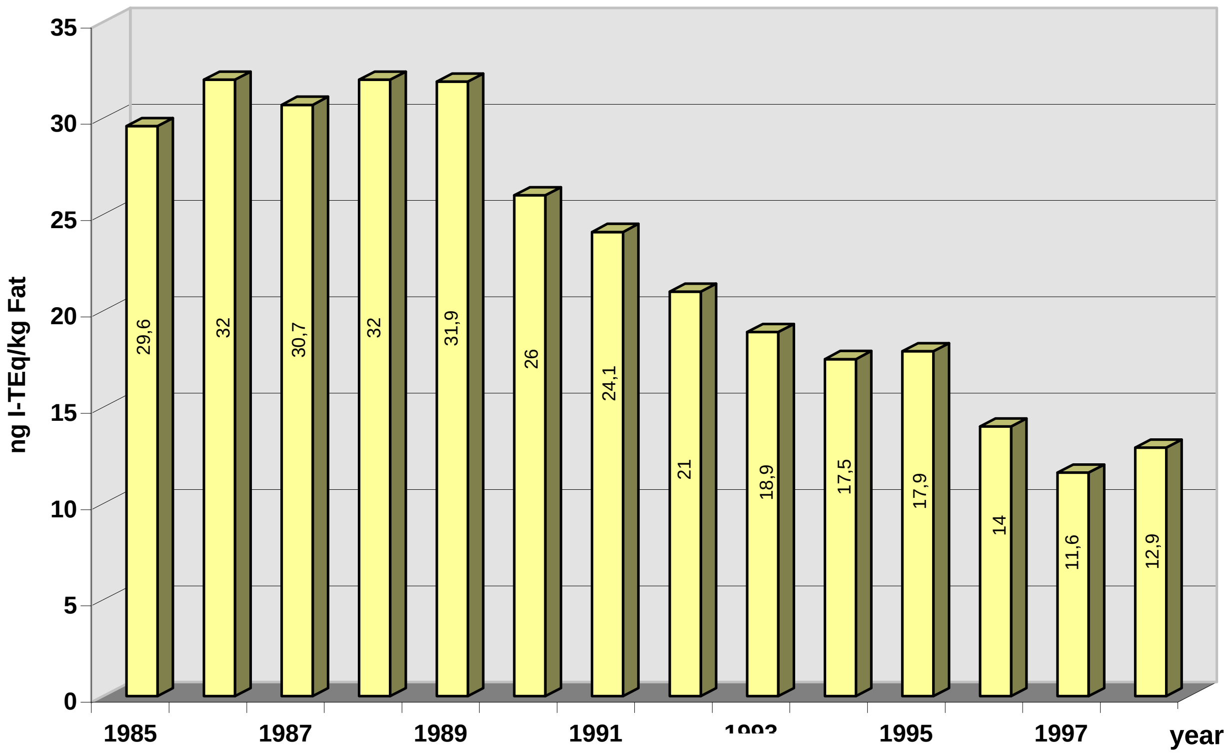
Considered by themselves the results from Baden-Wuerttemberg may be regarded as variations caused by accident which may possibly result from variabilities in the composition of the comparatively small test groups as regards the numerous factors influencing the dioxin content. However, as also in blood samples of children from Baden-Wuerttemberg (increase by approx. 20-40 %) and in blood samples of adults a parallel increase of the I-TEq values in time is to be observed this might be also due to a potential connection with a specific, additional exposure. Potential references will be discussed in Chapter 6.6.6.

It is remarkable that also in the case of high impacts a continuously declining trend is to be observed to a comparable extent (approx. 60 %), yet the evidential value is not documented due to the declining numbers of samples. Minimum and maximum values are within a decimal power. The maxima are, in general, dependent on the selection of samples caused by accident and the volume of samples.

However, the 95 percentile should be less affectable by the selection and volume of samples than the maxima. It was already referred to restrictions due to the data available on human milk. Also as to the 95 percentiles the decline of the values of a similar magnitude is clearly shown as in the case of averages and maxima. The values detected in human milk in 1997/1998 for the 95 percentile and also for the maximum are lower than the average dioxin contamination in the late 80-ies.

The temporal trend of the average dioxin contents in human milk in the years 1985 to 1997 is presented in Fig. 48.

Fig. 48: Temporal trend of the average dioxin contents (weighted averages) in human milk from the Federal Republic of Germany (data of 1991-1998 from Tab. 20; 1985 – 1990 from literature)



Dioxin contents in human milk exceeding today the range of 25-30 ng I-TEq/kg fat may be considered as an indication to an increased exposure of the mother exceeding the present background concentration where, however, also in elder mothers breast-feeding their first baby higher dioxin contents are to be expected.

The clear decline of the background concentration by dioxins in human milk samples from Germany by about 60 % observed in the last 10 years is confirmed by a similar reduction of the dioxin contents in blood samples (3,4). This is in the same order of magnitude as the decline of the dioxin contents in foodstuffs of animal origin observed in the same period and of the dioxin quantities daily taken in with food (1986-90: 1.9 pg I-TEq/kg of body weight and day; 1996-98: 0.7 pg I-TEq/kg of body weight and day) (5).

Influencing factors „age“ and „number of breast-feeding periods“

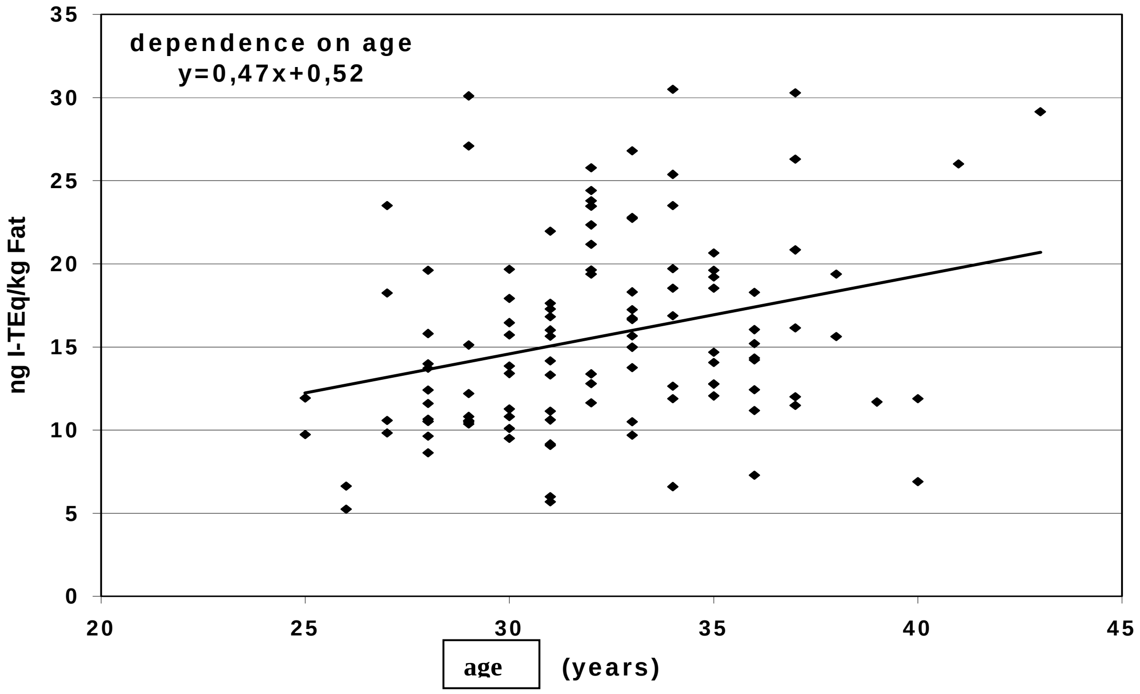
Assuming a normal diet the dioxin contents detected in human milk are affected, among others, by the factors age and number and length of the breast-feeding periods. Through food humans enrich their fatty tissue by dioxins which results in an increase of the body burden with increasing age. However, by breast-feeding dioxins are removed through fat-containing human milk from the body of the mother resulting in a decline of her body burden.

The data on age and number of breast-feeding periods transmitted by the Federal Laender since 1995 for the majority of the investigations allow an actual evaluation of these parameters.

To determine a possible dependence on age data from the years 1995 – 1998 were evaluated. Human milk from mothers at the age between 25 and 43 years who breast-fed their first baby were analyzed. In spite of the wide spreading of the data (s. Fig. 49) a correlation between PCDD/F contents in human milk and increasing age of the mothers is to be detected. Applying a linear regression an average increase of 0.47 ng I-TEq/kg fat per year may be estimated. The connection between dioxin concentration and increasing age is more pronounced in studies where blood and fatty tissue were analyzed (6).

To estimate the effects which preceding breast-feeding periods have on the dioxin contents detected the data of 1995 – 1998 as aggregate data were taken as basis. A differentiation according to sampling years was not possible as the test groups were too small for that.

