

Compilation of EU Dioxin Exposure and Health Data

Task 2 – Environmental Levels

Technical Annex

Report produced for
European Commission DG Environment
UK Department of the Environment, Transport and
the Regions (DETR)

October 1999

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A1 Austria

A1.1 SOIL

From Austria, dioxin results are available from a total of 90 soil samples (see Table A 1). Of these, 20 samples are the results from a special investigation program of the mid 1980s around the copper smelter at Brixlegg. The highest concentration found was 332 ng I-TEQ/kg d.m. for grassland in the Brixlegg region (Riss 1990, 1993, Riss *et al.* 1990). All samples were analysed according to given depth. The majority of samples were from forests and were from a survey performed in the Linz Region in 1989 (Weiss and Riss 1992). The highest concentrations were found in the O-horizons of forests with concentrations up to 63 ng I-TEQ/kg d.m. In the upper layers of the mineral soils (0-10 cm) from forest locations, the concentrations were much lower and ranged from 0.5 ng I-TEQ/kg d.m. to 5.8 ng I-TEQ/kg d.m. The concentrations decreased with increasing sampling depths. For grassland the median concentration from 13 pooled samples was 3.3 ng I-TEQ/kg d.m. with a maximum of 14.4 ng I-TEQ/kg d.m. (Umweltbundesamt 1993).

Table A 1: Austria - Soil. Concentrations in ng I-TEQ/kg d.m.

Type	Region	Type of Location	Horizon	Type of Sample	Date	N*	Min	Max	Mean	Median
Grassland	Linz Region	Urban	0-5 cm	Pooled (n** = 25)	Jul 89	13	1.6	14.4	5.8	3.3
Forest (mixed, managed)	Linz Region	Urban	O	Pooled (n = 6)	Jul 89	1			63.5	
Forest (mixed, managed)	Linz Region	Urban	0-5 cm	Pooled (n = 6)	Jul 89	1			3.7	
Forest (mixed, managed)	Linz Region	Urban	0-10 cm	Pooled (n = 6)	Jul 89	1			5.5	
Forest (mixed, managed)	Linz Region	Urban	10-20 cm	Pooled (n = 6)	Jul 89	1			0.07	
Forest (mixed, managed)	Linz Region	Urban	20-30 cm	Pooled (n = 6)	Jul 89	1			0.01	
Forest (mixed, managed)	Linz Region	Rural	O	Pooled (n = 6)	Jul 89	1			12	
Forest (mixed, managed)	Linz Region	Rural	0-5 cm	Pooled (n = 6)	Jul 89	1			3.1	
Forest (mixed, managed)	Linz Region	Rural	0-5 cm	Pooled (n = 6)	Jul 89	1			3.8	
Forest (mixed, managed)	Linz Region	Rural	10-20 cm	Pooled (n = 6)	Jul 89	1			0.01	
Forest (mixed, managed)	Linz Region	Rural	20-30 cm	Pooled (n = 6)	Jul 89	1			<0.01	
Forest (Norway spruce)	across Austria	Background	O	Pooled (n = 10)	Aug 93	25	1.6	31	6.9	4
Forest (Norway spruce)	across Austria	Background	0-5 cm	Pooled (n = 10)	Aug 93	5	2.9	5.8	4	3.3
Forest (Norway spruce)	across Austria	Background	5-10 cm	Pooled (n = 10)	Aug 93	5	0.5	2.5	1.4	0.7
Grassland	Brixlegg/Tyrol	Contaminated	0-5 cm	Pooled (n=30)	1987+1988	20	0	332		
Soil, unmanaged, undisturbed	Amstetten Area	Suburban	0-5 cm	Pooled (n=30)	May 93	12	1.3	8.9		

N* = number of samples analysed individually; n** = number of samples in a pool

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Table A 2: Austria - Air. Sampling time: 1992-1998. Concentrations in fg I-TEQ/m³