Fig. **49:** Dependence of the dioxin contents in human milk on age (Data of 1995-98; 1st breast-feeding period; N=116)

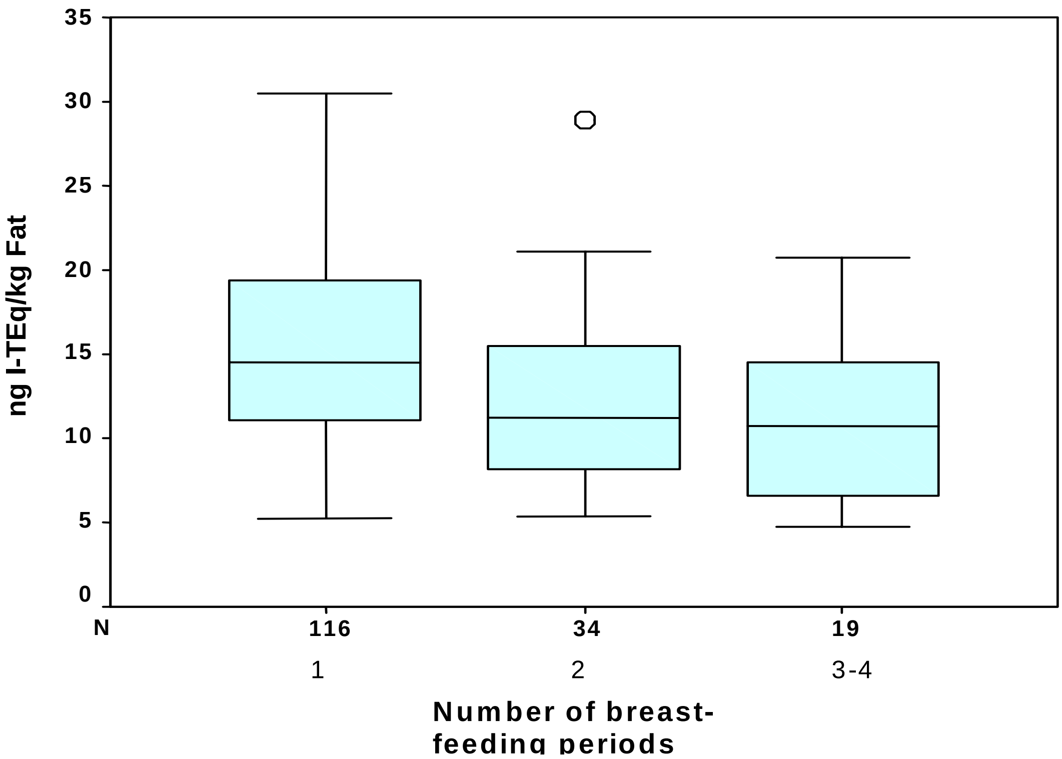


Data on the length of the individual breast-feeding periods are not available. To estimate the amount by which the body load of the mother was reduced by breast-feeding the more inaccurate parameter – number of breast-feeding periods – was therefore taken into account. The data are summed up in Tab. 21 and in Fig. 50.

Table 21: Dioxin contents in human milk (ng I-TEq/kg fat), differentiated according to the number of breast-feeding periods; data of 1995-1998 (*due to the small number of samples only orientation values of the 95 percentile)

Number of breast-feeding periods	1	2	3-4
Number of samples	116	34	19
Age	32.2 (25-43)	31.9 (24-40)	33.6 (28-40)
I-TEq [ng/kg fat]			
Average	15.6	12.3	10.9
Median	14.5	11.3	10.7
95 percentile	26.8	23.0*	20.7*
Maximum	30.5	28.9	20.7

Fig. 50: Dependence of the dioxin content in human milk on the number of breast-feeding periods; data of 1995/1998 (o = runaway)



The composition of age in groups with 1 and 2 breast-feeding periods may be regarded to be nearly equal. The average age of women breast-feeding their 3rd or 4th baby is, by 1.7 years higher as compared with women with two children results in a dioxin concentration higher by about 0.8 ng I-TEq/kg fat determined by age.

With the increasing number of breast-feeding periods the average dioxin content goes down. The same trends for the 95 percentile and the maxima are only relatively revealing due to the partly very small group sizes. In the case of an age-standardized consideration there may be estimated that the individual breast-feeding periods contribute, on average, with about 2.3 –3.3 ng I-TEq/kg fat to reducing the body burden of the mother. The dioxin contents in milk samples of women breast-feeding their 2nd baby are, on average, by about 20 % lower and of women breast-feeding their 3rd or 4th babies by about 30 – 40 % lower as compared with milk samples of mothers breast-feeding their first baby. These actual data confirm the results of earlier investigations (7,8).

Congener-specific data analysis

In Tab. 22 the contents of the 2,3,7-8-substituted congeners (average, minimum, maximum) and their contributions to the dioxin equivalents in per cent are summed up in an exemplary way for the years of investigation 1986-1990, 1994 and 1998.

Table 22: Contents of 2,3,7,8-substituted congeners in human milk

	1986-90 (N=728)		1994 (N=90)		1998 (N=69)		
	ng /kg fat MW (Minimum-Maximum)	% I-TEq	ng /kg fat MW (Minimum-Maximum	% I-TEq	ng /kg fat MW (Minimum-Maximum)	% I-TEq	% WHO TEq
2,3,7,8-TCDF	1.8 (0.15-12.0)	0.6	0.5 (0.1-1.4)	0.3	0.4 (0.1-1.5)	0.3	0.3
2,3,7,8-TCDD	3.2 (0.5-10.0)	10.0	1.9 (0.1-4,2)	10.9	1.5 (0.4-3.9)	11.5	9.9
1,2,3,7,8-PeCDF	0.6 (0.1-21.5)	0.1	0.2 (0.1-0.7)	0.1	0.2 (0.04-0.6)	0.1	0.1
2,3,4,7,8-PeCDF	28.3 (0.4-10.4)	46.0	16.4 (5.97-45.6)	46.7	12.1 (3.2-38.7)	47.0	40.5
1,2,3,7,8-PeCDD	10.4 (1.7- 35.0)	17.0	6.1 (0.1-14.6)	17.4	4.3 (1.4-7.8)	16.7	28.8
1,2,3,4,7,8-HxCDF	7.7 (1.1-28.0)		5.0 (1.6-15.4)	2.9	3.2 (1.2-7.7)	2.5	2.2
1,2,3,6,7,8-HxCDF	7.3 (1.0-24.0)		4.2 (1.6-12.5)	2.4	3.0 (1.2-8.0)	2.3	2.0
2,3,4,6,7,8-HxCDF	3.5 (0.1-16.0)		1.7 (0.2-4.3)	1.0	1.2 (0.2-2.6)	0.9	0.8
S(2,3,7,8)-HxCDF	18.5 (3.1-55.0)	6.0		6.3		5.8	5.0
1,2,3,4,7,8-HxCDD	8.5 (1.2-33.0)		4.0 (0.3-9.3)	2.3	3.2 (0.6-6.1)	2.5	2.1
1,2,3,6,7,8-HxCDD	37.9 (3.0-126)		20.9 (5.5-45.1)	12.0	15.2 (5.1-29.2)	11.8	10.2
1,2,3,7,8,9-HxCDD	7.0 (0.4-21.0)		3.1 (0.8-10.2)	1.8	2.5 (0.6-4.3)	1.9	1.7
S(2,3,7,8)-HxCDD	53,.4 (3.2-178)	17.0		16.0		16.2	14.0
1,2,3,4,6,7,8-HpCDF	7.3 (0.5-106)	0.2	3.6 (1.2-19.0)	0.2	2.2 (0.8-10.8)	0.2	0.2
1,2,3,4,6,7,8-HpCDD	47.2 (6.4-161)	1.5	25.9 (5.6-111.8)	1.5	19.0 (4.4-36.8)	1.5	1.3
OCDF	2.2 (0.1-232)	0.0	0.6 (0.1-5.5)	0.0	0.3 (0.02-0.7)	0.0	0.0
OCDD	226 (19.0-1300)	0.7	112.9 (32.2-444.8)	0.7	100.6 (28.2-224.0)	0.8	0.1
I-Teq	30.5 (5.6-87.1)		17.5 (4.9-43.9)		12.9 (4.7-28.9)		
PCDF		53.0		53.6		54.8	46.0
PCDD		47.0		46.4		45.3	54.0

As in the I-TEq values also in these 17 congeners the average and maximum contents are declining. The contribution of the individual congeners to the overall dioxin content (I-TEq values) in % is, however, equal with regard to the analytical spreading in the period under consideration. With approx. 45 % the 2,3,4,7,8 PeCDF contributes most strongly to the overall dioxin content (I-TEq value). Bigger shares result by 17 % from the 1,2,3,7,8 PeCDD and the group of hexadioxins respectively and by approx. 10 % from TCDD. The share of furans on the basis of the international toxicity equivalents dominates with approx. 55 % as against the share of dioxins with approx. 45 %.

Taking the WHO-TEF as basis for the calculation of the overall dioxin content and the contributions of the individual congeners in per cent the share of the individual congeners is shifted. The contribution of the 1,2,3,7,8-PeCDD goes up to approx. 29 % whereas the share of the 2,3,4,7,8-PeCDF declines to approx. 40 %. This affects the dioxin-furan ratio. In general, now dioxins dominate with approx. 55 %. The respective data calculated by way of example for 1998 may be taken from Tab. 22.

Dioxin exposure of the infant

In conformity with the declining dioxin contents in human milk also the daily dioxin intake of an infant connected with breast-feeding declined. For a 4 month-old infant exclusively breast-fed with mother's milk the average dioxin intake is calculated with 57 pg I-TEq/kg of body weight and day on the basis of the average dioxin content in human milk determined in 1998, an average quantity of milk intake of 821 ml (9), an average body weight of 6.5 kg (10) and an assumed fat content of 3.5 % in milk. As compared with adults (1996-98: 0.7 pg I-TEq/kg of body weight and day) the daily intake by dioxin of the breast-fed infant related to the body weight is by nearly 2 orders of magnitude higher. The TDI of 1 - 4 pg WHO-TEq/kg of body weight and day derived in 1998 by the WHO for the intake of dioxins and PCB similar to dioxin applies to a lifelong intake and does thus not provide a basis for assessing the dioxin intake during the short time of breast-feeding (11). As, on the other hand, breast-feeding has been proven to have positive effects on infant development the WHO and the National Commission for Breast-feeding recommend breast-feeding in spite of the increased dioxin impact.

Yet, the WHO and other expert commissions require further measures to be taken to control and minimize the dioxin input into the environment (11,12).