State/City/Region	Country	Type	Date/Sampling period	n	Min	Max	Mean	Median	Upper Quar.	Lower Quartile
GRAZ-Süd	Steiermark	suburban	Nov 1992-Oct 1993	17	8.7	587.3	119.3	67.8	159.6	25.5
LINZ-Orf-Zentrum	Oberösterreich	urban	Nov 1992-Oct 1994	15	21.8	322.4	74.4	48.0	85.7	31.5
STEYREGG-Weih	Oberösterreich	suburban	Nov 1992-Oct 1995	16	16.0	366.5	75.7	46.8	75.4	34.5
WIEN-9. Bez.	Wien	urban	Nov 1992-Oct 1996	16	12.5	435.6	79.0	62.8	81.1	27.9
WIEN-14. Bez.	Wien	urban	Nov 1992-Oct 1997	15	9.3	129.2	37.0	33.5	43.6	15.7
WIEN-22. Bez.	Wien	suburban	Nov 1992-Oct 1998	5	11.2	96.2	55.1	63.9	70.4	33.6
Ulmerfeld-Amstetten	Niederösterreich	rural	Sept. 1993 -Sept. 1994	12	10.8	110.4	36.4	24.1	47.2	16.2
Graz-Süd (*)	Steiermark	suburban	Dec. 1993 -Feb. 1994	4	245.7	365.4	311.2	316.8	334.5	293.5
Graz-Süd	Steiermark	suburban	Dec. 1993 -Feb. 1995	4	255.7	413.9	314.2	293.5	339.3	268.4
Graz-Mitte	Steiermark	urban	Dec. 1993 -Feb. 1996	4	140.2	375.3	250.9	244.0	277.4	217.5
Graz-Ost	Steiermark	urban	Dec. 1993 -Feb. 1997	4	138.9	302.4	197.7	174.7	215.9	156.5
Graz-Lustbühel	Steiermark	suburban	Dec. 1993 -Feb. 1998	4	72.4	201.7	121.4	105.7	132.0	95.1
Linz-Kleinmünchen	Oberösterreich	urban	Dec. 1994 -Mar 1995	3	61.5	127.5	104.0	123.1	125.3	92.3
Linz-ORF-Zentrum	Oberösterreich	urban	Dec. 1994 -Mar 1996	3	2.1	130.1	65.9	65.6	97.9	33.8
Linz-Ursulinenhof	Oberösterreich	urban	Dec. 1994 -Mar 1997	3	69.4	179.3	120.0	111.2	145.2	90.3
Steyregg	Oberösterreich	suburban	Dec. 1994 -Mar 1998	3	59.4	162.1	107.2	100.1	131.1	79.8
LinzPöstlingberg	Oberösterreich	suburban	Dec. 1994 -Mar 1999	3	1.3	46.1	25.5	29.2	37.6	15.3
Leoben-Werkskindergarten	Steiermark	urban	Jun. 1995 -Jan. 1996	3	120.6	324.6	206.9	175.6	250.1	148.1
Leoben-Moserhofstr.	Steiermark	urban	Jun. 1995 -Jan. 1997	3	43.7	141.0	77.8	48.8	94.9	46.2
Leoben-BFI	Steiermark	urban	Jun. 1995 -Jan. 1998	4	68.7	261.6	149.7	134.3	175.8	108.2
St. Peter Freienstein	Steiermark	urban	Jun. 1995 -Jan. 1999	2	61.5	306.8	184.1	184.1	245.5	122.8
Leoben-Göss	Steiermark	urban	Jun. 1995 -Jan. 2000	3	16.4	92.9	47.8	34.1	63.5	25.2
Graz-Marienhütte	Steiermark	urban	Feb. 1996 -Mar 1996	3	59.9	99.6	80.2	81.2	90.4	70.5
Graz-Händelstraße	Steiermark	urban		3	61.4	178.3	106.7	80.5	129.4	71.0
Raz-Peterstalstraße	Steiermark	urban		3	55.5	77.7	69.0	73.8	75.8	64.6

A1.2 SEDIMENT

There are no sediment data available from Austria.

A1.3 AIR

There are 155 results from air measurements available from Austria; most are from suburban or urban areas (Steiermark and Upper Austria); 12 from a rural area (Lower Austria)

(Table A 2). Sampling periods were from November 1992 until January 1998 (Umweltbundesamt 1993, Lorbeer *et al.* 1995, Thanner and Moche 1995a, 1995b, 1996a, 1996b, 1996c, Weis and Riss 1992, Weis 1998a, 1998b, 1998c). The lowest concentration measured was 1.3 fg I-TEQ/m³ in a suburban area, the highest at 587 fg I-TEQ/m³ in Graz (suburban) (Thanner and Moche 1996b). The media concentrations ranged from 24 fg I-TEQ/m³ to 317 fg I-TEQ/m³ (means: 26-314 fg I-TEQ/m³). The data as submitted did not allow to classify according to seasons.