6.6.4.2 Dioxin contents in human milk from a contaminated area

Investigations of milk of women living in an industrially contaminated area for dioxins were carried out by the former BgVV in the years 1990/91. A second series of measurement followed in 1997. A copper recycling mill with a cable carbonizing plant resulted in high dioxin emissions and depositions in this region over many years. This plant was shut down approx. 10 years ago.

Table 23: PCDD/PCDF contents in human milk from an industrially contaminated area (* mother has been living in this region for one year, coming from the old Federal Laender ((16,17)

Year	n	ng I-TEq/kg fat		
		Minimum	**Average**	**Maximum**
1990/91	9	36	59	86
1997	10	11*/19	41	81

In Tab. 23 averages, minima and maxima of the dioxin contents in human milk samples of both series of investigation are summed up. The first investigations carried through in 1990/91 confirmed the assumption that dioxin emission over many years resulted in remarkably high dioxin contents in human milk samples from this region.

Though the values determined in 1990/91 are in the range of the dioxin contents observed in the old Federal Laender before 1990 all 9 samples are, however, contaminated above average. The average calculated for this region is higher by a factor of 2 than in the old Federal Laender. In the distribution pattern of the congeners the clearly higher shares of HxCDF, 2,3,4,7,8-PeCDF, yet also of 1,2,3,7,8-PeCDD and TCDD, attract attention which points to a specific contamination source. The main contamination pathway for the inhabitants was, in particular, the regional food supply preferred at that time and the traditonally higher share of self-supply with such foodstuffs as e.g. eggs, chicken meat, fruit and vegetable. Due to dioxin emission over many years they were clearly contaminated. Appropriate recommendations for consumption were given to the population of this region.

The follow-up investigation in 1997 showed dioxin contents of milk above average for all samples from mothers living in this region for a longer time in spite of the emission sources having been shut down, the recommendation to change the habits of nutrition and an increasing transregional food supply. The lowest value from the series of measurement of 1997 comes from a mother living there only for 1 year and having moved there from the old Federal Laender.

An analysis of the exposure factors documented in the questionnaire shows a positive correlation with the traditional habits of nutrition (self-supplier) as well as with the duration of living in the vicinity of emission sources. The highest dioxin contents were detected in women eating food from the own production and having lived for a long time (partly lifelong) in the vicinity of emission sources (< 4 km).

6.6.5 Evaluation of data on dioxins in the blood of 9– to 11-year old children from Baden-Wuerttemberg

6.6.5.1 Collection and composition of samples

The investigations in the framework of the project „monitoring health departments in Baden-Wuerttemberg“ are carried out as repeated interdisciplinary investigations of pupils in the 4th class in fixed study areas in the competence of these monitoring health departments. The basis for selecting the monitoring health departments were a sufficient quantity of data covering the various environmental compartments (e.g. air) and aspects of the spatial differentiation (categories of regional planning according to the Land development plan). The four monitoring health departments Mannheim, Stuttgart (as of 1995/96), Ortenaukreis and Ravensburg chosen cover largely the range of the contaminations met in Baden-Wuerttemberg and the spatial structures. Mannheim and Stuttgart represent the biggest urban areas of Baden-Wuerttemberg marked by a high industrial and transport density. The Ortenaukreis (investigation area Kehl) is an urban area in the rural area showing an area of a high industrial density in the region of Kehl. The district of the Ravensburg health department (study area Aulendorf/Bad Waldsee) characterizes the rural area without industrial agglomerations.

The test group consists of 9- to 11-year old children. The collecting blood samples consist of the individual samples of children living in the respective place at least 2 years and born in Germany. The rate of participation of the children in the investigations between 1993/1994 and 1998/1999 was altogether 78.3 %.

The collecting blood samples were compiled according to various criteria in the individual years of investigation. Whereas in 1993/1994 differentiation criteria were not considered since 1994/1995 a differentiation has been partly made according to the sex. In 1998/1999 samples differentiated according to the status of breast-feeding were additionally compiled. Tab. 24 gives a survey of the composition of the samples.

Table 24: Characaterization of the collecting blood samples of 9 - to 11-year old children from Baden-Wuerttemberg

	Year of investigation	Place of investigation	Specification of the sample	Number of individual samples in the collecting sample
1	1993/94	Mannheim	Boys and girls	79
2	1993/94	Kehl	Boys and girls	44
3	1993/94	Aulendorf/Bad Waldsee	Boys and girls	33
4	1994/95	Mannheim	Boys	125
5	1994/95	Mannheim	Girls	120
6	1994/95	Kehl	Boys	83
7	1994/95	Aulendorf/Bad Waldsee	Boys	69
8	1994/95	Aulendorf/Bad Waldsee	Girls	55
9	1995/96	Stuttgart	Girls	86
10	1995/96	Stuttgart	Boys	97
11	1996/97	Aulendorf/Bad Waldsee	Girls	54
12	1996/97	Aulendorf/Bad Waldsee	Boys	48
13	1996/97	Stuttgart	Girls	53
14	1996/97	Stuttgart	Boys	50
15	1996/97	Kehl	Boys and girls	105
16	1996/97	Mannheim	Boys and girls	97
17	1998/99	Kehl	Girls	39
18	1998/99	Kehl	Boys	35
19	1998/99	Kehl	Breast-fed children	48
20	1998/99	Kehl	Children not breast-fed	18
21	1998/99	Mannheim	Girls	58
22	1998/99	Mannheim	Boys	75
23	1998/99	Mannheim	Breast-fed children	89
24	1998/99	Mannheim	Children not breast-fed	41
25	1998/99	Aulendorf/Bad Waldsee	Girls	54
26	1998/99	Aulendorf/Bad Waldsee	Boys	51
27	1998/99	Aulendorf/Bad Waldsee	Breast-fed children	88
28	1998/99	Aulendorf/Bad Waldsee	Children not breast-fed	16
29	1998/99	Stuttgart	Girls	46
30	1998/99	Stuttgart	Boys	46
31	1998/99	Stuttgart	Breast-fed children	66
32	1998/99	Stuttgart	Children not breast-fed	28

6.6.5.2 Consideration of various influencing factors

According to the agreement in the working sub-group on the Reference Measuring Program the basis of the following evaluations are the I-TEq values, with values below the detection limit being included with half of the value of the detection limit. In all results there should be taken into account that an individual runaway may have strong effects on the concentration detected in a collecting blood sample.

Important influencing factors on the dioxin contents of blood samples of children of the investigated age group will be discussed hereinafter.

Influence of the sex

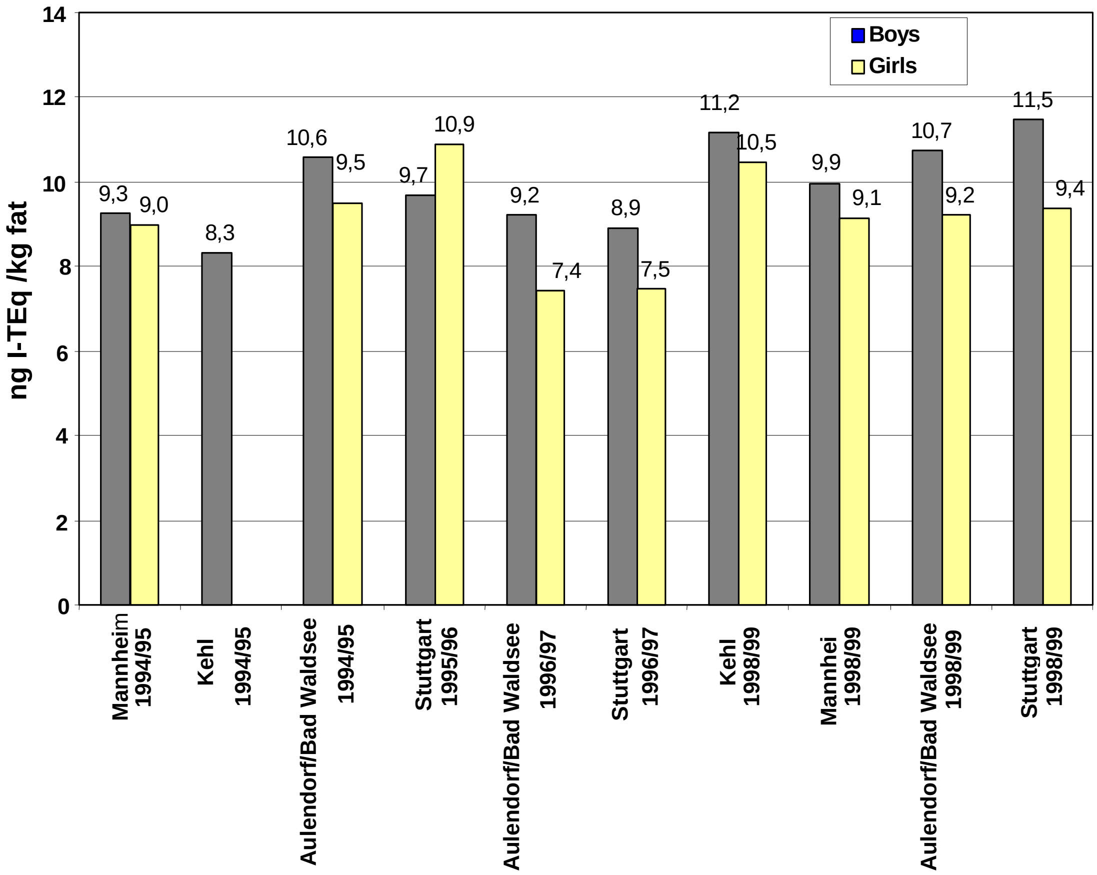
The dioxin contents of the collecting blood samples differentiated according to sex are represented in Tab. 25. Here there becomes evident that – with the exception of Stuttgart 1995/1996 – the blood of boys shows higher values than the blood of girls. The difference totals, on average, 11 %.

Table 25: Comparison of the dioxin contents (I-TEq) in collecting blood samples of 9– to 11-year old boys and girls

Year	Place of investigation	Boys ng I-TEq /kg fat	Girls ng I-TEq/kg fat	Increase for boys by %
1994/95	Urban area 1 (Mannheim)	9.3	9.0	2.8%
1994/95	Urban area in the rural area (Kehl)	8.3		
1994/95	Rural area (Aulendorf/Bad Waldsee)	10.6	9.5	11.2%
1995/96	Urban area 2 (Stuttgart)	9.7	10.9	-11.2%
1996/97	Rural area (Aulendorf/Bad Waldsee)	9.2	7.4	24.3%
1996/97	Urban area 2 (Stuttgart)	8.9	7.5	18.9%
1998/99	Urban area in the rural area (Kehl)	11.2	10.5	6.9%
1998/99	Urban area 1 (Mannheim)	9.9	9.1	8.7%
1998/99	Rural area (Aulendorf/Bad Waldsee)	10.7	9.2	16.5%
1998/99	Urban area 2 (Stuttgart)	11.5	9.4	22.4%
		Average		11.2%

The distribution of the breast-feeding status is, as a rule, balanced for boys and girls, however differing in the share depending on the place and year of investigation up to 15 %. A varying ratio as to the breast-feeding status or breast-feeding period of the various groups might also explain the big spreading of the values (spreading area: 2.8 % - 24.3 %). Nevertheless the qualitative data (in 8 of 9 data pairs the blood of boys shows higher contents) is documented (Fig. 51)

Fig. 51: Influence of the sex on the dioxin contents (ng I-TEq/kg fat) in collecting blood samples of 9– to 11-year old children from Baden-Wuerttemberg, differentiated according to location and sampling period

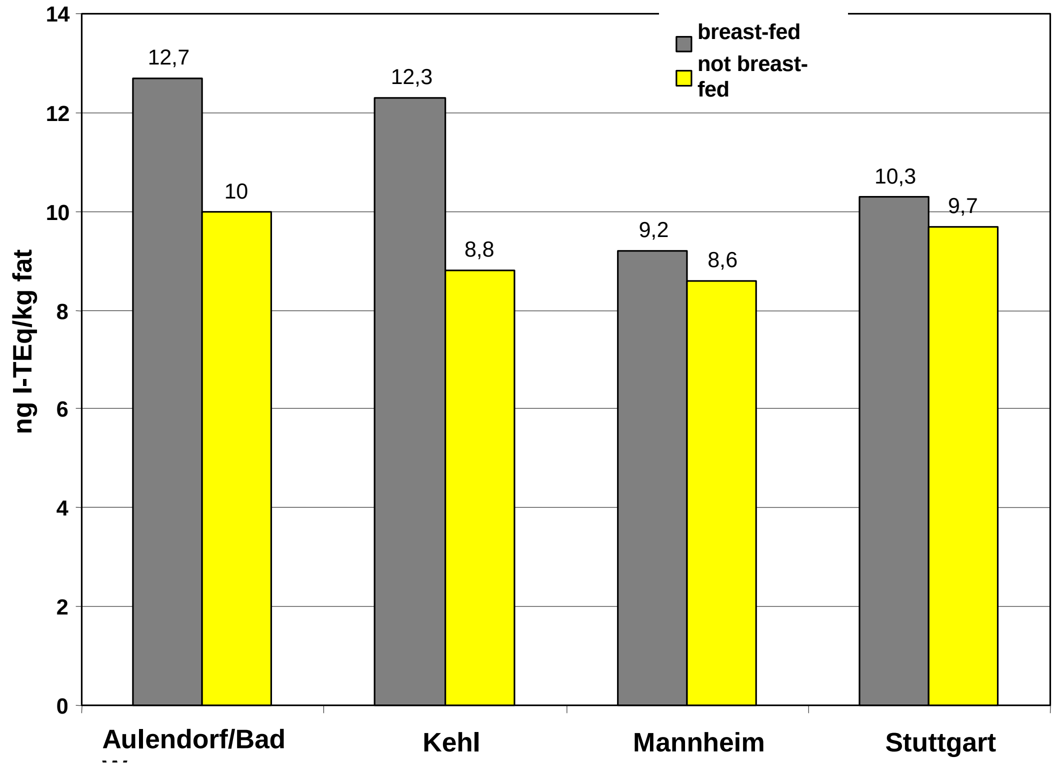


An influencing factor causing sex-specific differences are the different habits of nutrition of boys and girls. In accordance with the VERA study (joint study on investigation of nutrition, risk factors, analyzing) the daily intake of boys at the age of 10-12 years is, on average, 103.8 g fat, whereas girls of the same age eat only 92.3 fat a day (difference of 10 %). In addition, girls have a higher body fat share in per cent than boys which referring to the dioxin contents detected results in a „dilution effect“. Analoguous differences are to be detected also in actual investigations of PCB contents in the blood of adults. On average, women have lower PCB contents in their blood than men, with the effect of breast-feeding having to be considered for women.

Effect of breast-feeding

A differentiation according to the breast-feeding status was made in the blood samples of 1998/1999. An additional dioxin intake in the infant age is connected with breast-feeding. It is still well detectable in the age group of 9 to 11 years investigated here. The group of breast-fed children shows, on average, by 20 % higher dioxin contents in the collecting blood samples (Tab. 26, Fig. 52). Obviously the breast-feeding status has a stronger influence than the sex in this age group.

Fig. 52: Influence of breast-feeding on the dioxin contents (ng I-TEq/kg fat) in collecting blood samples of 9- to 11-year old children; data of 1998/1999



The distribution of the sex in the collecting blood samples according to the breast-feeeding status is balanced in Kehl and Aulendorf/Bad Waldsee. In Mannheim, however, the share of girls is clearly higher in breast-fed as well as in not breast-fed children. In Stuttgart the share of girls is also higher in breast-fed children, in not breast-fed children, however, the share of girls is clearly smaller.

Table 26: Effects of breast-feeding on the dioxin contents in blood (ng TEq/kg fat) of 9- to 11-year old children from Baden-Wuerttemberg; data of 1998/99

Effects of breast-feeding	Rural area (Aulendorf/ Bad Waldsee)	Urban area in the rural area (Kehl)	Urban area 1 (Mannheim)	Urban area 2 (Stuttgart)	Average
breast-fed	12.7	12.3	9.2	10.3	11.1
not breast-fed	10.0	8.8	8.6	9.7	9.3
additional contribution by breast-feeding	27.0 %	39.8 %	7.0 %	6.2 %	19.9 %

By this unequal distribution of sex in the collecting blood samples of children from Stuttgart the effect of breast-feeding on the dioxin contents in the urban area 2 is rather underestimated. Considering the distribution of the sex (provided the breast-feeding period is equal or a similar time pattern as to additional feeding is applied) there may be assumed that the effect of breast-feeding is rather of an order of magnitude determined for Kehl and Aulendorf/Bad Waldsee.

There is to be expected that the dioxin intake of children is affected by the duration of the breast-feeding period in addition to the sex. Data of 1996/1997 document that mothers from Mannheim stop breast-feeding a little earlier than mothers from other places of investigation. The lower dioxin contents in the collecting blood samples of the breast-fed children from Mannheim could thus be due to a shorter breast-feeding of the children.

6.6.5.3 Temporal Trends

Influencing factors and comparability of the test groups

A prerequisite to the evaluation of the temporal trends is the comparability of test groups with regard to important influencing factors such as place of investigation, sex and breast-feeding status. Short remarks on the comparability of these factors are described below.

Place of investigation

The explanations in the preceding chapters point to the differences between the place of investigation for which, proceeding from the available data, a final clarification whether they are exclusively due to different effects of the breast-feeding status or differences in the breast-feeding attitude is not possible. To exclude this factor only comparisons in the individual places of investigation are made to determine the trend.

Sex:

In the framework of investigations, inter alia, collecting blood samples were taken where the blood of girls and boys was mixed. The ratio between the sexes was balanced in these samples. For the years of investigation with collecting blood samples differentiated according to sex available the dioxin contents were calculated as averages from the contents of the sample pools for boys and the sample pools for girls (not weighted, to represent a 1:1 ratio) per place and period of investigation.

Breast-feeding status:

In Aulendorf/Bad Waldsee and Stuttgart, as a rule, the observed breast-feeding rate is 10 % higher than in other places of investigation. The share of the breast-fed children remains nearly unchanged over the period in the four places of investigation. However, these data refer to the whole group and not to the partial group from which the collecting blood samples were composed. Yet, a transferability of the data to the partial group is assumed. Therefore a temporal consideration of breast-feeding seems to be not necessary.

Trend assessment

Due to the aspects mentioned above the comments on the trend assessment should be treated with caution. Therefore the comments should be rather understood as information. All data sets of mixed collecting blood samples of girls and boys and of the sex-specific collecting blood samples were included in the trend assessments.

The temporal trend of the dioxin concentrations in blood is represented in Fig. 53. Here, a slightly declining trend is shown until 1996/1997 and subsequently an increase in the year of investigation 1998/1999. This trend is to be observed in all 4 locations of investigation (Tab. 29) and is proved by separate investigations of the trend of dioxin contents in collecting blood samples of boys and girls (Tabs. 27 and 28).