A1.4 VEGETATION

In Austria, biomonitoring with fodder grass and Norway spruce needles was applied in the past. The results from 95 samples are summarised in Table A 3. It can be seen that in situations of local point sources, such as the copper smelter in Brixlegg, Tyrol, extremely high PCDD/PCDF concentrations were found in grass and spruce needles (Riss 1993, Riss *et al.* 1990). More data from the Brixlegg monitoring programme are discussed in chapter A1.5 in connection with the cow's milk data.

The results from the spruce needles indicate that the high concentrations in ambient air lasted for several years as, with increasing exposure time, the concentrations in the spruce needles increased. On the other hand, the monitoring performed in the suburban area of Amstetten, sample collection in October 1993, does not show the increase of dioxin concentrations with increasing exposure time. The concentrations in Amstetten were 20 to 40-times lower than the results from Brixlegg for comparable times of exposure. The concentrations of PCDD/PCDF in spruce needles from background stations across Austria are relatively low; an increase in concentration with the duration of exposure cannot be established as all concentrations are in a quite narrow range (0.3-1.9 ng I-TEQ/kg d.w.) (Riss *et al.* 1993).

Table A 3: Austria - Vegetation. Concentrations in ng I-TEQ/kg d.m.

Type Vegetation	Location	Exposure Time (years)	Date	n	Min	Max	Mean	Median
Fodder grass	Brixlegg, contamin.		5/88	5	13	23		
	Brixlegg, contamin.		5/92	11	0.8	104.6	16.7	2.4
	Brixlegg, contamin.		Since 1987	29	1.6	33		
Spruce needles (Norway spruce)	All Austria, backgr.	0.5	10/93	25	0.3	1.9	0.7	0.6
	All Austria, backgr.	1.5	10/93	5	0.3	0.9	0.6	0.6
	All Austria, backgr.	2.5	10/93	5	0.6	1.2	0.8	0.7
	Brixlegg, contamin.	1	Since 1989	8	7	87		
Spruce needles (Norway spruce)	Brixlegg, contamin.	0.5	10/87	1		51		
	Brixlegg, contamin.	1.5	10/87	1		55		
	Brixlegg, contamin.	2.5	10/87	1		75		
	Brixlegg, contamin.	3.5	10/87	1		86		
	Amstetten, suburban	1	5/93	1		5.4		
	Amstetten, suburban	2	5/93	1		2.7		
	Amstetten, suburban	3	5/93	1		2.8		
	Amstetten, suburban	4	5/93	1		2.1		

A1.5 COW'S MILK

In 1987, high PCDD/PCDF concentrations were detected in vegetation and cow's milk in the neighbourhood of a copper reclamation plant located at Brixlegg (Riss 1990, 1993). Ten samples of cow's milk, analysed in early 1988, had elevated concentrations from 14 to 69.5 pg I-TEQ/g fat. The data are displayed in the upper part of Table A 4. A monitoring programme was initiated around the copper plant with a special emphasis on the terrestrial food-chain grass • cow milk → man. PCDD/PCDF concentrations have been analysed in grass fed to dairy cow's as well as the cow's milk. The grass samples were analysed from a farm at the times of the harvest. The milk samples were taken at the end of the winter feeding period and collected from the tank of the farm (Riss 1993). The results for both matrices and the five year biomonitoring are displayed in the lower part of Table A 4. It can be seen that the concentrations decreased sharply in grass and slower in cow's milk. It took about five years to be close to background concentrations. After the dioxin concentrations in cow's milk had reached a level that would allow consumption of dairy products according to the German and the Dutch guidelines, the monitoring programme was terminated in Spring 1993.