Table 27: PCDD/PCDF concentrations in collecting blood samples of boys, differentiated by the place and year of investigation (ng I-TEq /kg a fat)

Boys	1994/95	1995/96	1996/97	1998/99	Change 1996/97 till 1998/99
Urban area1 (Mannheim)	9.3			9.9	
Urban area 2 (Stuttgart)		9.7	8.9	11.5	29.2 %
Urban area in the rural area (Kehl)	8.3			11.2	
Rural area (Aulendorf/Bad Waldsee)	10.6		9.2	10.7	16.3 %

Table 28: PCDD/PCDF concentrations in collecting blood samples of girls, differentiated by place and year of investigation (ng I-TEq /kg fat)

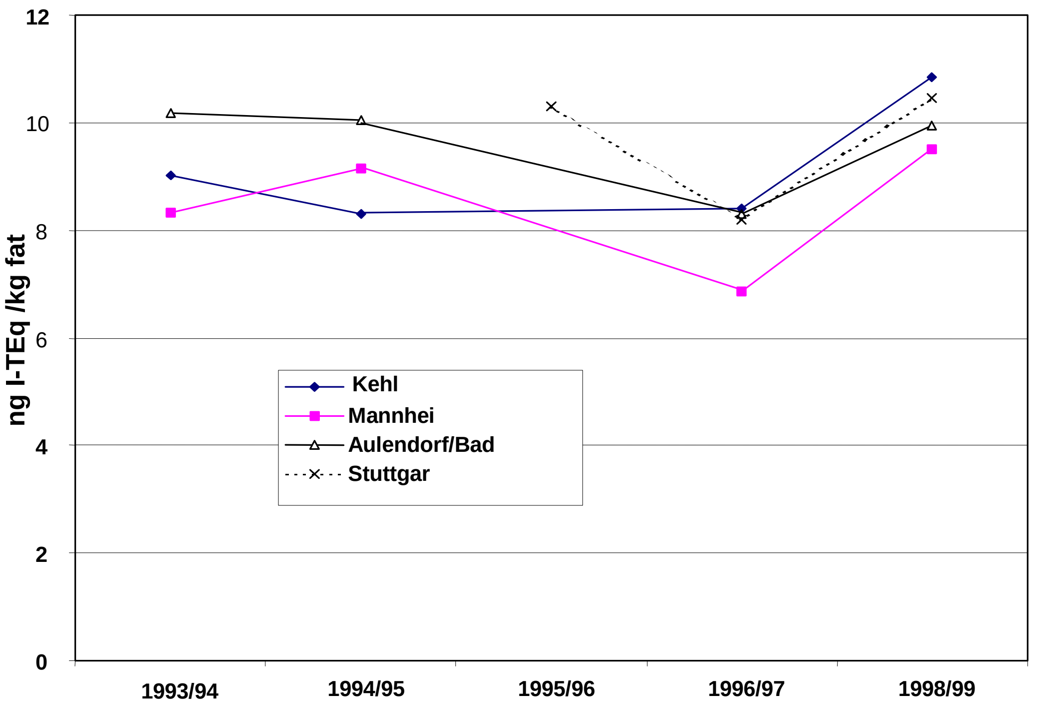
Girls	1994/95	1995/96	1996/97	1998/99	Change 1996/97 till 1998/99
Urban area1 (Mannheim)	9.0			9.1	
Urban area 2 (Stuttgart)		10.9	7.5	9.4	25.3 %
Urban area in the rural area (Kehl)				10.5	
Rural area (Aulendorf/Bad Waldsee)	9.5		7.4	9,.	24.3 %

The decline of the dioxin content in the blood of 9– to 11-year old children observed before 1996/1997 corresponds altogether to the declining background concentration in the population in Germany as it has been also documented by the results of blood samples of adults and human milk and dioxin intake assessments before 1997.

Table 29: PCDD/PCDF concentrations in collecting blood samples, data of boys and girls summed up (partly calculated as average) according to place and year of investigation (ng I-TEq /kg fat)

Place of investigation	**Year of investigation**					**Change**
	1993/94	**1994/95**	**1995/96**	**1996/97**	**1998/99**	**1996/97 till 1998/99**
Urban area1 (Mannheim)	8.3	9.2		6.9	9.5	38.2 %
Urban area 2 (Stuttgart)			10.3	8.2	10.5	28,0 %
Urban area in the rural area (Kehl)	9.0	8,3		8.4	10.9	29.3 %
Rural area (Aulendorf/Bad Waldsee)	10.2	10.1		8.3	10.0	20.2 %

Fig. 53: Temporal trends of dioxin contents in collecting blood samples (boys and girls) in the 4 places of investigations (ng I-TEq/kg fat)



The decline of the dioxin contents observed until 1996/1997 is confronted with an increase in the following year. The trend synchronous in all 4 places of investigation points to the fact that the increase observed between 1996/1997 and 1998/1999 is not only due to a variability caused by accident in the compositions of the test groups as to the confounders but possibly also to other factors contributing to this increase.

An important factor for the dioxin content are the analyzing methods to be used. Immediately after the period of investigation, i.e. in a few measuring cycles, the samples were analyzed by the same laboratory. There should be considered that the concentrations of many congeners measured may lay only slightly above the detection limit and additionally this limit may vary between the measuring cycles.

Parallels with the increase in the dioxin contents in human milk samples from Baden-Wuerttemberg (approx. 20 % increase) and from North Rhine—Westphalia (approx. 15 % increase) observed in 1998 may point to a connection with the citrus pulp problems.

6.6.6 Connection between citrus pulp contaminated by dioxins and dioxin contents in human samples

In the years 1990-1997 a continuous decline of the dioxin contents was detected in human milk as well as in blood. Thus, in 1998 the average dioxin contents (I-TEq) in human milk from the Laender Baden-Wuerttemberg and North Rhine-Westphalia were by about 22 or 16 % higher than in the preceding year. The number of the samples investigated in this period is, however, small. In Chapter 6.6.4 there was referred to potential variabilities as to the various influencing factors in the composition of the test groups.

Also a temporally parallel rising trend of the dioxin content in the blood of 9- to 11-year old children from Baden-Wuerttemberg is remarkable. In the last period of investigation 1998/1999 an increase in the dioxin contents (I-TEq) by 20 to 40 %, on average, is to be observed as compared to the preceding period of investigation (1996-1997) in all 4 locations of investigation. An analoguous trend of the background contamination by dioxins is detected for blood samples of adults.

The various investigations of human milk and blood were carried out independent of each other and by different laboratories. Thus, laboratory-associated effects on the results are unlikely. The question whether in 1998 an additional increment may have been present in the exposure shall be discussed here by means of the dioxin data in human milk reported from Baden-Wuerttemberg and NRW and the dioxin contents in collecting blood samples of children from Baden-Wuerttemberg.

The citrus pulp problems are in a temporal connection with the observed increase in the dioxin contamination. Since September 1997 and also in spring 1998 citrus pulp contaminated by dioxins from Brazil used as a component of feedingstuffs has shown dioxin contents up to 10 times higher and characteristic shifts in the congener pattern of cow's milk and milk products, beef and veal. The increase of TCDD and 1,2,3,7,8-PeCDD was most remarkable. Furthermore the contents of hexafurans and 2,3,4,7,8 PeCDF were slightly increased as compared with the usual background concentration (see Chapter 6.5.8, Fig. 42). How big the share of higher contaminated foodstuffs coming on the market was is not yet known.

The increase of the dioxin content referred to cow's milk, milk products, beef and veal. These are products which are contributing to the daily dioxin intake in essential shares. In addition, this increase was to be detected in many Federal Laender and European states over a few months with the usual trade routes from the main import port Rotterdam resulting in regional differences. Thus, the question is raised whether this contamination detectable in important foodstuffs of animal origin may have resulted in an insignificant temporary trend reverse also in human samples.

In connection with the increase in the dioxin contents in the human samples from Baden-Wuerttemberg and North Rhine-Westphalia thus modifications in the congener pattern might give information on a potential connection with the consumption of such contaminated foodstuffs. In contrast to the total dioxin content (I-TEq) congener-specific shifts should be less affected by factors such as age and number of breast-feeding periods (human milk) or sex and breast-feeding status (blood).

In the human milk samples from NRW and Baden-Wuerttemberg changes in the content of the congeners relevant to the citrus pulp contamination show a uniform basic pattern: continuously declining values between 1994 and 1997 and an increase in 1998 by 12 – 30 % depending on the congener (Tab. 30). Primarily 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD (Fig. 54) and 2,3,4,7,8-PeCDF (Fig. 55), are remarkably, hexadioxins are less remarkably increased. In addition the hexafuran contents have slightly increased in Baden-Wuerttemberg. However, an elevated contribution of TCDD to the total dioxin content – as has been observed in cow's milk – was not found in human milk samples.