Table A 4: Austria - Fodder grass and cow's milk from Brixlegg, reference time: 1987/88-1992/93. Concentrations in ng I-TEQ/kg dry mass or pg I-TEQ/g fat, respectively

Year	n	Grass ng I-TEQ/kg d.m.	Cow's Milk pg I-TEQ/g fat
02/88	6		21.1-69.5
04/88	4		14.0-28.3
05/88	5	13-23	
1987/88		33	49
1989/90		16	30
1990/91		5	22
1991/92		5	12
1992/93		4	5
8/1992		2.9	
5/1993		1.6	
Spring 1993			1.0-2.1

A1.6 SEWAGE SLUDGE

Dioxin results from 16 samples of municipal sewage sludge are available from Austria. The samples were generated during surveillance and obtained from across Austria. The concentrations ranged from 8.1 ng I-TEQ/kg d.m. to a 38.1 ng I-TEQ/kg d.m. with a median of 13 ng I-TEQ/kg d.m. (Table A 5) (Scharf *et al.* 1997), All concentrations are below the legal limit value of 100 ng I-TEQ/kg d.m. for use as fertiliser in agriculture (see Task 1).

Table A 5: Austria - Sewage sludge; sampling period: 11/94 until 03/95. Concentrations in ng I-TEQ/kg d.m.

	n	Min	Max	Mean	Median
Municipal sewage sludge	16	8.1	38.1	14.5	13.1

A2 Belgium

There is information regarding PCDD/PCDF ambient air, soil, and deposition from air in Belgium however there seems to be no information on concentrations in other media. The information identified is summarised below and is only from one of the three regions of Belgium; Flanders. No useful information has been found relating to the other two regions; Brussels and Wallonia.

A2.1 SOIL

PCDD/PCDF concentrations in soils were determined for the same six sites that were sampled for ambient air (de Fré and Wevers 1998). The study on the concentrations of PCDD/PCDF in soils (de Fré and Wevers 1998) gives concentrations in six locations covered in the study on ambient air (Wevers *et al.* 1993). The soil samples were collected in May-June 1992 from areas as undisturbed as possible. The results are shown in Table A 6. From the results a average figure for the rural locations in Flanders, Mol and Moerkerhe, was 2.21 ng/kg I-TEQ. As the urban/industrial locations were so different it would not be representative to give an average for these areas; nevertheless, the numbers are low for urban areas in a densely populated and industrialised country.

Table A 6: Belgium - Soil (ng I-TEQ/kg d.m.)

	Conc. (ng I-TEQ/kg d.m.)
Mol	2.14 ±0.24
Moerkerhe	2.27 ±0.06
Berendrecht	3.18 ±0.57
Zelzate	8.94 ±1.06
Ham	2.72 ±0.20
Vilvoorde	5.76 ±0.21

A2.2 AIR

A1.1.1 Ambient Air

Measurement of the concentration of PCDD/PCDFs in ambient air in Belgium has been studied at selected locations in Flanders (Wevers *et al.* 1993). A description of the six sampling sites is shown below. Sampling times were 3-times 24 hours. Two different sampling devices have been utilised: A commercial high volume sampler (from Ströhlein) capable to collect particles on a preconditioned glass fiber filter and a two-phase sampler equipped with filter and a polyurethane foam. The commercial sampler was set to a sampling rate of 75 m³/h and the two-phase sampler at 24 m³/h.

The average PCDD/PCDF concentrations for the six sampling sites are shown in Table A 7.

Sampling Site	Site Description.
Mol	A rural location, further characterised by a coal fired power station and some nuclear industry at 1-4 km east of the sampling site.
Berendrecht	This site is situated in the Antwerp harbour area, with industry stretching out 2-15km SE to SW from the sampling site and the city of Antwerp about 15 km SE.
Zelzate	This site is situated in the vicinity of a metallurgical and chemical industry at the Gent-Terneuzencannal canal with a highway at 1-15 km SW from the sampling site and the city of Gent at 20 km SW.
Moerkerke	A rural location with a large municipal waste incinerator (175,000 ton/yr.) about 10 km west of the sampling site as the nearest known source.
Vilvoorde	Situated about 15 km NE from the Brussels metropolitan centre, characterised in addition by a coal fire power station at 1 km W and industry extending 1-7 km SW-W, including a MWI at about 6 km SW from the sampling site.
Ham	A location characterised by a chemical industry and a highway at 2-7 km SE to W from the sampling site.