Fig. 54: Temporal trends of the content of TCDD and 1,2,3,7,8-PeCDD in human milk samples from Baden-Wuerttemberg and North Rhine-Westphalia

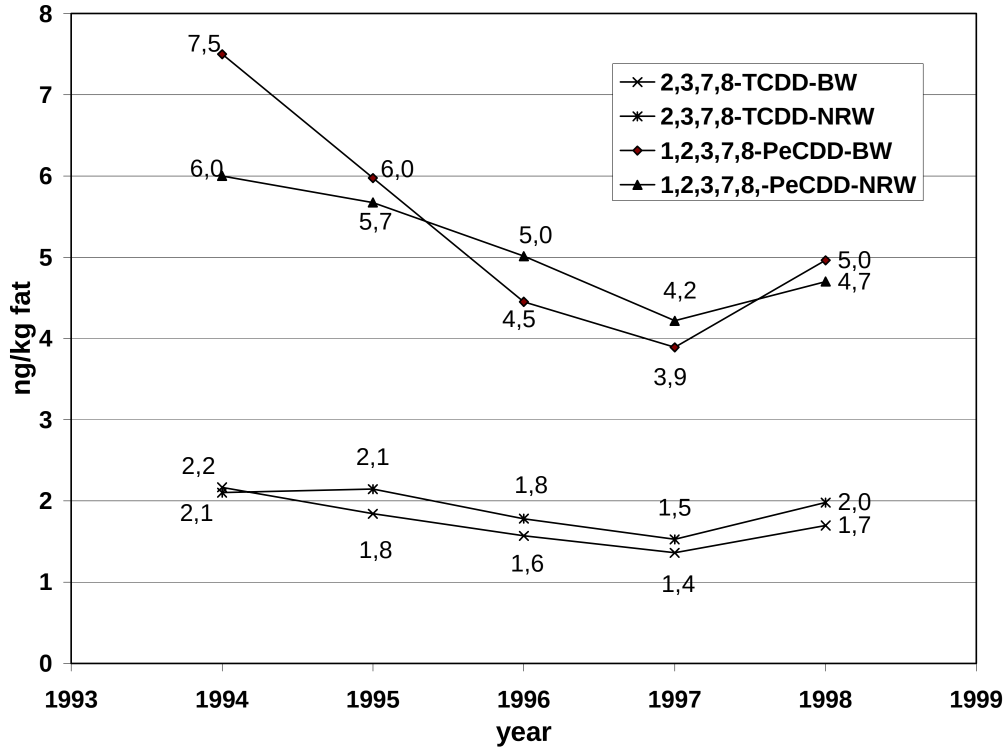


Fig. 55: Temporal trends of the 2,3,4,7,8-PeCDF contents in human milk samples from Baden-Wuerttemberg and North Rhine-Westphalia

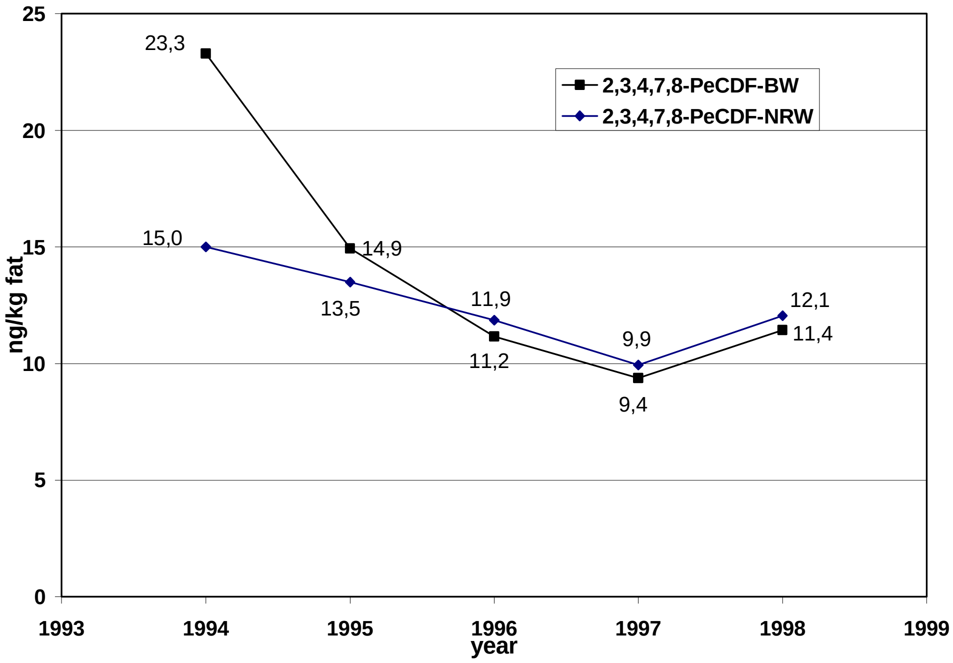


Table 30: Contents of various congeners in human milk from NRW, BW and LS for the period between 1994 and 1998 (data in ng/kg fat)

	Year	2,3,7,8 -TCDF	**2,3,7,8 -TCDD**	1,2,3,7,8 -PeCDF	**2,3,4,7,8 -PeCDF**	**1,2,3,7,8 -PeCDD**	1,2,3,4,7,8 -HxCDF	1,2,3,6,7,8 -HxCDF	2,3,4,6,7,8 -HxCDF	1,2,3,4,7,8 -HxCDD	1,2,3,6,7,8 -HxCDD	1,2,3,7,8,9 -HxCDD	1,2,3,4,6,7,8 -HpCDF	1,2,3,4,6,7,8 -HpCDD	1,2,3,4,6,7,8,9 -OCDD	**I-TEq**
NRW	1998	0.70	2.0	0.20	12.0	4.7	3.8	3.0	1.3	3.4	17.4	2.1	2.3	16.7	100.1	13.8
NRW	1997	0.58	1.5	0.24	9.9	4.2	4.0	2.9	1.4	3.6	14.6	2.2	2.2	21.6	109.6	11.9
NRW	1996	0.67	1.8	0.21	11.9	5.0	4.0	3.2	1.4	4.0	18.9	2.8	5.5	19.2	98.6	14.1
NRW	1995	0.56	2.1	0.26	13.5	5.7	4.9	3.7	1.7	4.8	21.8	2.9	2.8	21.9	121.8	16.1
NRW	1994	0.60	2.1	0.20	15.0	6.0	5.3	4.0	1.9	4.9	22.1	32	3.0	23.6	110.4	17.2
Increase 1997/98		21.3%	**29.6%**	-19.2%	**21.3%**	**11.4%**	-6.1%	2.2%	-5.8%	-4.9%	19.6%	-7.5%	7.7%	-22.6%	-8.7%	**16.3%**
BW	1998	0.51	1.7	0.27	11.4	5.0	3.5	3.2	1.4	4.0	16.6	3.2	2.4	21.9	113.5	13.5
BW	1997	0.44	1.4	0.22	9.4	3.9	3.0	2.8	1.2	3.3	13.0	3.1	3.0	20.3	85.8	11.0
BW	1996	0.57	1.6	0.32	11.2	4.4	3.4	3.0	1.4	3.7	13.8	3.7	2.7	26.6	103.7	12.8
BW	1995	1.35	1.8	1.24	14.9	6.0	6.6	4.8	2.5	5.5	19.1	4.3	5.6	34.8	117.8	17.5
BW	1994	0.74	2.2	0.33	23.3	7.5	7.0	5.7	2.6	6.3	27.4	5.6	6,2	50,6	213,1	23,9
Increase1997/98		14.7%	**24.6%**	22.0%	**21.9%**	**27.5%**	15.9%	16.8%	15.4%	22,.%	27.1%	2,.%	-19.5%	8.0%	32.2%	**22.6%**
NI	1998	0.24	1.1	0.17	13.6	3.6	2.7	3.0	0.8	2.2	12.8	2.0	1.6	14.9	69.0	12.3
NI	1997	0.28	1.3	0.14	12.5	4.0	3.0	3.2	1.0	2.2	16.3	2.4	2.4	19.4	107.17	12.7
NI	1996	0,.8	1.6	0.22	17.6	5.0	3.8	3.9	1.4	3,3	18.5	2.6	2.9	23.9	122.2	16.6
NI	1995	0.38	2.2	0.32	19.1	6.7	6.0	5.8	1.5	4.0	13.2	2..4	5.0	22.3	88.8	18.7
NI	1994	0.29	1.6	0.16	17.1	5.9	4.3	4.4	1.2	2.3	18.2	2.5	4.2	24.8	98.9	16.8
Increase1997/98		-14.6%	**-18.1%**	20.6%	**8.7%**	**-9.1%**	-8.3%	-6.2%	-13.7%	4.0%	-21.6%	-19.6%	-32.0%	-22.9%	-35.6%	**-3.3%**
BY	1998	0.25	1.1	0.25	8.6	3.8	2.6	2.3	1.4	3.0	14.5	2.9	3.4	30.5	184.9	10.5
BY	1997	0.25	1.2	0.25	11.3	4.9	3.0	2.8	1.7	4.3	15.2	2.8	2.8	23.8	109.7	12.8
Increase1997/98		0%	**-11.3%**	0%	**-25.9%**	**-22.5%**	-13.3%	-17.9%	-17.7%	-30.3%	-4.6%	3.6%	21.4%	28.2%	68.6%	**-18.0%**

Also in collecting blood samples of 9 to 11-year old children analoguous modifications of the congener pattern were detected. An equal trend – comparative constancy in the framework of analytical variation or decline of the contents between 1993/1994 and 1996/1997 and an increase by 30 – 125 % in the year 1998/1999 - is to be detected for TCDD in all 4 locations of investigation (Tab. 31, Fig. 56). This clear increase in TCDD is also reflected in a higher contribution of TCDD to the I-TEq. It rises to approx. 18-24 % in all 4 locations of investigations in 1998/1999. Also for 1,2,3,7,8 pentadioxin, 2,3,4,7,8 pentafuran and the hexafurans a comparable increase is to be observed in all 4 locations between 1996/1997 and 1998/1999 (Tab. 31). Yet, the actual contents are mostly in the range of the preceding periods of investigations. Thus, this picture is less clear for these congeners.