Table A 7: Belgium - Ambient air (fg I-TEQ/m³)

Sampling Location	High Volume sampler 75 m ³ /h (fg TEQ/m ³)	2-Phase Sampler 24 m ³ /h + vapour phase (fg TEQ/m ³)
Mol	99.9 ± 66.7	125 ± 72.7
Berendrecht	111 ± 46.1	86.2 ± 24.7
Zelzate	173 ± 18.2	118 ± 41.7
Moerkerke	121 ± 9.8	69.6 ± 10.9
Vilvoorde	214 ± 136	115 ± 89.5
Ham	254 ± 118	129 ± 167

The result from the commercial air sampler showed that the average PCDD/PCDF concentration at typical rural locations in the study is 100 fg TEQ/m³. However as the sampling took place at different times it is not very representative. The differences between the sites were not as large as the differences found between days at each site suggesting that the major influence on concentration is meteorology rather than sources in the immediate vicinity. The comparison of the two samplers would suggest that the PCDD/Fs were predominately associated with the particle phase.

The impact of automobile emissions was studied in a tunnel (Wevers and de Fré 1992). In this study, which was carried out near Antwerp, four background samples were taken around the vicinity of the tunnel however their locations are not included in the study. The results for the background concentrations around the tunnel are shown Table A 8 for both the samplers used simultaneously in the study.

Table A 8: Belgium - Tunnel air (fg I-TEQ/m³)

Site type	Filter + PUF Sampler (fg TEQ/m ³)	Filter Sampler (fg TEQ/m ³)
Background	21	/
Background	55	71
Background	24	/
Background	35	45
Mean	35 ± 18	58 ± 19

The means for the background measurements were 35 fg TEQ/m³ and 58 fg TEQ/m³ for the particulate sampler and the particulate/vapour sampler respectively. These concentrations cannot be considered as being representative of the background concentrations of PCDD/PCDF in Belgium ambient air.

A1.1.2 PCDD/PCDF in Deposition

PCDD/PCDF in deposition were determined for locations in Flanders (de Fré and Wevers 1998). Deposition measurements using Bergerhoff gauges have been carried out in Flanders since 1993 on behalf of the VMM (Vlaamse Milieumaatschappij), the agency in charge of environmental monitoring. In the study the majority of the sampling sites are positioned where specific PCDD and PCDF sources are located however some sampling sites have a mix of urban and background locations. The results are summarised in Table A 9 together with the sampling time and the type of the site.

Table A 9: Belgium - Deposition samples in Flanders

Site Name	Site Type	Date of Sampling	ng TEQ/m ² ·a	pg TEQ/m ² ·d
Eksel	Background	04/97-05/97	1.13	3.1
Mol	Background	04/97-05/97	0.26	0.7
Merksem	Urban	08/96-09/96	4.39	12.0
Antwerpen	Urban	04/97-05/97	0.33	0.9

The results for Merksem are not comparable with the other data for 1997 as the sampling period was not only in a different year but also at a different time of year.

A2 Denmark

In 1997, the Danish Environmental Protection Agency published a Working Paper (Ministry of Environment and Energy 1997) on the status of dioxin emissions and concentrations of these substances in the environment. An exposure assessment was performed as well (see Task 4 for dietary intake and Task 6 for human levels). The present report is an update of a previous report published in 1995 (in Danish only). Since then only a few new original Danish dioxin investigations have been conducted. The new data is mainly based on very few measurements (spot tests) of a few items, such as wood stoves, biofuels, sewage sludge, and textiles. Therefore, there is still a rather limited database on which to base assessment of sources and exposure to PCDD/PCDF in Denmark. The present database does not include Greenland and the Faroe islands. An update – including these areas – is planned for the year 2001.

A further draw-back of the data is that most of the Danish investigations were homologue-specific with low-resolution mass spectrometry (LRMS), which may be sufficient for samples with high dioxin concentrations but not for low concentrations.

Here, we summarise the results obtained for environmental matrices.

A2.1 SOIL

There is one old Danish investigation of PCDD/PCDF in soils but it is outdated. At the time of the investigation, there was no congener-specific analysis performed. Background levels of dioxins in Danish topsoil (9 cm of depth) from different areas (forest and agricultural land) have been estimated to between 51 and 515 ng.

A small, not published, Danish investigation from a locality in Jutland, where cable scrap had been illegally burned, showed concentrations of 25,700 ng PCDD+PCDF/kg d.m. This concentration was about fifty times background levels. Only hepta- and octa-congeners were analysed (Wrang and Worsøe 1991).

Denmark recognises a need for new investigations, since it is impossible to assess the possible consequences of applying sewage sludge and compost contaminated with dioxins to agricultural soils, without knowing the background concentration of dioxins in these soils.

A2.2 SEWAGE SLUDGE

There were a few sewage sludge samples analysed for PCDD/PCDF. The results for a single sludge sample from each of three waste water plants gave 10, 16, and 36 ng I-TEQ/kg d.m. (mean: 21 ng I-TEQ/kg d.m.). The authors found a pattern similar to pentachlorophenol contamination (Grüttner *et al.* 1996)

In a later study, 35 more municipal sewage sludge samples were analysed. The PCDD/PCDF concentrations were in the range of 0.7-54.7 ng N-TEQ/kg d.m. The average of all 38 samples was 9.5 ng N-TEQ/kg d.m. (Vikelsøe, personal information, 1996).

The total amount of PCDD/PCDF contained in Danish sewage sludges – annual production of about 925,000 ton f.w. or 170,000 ton d.m. – is around 1.7 ng I-TEQ/kg d.m.

A2.3 VEGETATION, WILDLIFE, AIR, AQUATIC ENVIRONMENTS, COMPOST

There are no Danish studies of PCDD/PCDF in vegetation, wildlife, air, the aquatic environment, municipal or garden compost.

A2.4 COMMODITY PRODUCTS

There was one study on commodity goods in Denmark: the analysis of 24 T-shirts gave concentrations from 0.02 to 2.6 ng N-TEQ/kg with an average of 0.35 ng N-TEQ/kg (Vikelsøe and Johansen 1996). The levels in the samples ranged from 0.02-2.6 ng N-TEQ/kg. In general, the findings of the study correspond to the findings in the German studies, although the few samples with very high contamination were not identified.

A3 Finland

There is a large quantity of data available concerning environmental levels of PCDD/PCDF in Finland. In particular there is detailed coverage of levels in sediments, fish and some other wildlife.

Sediments have been studied in detail because of the problem of historical contamination by the chemical industry, and the continuing risks linked to the re-mobilisation of the sediments. Fish are also studied in detail, because of the dominance of fish in the diet of the Finns, and as indicators of contamination through the aquatic system.

Information on soils and vegetation has also been included below for completeness, although the data is sparse.

A3.1 SOURCES OF CONTAMINATION

The widespread use of wood preservatives in the forestry industry and also chlorine-based bleaching processes in the pulp and paper industry have left large areas of contamination across the country. The use of 'Ky-5', a commercially available fungicide and wood preservative, is reported. This chemical is a mixture of 2,3,4,6-tetrachlorophenol (2,3,4,6-TeCP), pentachlorophenol (PCP) and 2,4,6-trichlorophenol (2,4,6-TCP). It was used from the 1940s until its ban in 1984. The contamination by PCDD/PCDF in the various fungicides and their derivatives depends on the nature of production processes and conditions.

Assmuth and Vartiainen (1994) have described the mechanisms of contamination by this type of chemical. Congeners of PCDD/PCDF are readily formed from 2,4,5-trichlorophenol (2,4,5-TCP) and its derivatives which have commonly been used in herbicides. Contamination by 2,3,7,8-Cl₄DD has been observed at production facilities for such compounds as a result of both 'normal' emissions and accidents, such as at Seveso.

Through natural transport processes these contaminants in effluents and runoff have been distributed through the aquatic environment, and have been deposited in sediments in rivers and the sea. Contamination of sediments in some instances is severe that the sediment contains levels of PCDD/PCDF equivalent to that of hazardous waste. Although many of the sources of pollution no longer exist, the contaminants continue to pose a risk in the present day, as the sediments can be disturbed and re-mobilised and the PCDD/PCDF again become available for uptake by fish and other aquatic life.

A3.2 SOILS

High concentrations of PCDD/PCDF have been found in soil samples from sites where wood preservatives have been used (Assmuth and Vartiainen 1994; 1995). Three such sites were analysed, along with samples of waste, leachate and runoff from municipal mixed waste sites for comparison.