Fig. 56: Temporal trends of the TCDD contents in collecting blood samples of 9- to 11-year old children from Baden-Wuerttemberg, differentiated by the 4 locations of investigation

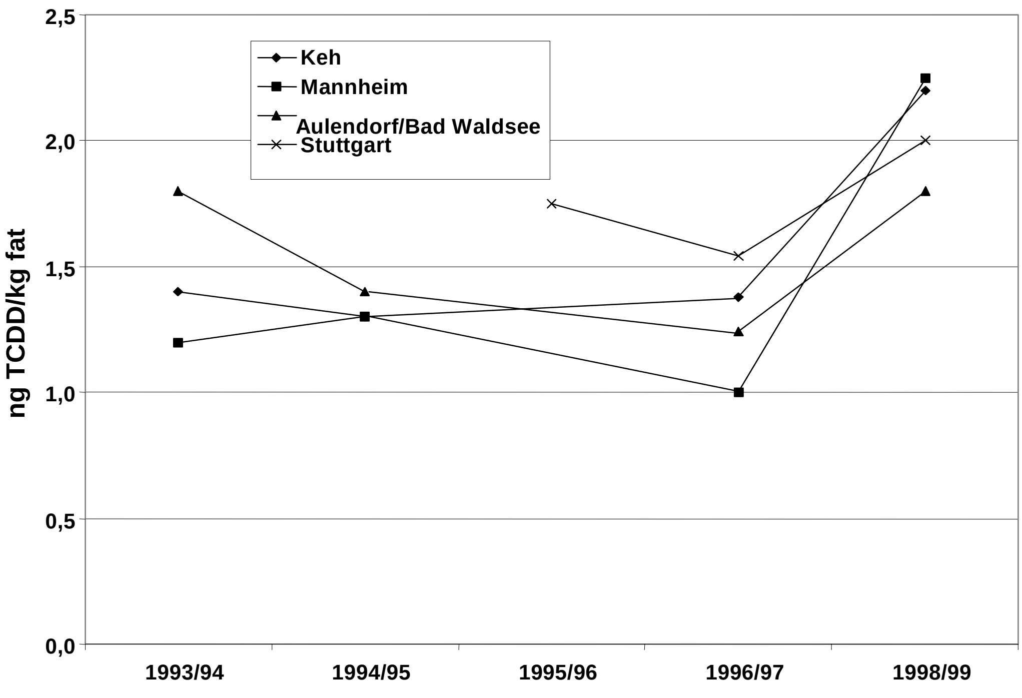


Table 31: Contents of essential congeners in collecting blood samples of 9– to 11-year old children from Baden-Wuerttemberg (data in ng/kg fat)

Location	Year	**2,3,7,8 -TCDD**	**2,3,4,7,8 -PeCDF**	**1,2,3,7,8 -PeCDD**	1,2,3,4,7,8 -HxCDF	1,2,3,6,7,8 -HxCDF	2,3,4,6,7,8 -HxCDF	1,2,3,4,7,8 -HxCDD	1,2,3,6,7,8 -HxCDD	1,2,3,7,8,9 -HxCDD	1,2,3,4,6,7,8 -HpCDF	1,2,3,4,6,7,8 -HpCDD	OCDD	**I-TEq**	TCDD share in I-TEq
Rural area	93/94	1.8	5.5	3.6	3.8	3.5	1.9	2.7	13.0	4.6	13.7	30.2	247.2	10.2	17.7%
Aulendorf/	94/95	1.4	5.7	3.6	4.7	3.7	2.4	2.7	13.7	3.6	17.0	36.9	235.6	10.0	14.0%
Bad Waldsee	96/97	1.2	5.2	3.0	3.1	2.9	1.5	1.7	11.5	2.7	11.3	25.4	189.8	8.3	14.9%
	98/99	1.8	5.	4.0	5.1	3.2	2.0	3.1	9.1	3.5	12.2	34.9	236.2	10.0	18.0%
Increase 1996/97 to 1998/99		**45%**	**3%**	**31%**	66%	11%	33%	80%	-21%	28%	8%	37%	24%	**20%**	
Urban area in the rural area	93/94	1.4	4.9	3.2	4.5	2.9	1.9	2.8	12.2	4.0	12.3	26.3	180.5	9.0	15.5%
Kehl	94/95	1.3	4.9	3.4	4.1	3.2	2.9	0.7	7.8	2.3	7.2	29.7	209.9	8.3	15.6%
	96/97	1.4	5.3	2.9	3.3	2.9	1.7	2.1	9.7	3.1	9.0	25.1	151.1	8.4	16.5%
	98/99	2.2	6.4	3.7	4.4	4.0	2.8	3.0	10.0	3.2	10.5	29.9	247.5	10.8	20.3%
Increase 19 96/97 to 1998/99		**59%**	**20%**	**28%**	33%	38%	62%	43%	3%	2%	16%	19%	64%	**29%**	
Urban area1	93/94	1.2	4.8	2.7	3.9	2.3	1.8	2.3	11.0	3.8	11.7	31.5	237.6	8.3	14.4%
Mannheim	94/95	1.3	4.8	3.0	4.9	3.4	2.5	2.3	13.2	4.0	12.0	36.5	281.4	9.1	14.2%
	96/97	1.0	3.9	2.4	3.2	2.8	2.0	2.0	8.3	2.4	8.7	24.9	184.8	6.9	14.5%
	98/99	2.3	5.5	3.0	4.3	3.6	2.6	2.2	6.9	2.7	9.2	27.5	252.3	9.5	23.6%
Increase 1996/97 to 1998/99		**125%**	**40%**	**25%**	34%	27%	30%	10%	-17%	13%	5%	10%	36%	**39%**	
Urban area2	95/96	1.8	6.3	3.8	4.6	3.3	2.4	2.8	12.1	3.4	8.9	20.4	186.6	10.3	17.0%
Stuttgart	96/97	1.5	5.0	2.7	3.3	2.7	2,0	2.0	9.2	2.6	9.8	22.8	184.5	8.2	18.9%
	98/99	2,0	6.5	3.7	4.7	4.2	2.7	2.1	9.7	2.8	10.5	24.9	226.6	10.4	19.2%
Increase 19 96/97 to 1998/99		**29%**	**29%**	**35%**	43%	58%	38%	5%	5%	10%	8%	9%	23%	**27%**	

The qualitative modifications of the congener pattern of human samples from Baden-Wuerttemberg and North Rhine-Westphalia shows analogies with the specific modifications described for contaminated cow's milk and milk products caused by citrus pulp. Also the relative share of the cow's milk samples additionally contaminated by citrus pulp is the highest in these two Federal Laender (see Chapter 6.5.8, Tab. 17). Additionally to the temporal coincidence this points to a potential connection between the consumption of foodstuffs where Brazilian citrus pulp in feedingstuffs resulted in higher dioxin contents of foodstuffs and an increase of dioxin contents observed in human milk and blood from Baden-Wuerttemberg and North Rhine-Westphalia.

The question arises why the dioxin contents detected in 1998 and the congener pattern observed in human milk from Lower Saxony do not show the same characteristics. Differences in the regional and temporal distributions of the Brazilian citrus pulp contaminated by dioxin in the various Federal Laender may be an explanation. The data of the contaminated cow's milk samples in the individual Federal Laender allow this interpretation.

To sum up there should be mentioned that there cannot be excluded that the elevated dioxin input into the food chain caused by Brazilian citrus pulp throughout Europe contributed to the slight increase of the dioxin contents in human samples from various Federal Laender observed in 1998. Though this connection may not be proved free of doubts – also the small numbers of samples in this period should be taken into account – various facts of evidence point to it. As a consequence efforts shall be further supported to record potential input pathways for dioxins into the food chain and to eliminate them.

6.6.7 Evaluation of human data

The data available in the human milk and dioxin human data base allow a well-founded assessment of the trend of the contamination of humans by dioxins. The dioxin data document that the contents in human milk have decreased by approx. 60 % since 1990. This refers to average as well as maximum contaminations (1998: MW = 12,9 ng I-TEq/kg fat, maximum = 28,9 ng I-TEq/kg fat). Declining dioxin contents have also been detected in blood samples of 9 - to 11-year old children from Baden-Wuerttemberg.

The declining dioxin contamination detected in human samples is due to declining dioxin contents in foodstuffs and a smaller dioxin intake resulting from it. This trend shows the positive effects and the success of numerous measures aimed at minimizing the dioxin emission into environment and reducing the exposure of the population.

In spite of the successes reached so far the average daily dioxin intake of a 4-month-old breast-fed infant is with 57 pg I-TEq/kg of body weight by nearly two orders of magnitude higher than that of adults. The increased dioxin intake by breast-feeding causes still a 20 % higher dioxin contamination detected for 9 - to 11-year old children. Since on the other hand breast-feeding has been proven to have positive effects on infant development, it is nevertheless still recommended by the WHO and the German National Commission for Breast-feeding of the former BgVV.

In addition, the data available document that still individual local sources (contaminated industrial sites) or other factors (e.g. citrus pulp) cause increased dioxin contents in human samples.

The search for potential causes for higher dioxin contents in human matrices as human milk presupposes further accompanying information on the individual data sets. Data of the mother as e.g. on the establishment of industries near the residence, on special habits of nutrition (vegetarian, high consumption of fish, self-supply with foodstuffs) or on potential occupational exposures are imperative for that, yet also for checking the comparability of test groups. Tests of human milk samples from an industrially contaminated area underline that considering most various influencing or exposure factors based on the concerted questionnaire are a helpful instrument for detecting the causes in the case of remarkably high dioxin contents in human milk. Purposeful, source-related measures should be taken only after clarifying the exposure factors and determining the transfer pathways.

The discussion of the human data allowing to recognize or assume an additional exposure exceeding the present background concentration shows the special necessity of inter-compartment considerations. In the discussion there was shown that the transfer pathways „contaminated feedingstuffs – foodstuffs man“ or „increased dioxin emission – deposition – foodstuffs man“ are even today relevant to tracing back and clarifying higher dioxin contaminations of humans.

The documentation of the data on dioxins in human milk and other human samples based on the concerted and harmonized questionnaire in the human milk and dioxin human data base and their linkage with the dioxin data base (environmental and foodstuff data) provided the required prerequisites for these inter-media considerations of the transfer pathways.

Taking further measures aimed at identifying the sources and reducing the dioxin exposure and thus reducing the dioxin body burden is necessary. This requirement is also raised by the WHO. It is derived from the necessity to minimize the postnatal exposure of the breast-fed infant which due to the prenatal exposure is in the background concentration area. Human milk or blood tests offer themselves also for checking the success and indicating additional exposures.