The maximum concentration found in the soils at those sites contaminated with wood preservatives was over 90,000 ng I-TEQ/kg d.m., and the mean value for all soil on these sites was 19,000 ng TEQ/kg (Assmuth and Vartiainen 1995). The congener profile of these samples was similar to that of the wood preservative, with the three most abundant congeners being 1,2,3,4,7,8-Cl₆DF, 1,2,3,4,6,7,8-Cl₇DF and Cl₈DF. The lowest concentrations, at 9.1 ng I-TEQ/kg, were found in the topsoil, possibly because of the mechanisms of relocation and decay of the wood preservative compounds. No relationship was found between concentrations of chlorophenol and PCDD/PCDF, resulting from the different behaviour of these compounds in sediment. Chlorophenols are more water-soluble than PCDD/Fs and are therefore more mobile and do not remain in the surface layers. The concentration found in the municipal waste was 50 ng I-TEQ/kg d.m.

The study by Assmuth and Vartiainen (1995) also considered the effects of this contamination on the levels of PCDD/PCDF in fish. These results are describe in the section below on fish.

Much higher concentrations have also been found in other areas of contamination. Surface layers (0-20 cm) of soils at sites where wood preservative had been used, sampled by Sandell and Tuominen (1993) (cited in IARC 1997), were found to contain concentrations of 1.7-85 µg I-TEQ/kg d.m. (1,700 to 85,000 g I-TEQ/kg d.m.). There has not been any analysis of background concentrations of PCDD/PCDF in Finnish soils.

A3.3 SEDIMENTS

As has already been mentioned, there is a substantial amount of data available relating to concentrations of PCDD/PCDF in sediments in Finland. The sediments analysed range from those in isolated lakes in Lapland, to those in a major river contaminated over many decades by chemical industries. A summary of the data available in published documents is shown in Table A 10. The table firstly includes those sediments considered to be in 'background' locations and reference sites chosen for comparison with contaminated sites. In these samples the concentrations of PCDD/PCDF range from 0.71 to 100 ng I-TEQ/kg d.m. The second section of the table contains details of the concentrations found in areas of contamination, with details of the known or suspected sources of contamination. In these samples the concentrations range from 3.37 to 80,000 ng I-TEQ/kg d.m. The majority of studies has focused on areas of contamination.

A3.3.1 Lake Sediments

The work of the Arctic Monitoring and Assessment Programme (AMAP) has included monitoring of lakes in uninhabited areas, in order to assess background concentrations of PCDD/PCDF. The AMAP report (1998) gives a broad description of analyses in Arctic lakes (AMAP 1998). The PCDD/PCDF profile was dominated by Cl₈DD. Concentrations of 2,3,7,8-Cl₄DD/Cl₄DF were low or non-detectable in most of the sediment samples, however, other non-2,3,7,8-substituted tetrachlorinated dibenzodioxins and dibenzofurans were present. This high proportion of Cl₈DD in all Arctic lake sediments suggested that the major source of PCDD/Fs is combustion related (AMAP 1998). Concentrations in Finnish lakes were similar to those found in Sweden, and similar to ‘background’ concentrations in some other areas of Europe such as Lake Ladoga in Russia.

The top layers of the sediment cores of Lake Pahtajärvi, Lake 222 and Lake Sierram were analysed in winter 1994 (Vartiainen *et al.* 1997). The lakes are relatively undisturbed catchments in Lapland, receiving atmospheric inputs of contaminants, showing historical inputs into the Arctic environment (AMAP 1997). Current concentrations in the sediments were found to range from 1.4 to 4.2 ng I-TEQ/kg d.m. The sections of the cores that correspond to sediments laid down in 1900 in Lakes Pahtajarvi and Sierram had estimated concentrations of 0.7 and 0.9 ng I-TEQ/kg d.m. However, more detailed time-trends data provided in AMAP (1997) show that there has been a recent decline in PCDD/PCDF deposition in Lake Pahtajärvi, following major increases in the 1940s. The contamination was considered to originate from a variety of sources: air deposition, chemical and paper industry discharges and wide use of the fungicide Ky-5. The pre-industrial contribution of PCDD/PCDF, presumably due to combustion sources such as forest fires and wood burning, is evident in these Finnish cores, particularly Lake 222 (Vartiainen *et al.* 1997).