6.6.8 References for the Chapter „Human data“

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7. Summary

The dioxin reference program is founded on a decision by the 37th conference of Federal and Länder Ministers for the Environment of the Federal Republic of Germany, 21-22 November 1991. At that time, there were calls for further studies to improve the data on dioxins in soil, water, air, foodstuffs and feedingstuffs, for the initiation and coordination of future government and Länder measuring programs and for a centralized documentation and evaluation of the results. To implement this it was agreed within the framework of the Government/Länder working group on DIOXINS, *inter alia*, to carry out a nationwide dioxin measuring program of several years duration. By observing dioxin contamination in the environment over a longer period it should be possible to make statements on the trend. In particular, the effects on contamination in foodstuffs and in the different environmental media of the dioxin emitters' reduction measures, then introduced, were to be determined. The dioxin reference measuring program was started in mid 1994. For the individual compartments (ambient air, cow's milk, human milk) the evaluation also referred to data up to the year 1989. This also allows the temporal progress of dioxin contamination to be documented for longer periods.

Air emissions

For the compartment air emissions, the database DIOXINS contains measurements from several Länder on the dioxin concentrations in the air and on dioxin deposition. In general the data groups for each location are not from individual samples but from a series of samples which allow the average dioxin concentrations or dioxin deposition to be described for the respective year. The locations for the dioxin sampling are primarily in areas with a high industrial density or in centres of population with high emission density; these results from the dioxin samples are therefore less suited for estimating the average dioxin concentrations in the Laender, but describe primarily the situation in focal areas of impact.

Dioxin measurements for the compartment air emissions have been documented in the database DIOXINS since 1990. For individual sampling locations there are homogenous series of samples covering many years, enabling statements to be made on the trend of dioxin concentrations. An example of this is the presentation of the trend of dioxin concentrations at stations in Hesse and Thuringia for the sampling period 1990-1999, based on the annual average values (fig. 4, p.14). This reveals that there has been a clear decline in the dioxin concentrations in the air since sampling began in 1990; in industrial centres the annual average decreased from over 100 fg I-TEQ/m^3 to below 50 fg I-TEQ/m^3, and in rural areas the annual average dropped from over 40 fg I-TEQ/m^3 to below 20 fg I-TEQ/m^3.

Small fluctuations in concentration levels overlie the decrease in dioxin concentrations evidenced by the series of samples taken over several years. These can essentially be explained by variations in meteorological conditions occurring during the years of sampling; they can, however, also be influenced by particular emissions circumstances in the area of the sampling station. Further evaluations consider seasonal differences in dioxin concentrations. Examples are included for average homology profiles for dioxin concentration and the dioxin deposition.

Soil

Soil represents a link between the environmental compartments and the food chain. Dioxin concentrations in soils can essentially be traced to inputs via air pathways and inputs from sewage sludge and compost. Soils are the substrate for plants, and as such can influence the dioxin concentrations of vegetable and animal foodstuffs.

Due to the short time-period covered by the study, the data obtained under the dioxin reference measurement program concerning the dioxin concentrations in soils is not suitable for statements on the trend; they do, however, give a good overall picture of the dioxin contamination situation. The dioxin reference measurement program distinguished between 3 area types of settlement structures: urban areas, urban fringe and rural areas. The large number of sample results shows clear differences between these three area types. As expected, the dioxin concentrations in urban areas are higher that those at the urban fringe, which in turn are higher than those in rural areas.

If no distinctions are made between uses, the median of the PCDD/F concentrations in soil is 1.08 ng I-TEQ/kg dry matter (dm) in urban areas, 0.49 ng I-TEQ/kg dm at the urban fringe, and 0.24 ng I-TEQ/kg dm in rural areas (fig 11 p. 28). In rural areas, the organic layer of soil in woodland has the highest concentrations, with a median of 12.12 ng I-TEQ/kg dm, while topsoil concentrations in arable land and pasture is below 1 ng I-TEQ/kg dm. It should be noted that for these values the calculation of the TEQ did not take a detection limit into account.

Sediments and suspended matter

As in the case for soil, the duration of the study is not sufficient to allow a statement on the trend of dioxin concentrations in sediments /suspended matter. Resuspensions are probably one of the reasons why the recordable decline of dioxin concentrations in the air cannot yet be similarly observed in surface waters. The rivers Rhine and Elbe show dioxin concentrations of the same magnitude.

Bioindicators

Biomonitoring allows us, with the help of bioindicators, to observe the temporal progress of environmental influences on the plant environment. Spruce needles, green cabbage, lettuce and grass samples are particularly suited for studying dioxin contamination in the environment. Green cabbage is a suitable indicator plant for airborne pollutants, due to its large surface and special leaf structure, and its long growing period. In some locations, seasonal fluctuations in dioxin concentrations are clearly apparent; the increase in the burning of fuel during the winter months has a notable influence on dioxin emissions and ultimately on the bioindicators.

Cow's milk

Under the dioxin reference measurement program, studies were made on dairy and collecting tank milk, farm milk and dairy products. The study of dairy milk and other dairy products takes in larger catchment areas, while studying farm milk permits the observation of local influences.

Due to the vast extent of the data, which in some cases covers more than ten years, it is possible to make definite statements on the temporal trend of dioxin contamination in cow's milk, on seasonal fluctuations and on the influence of contaminated feedingstuffs (citrus pulp).

The dioxin concentrations in cow's milk have declined by more than 60% over the total study period. This trend was only interrupted temporarily by the use of contaminated citrus pulp as feedingstuff in 1997/97. At this time dioxin concentrations on average more than doubled.

The decline of dioxin contamination is to be observed equally in urban areas and rural areas. As expected, the dioxin concentrations in milk from urban areas are higher than those in milk from rural areas. In both area types, however, dioxin contaminations are in decline. The difference in concentration levels between urban areas and rural areas is also becoming smaller, although a total equality in the levels is not to be expected. This is because the diffuse dioxin contamination in urban areas arising from the population density will continue in future to be higher that in rural areas.

The Government/Länder working group on DIOXINS proposed in its 2nd report 1993 that for dioxin concentrations in milk, the target parameter of 0.9 pg I-TEq/g fat should be aimed for. This should be achieved or undercut in the long term by reducing dioxin inputs into the environment. At the beginning of the 90s, 95% of the milk samples failed to meet this parameter. In recent years, however – due to the general decline of dioxin inputs into the environment – the target value for milk and milk products has only been exceeded in isolated cases.

Based on a location, the dioxin concentrations in cow's milk reveal seasonal fluctuations, depending on feeding. Concentrations are highest at the end of the winter feeding, and lowest after the summer grazing. These fluctuations were taken into consideration during the evaluation of the time series.

Human data

The data available in the database on „Dioxins in Human Milk and Blood Plasma“ indicates a decline of about 60% in the past ten years of both the average and the maximum dioxin concentrations in human milk from Germany. The average background concentration has been continuously decreasing since the end of the 1980s, from 30-32 ng I-TEQ/kg fat to 13 ng I-TEQ/kg fat in 1998 (figure 48 p. 87). A study program in Baden-Wuerttemberg, which measured *i.a*. the dioxin concentrations in blood samples of school children between 9 and 11 years, also ascertained a decline.

4-month-old infants being breast-fed take in 57 pg I- TEQ/kg body weight via the mother's milk. The average daily dioxin intake is therefore clearly higher than that of adults, and exceeds the TDI values derived by the WHO of 1-4 pg WHO-TEQ/kg body weight and day – although these apply to adults and refer to intake over a lifetime. The increased dioxin intake connected with breast-feeding leads to children of 9-11 years who were breast-fed having on average 20% higher levels of dioxin in the body than children who were not breast fed. Since, on the other hand, breast-feeding has been proven to have a positive effect on infant development, it is nevertheless still recommended both by the WHO and Germany's National Commission for Breast-feeding.

8. Conclusions

The aims of data collection on “Dioxin concentrations in the environment” (see 3rd report of the Government/Länder working group on DIOXINS), and those of the “Dioxin reference measuring program” (cf 4th Report) were achieved to a large degree. This was mainly due to the data collected by the Länder and to the central database DIOXINS run by the Federal Environmental Agency (UBA) and the Federal Institute for Consumer Health and Veterinary Medicine (BgVV). The aims in question were:

- to create an overview of the level of environmental pollution and contamination of humans from dioxins,
- to make statements on trends of dioxin contamination of environmental media, biota and humans,
- to controll and document the success of environmental protection measures taken to reduce the input of dioxins into the environment, and the exposure of humans.

These reports show that the dioxin contamination of humans and the environment has declined considerably over the past ten years. The administrative and technical measures introduced over a decade ago have had a lasting effect.

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 hazardous incidents.

Data collected in the database DIOXINS reflect the status of environmental pollution by dioxins. It is possible to trace exposure from the source to humans by using the information on emissions, ambient air, deposition, feedstuffs, foodstuffs and human data. However, there are almost no data at all on feedstuffs. Feedstuffs were not investigated within the “Dioxine reference measuring program”, because this compartiment is not sufficient to make statements on trend of dioxin contamination of the environment. On the other hand, feedstuffs are an essential link in the chain. Implementation of the pilot project “dioxin-feedstuffs monitoring” (resolution of the Conference of Agricultural Ministers, 23 March 2001) would remedy the existing deficit.

Although pollution of humans was reduced by approx. 60%, further efforts are necessary in order to ensure sufficient protection from dioxins for humans and the environment. In order to promptly identify incidents, contaminated feedstuffs and foodstuffs and to avert damage, the Government/Länder working group on DIOXINS proposes

- to continue the reference measuring program,
- to continue the operation of the database DIOXINS (data input, evaluation, reporting),
- and to inform the public also in future about the results.