Further evidence of a recent decline in concentrations of PCDD/PCDF has been presented by Vartiainen *et al.* (1995). Two sediment cores from Lake Valkjärvi near to Kärkölä in Southern Finland were analysed. The catchment of this lake has been associated with chlorophenol contamination from a saw mill of Kärkölä, as reported by Lampi *et al.* (1990). The study aimed to determine whether this contamination was accompanied by contamination with PCDD/PCDF. The sediment data shows low traces of contamination by Ky-5, and it was found that the maximum concentrations of PCDD/PCDF occurred in sediments that were laid down in the years 1960-70, at concentrations of 1.01 ng I-TEQ/kg d.m. and 1.14 ng I-TEQ/kg d.m. in the two separate cores. These concentrations were considered to be similar to those in background locations in Finland, and lower than in rural areas in other countries. Fish samples were also collected from this location (see section A3.4.1).

Geographical variations in concentrations across space have also been studied (Koistinen *et al.*, 1990). Analysis was carried out to establish whether contaminants in lake sediments could be linked to pulp mill effluents. The 18 samples were all from a lake system in Central Finland, about 150 km in length south of Äänekoski, and downstream from a pulp mill, taken in 1988/89. Concentrations in the sediments were relatively low, and ranged from below the limit of detection to 13 pg I-TEQ/g d.m. The congener 2,3,7,8-Cl₄DD was not detected in any of the samples, but other Cl₄DD were evident and showed a clear gradient downwards from the pulp mill effluent. Other 2,3,7,8-substituted compounds were present, but not in this pattern, and were therefore concluded to be not of pulp mill origin. Overall the PCDD/PCDF

congener profile was described as more “background” than “bleaching”, indicating that pollution from this mill had been very low in recent times.

A3.3.2 River Sediments - The Kymijoki River

The Kymijoki River is the fourth largest river in Finland, and its catchment has a long history of pulp and paper production and other chemical industries, such as the manufacture of Ky-5 wood preservative from 1940 to 1984. It has been estimated that up to several kilograms of PCDD/PCDF have been released into the river over a 40 year period, originating from production equipment cleaning and one accidental fire (Verta *et al.* 1997).

There have been improvements in water quality over the last 15 years, but the river sediments still retain high levels of contamination, which is re-released into the aquatic system and there is therefore a continuing risk to the ecosystem and human health.

High concentrations of PCDD/PCDF were first reported by Koistinen *et al.* (1995b). A joint research project was later initiated in order to examine the extent of the contamination. This study considered the occurrence and impacts of organochlorine compounds and heavy metals in the River Kymijoki, as described by Verta *et al.* (1997). Broadly, the aims of the research project were to survey the extent of contamination in the river; to study the mechanisms of fate, transport and toxicology of the contamination within the ecosystem and in relation to human health; and to assess the preconditions for restoration of the ecosystem. In particular there have been many studies on levels of contaminants in sediments and fish. PCDD/PCDF were among many chemicals under investigation, and in particular PCDE were of interest, to assess their relative toxicity compared to PCB and PCDD/PCDF.

Sediment samples were collected along the Kymijoki River and its estuary in a number of different studies (Koistinen *et al.* 1995b and 1997b; Verta *et al.* 1997). Verta *et al.* (1997) described river sediment samples collected from 16 locations along the Kymijoki River during the spring of 1996. At least two cores from each location were analysed. As found by Koistinen *et al.* (1990), there was a pattern of decreasing concentrations downstream. The highest I-TEQ results were found in the upper course of the river, where there were concentrations of 0.006 to 0.08 mg I-TEQ/kg d.m. (6,000-80,000 ng I-TEQ/kg d.m.). On the lower course the concentrations were 0.0005-0.0035 mg I-TEQ/kg dry matter (500-3,500 pg I-TEQ/g d.m.). Highly chlorinated (hepta- and octa-) congeners, typical of Ky-5, dominated in river sediments. The highest PCDD/PCDF concentrations were up to two orders of magnitude higher than the limit values proposed for contaminated soils.

Very high levels of contamination were also found by Koistinen *et al.* (1995b). Samples were collected from one location upstream of a source of contamination (reference site) and two downstream. Concentrations of PCDD/PCDF in the river sediments were so high that the sediment should be treated as hazardous waste, with up to 59,000 ng I-TEQ/kg d.m. The limit for a hazardous waste classification is 1 ng I-TEQ/g dry waste. The congener profile was very similar to that of Ky-5, a wood preservative that used to be manufactured at a plant on this river. Concentrations in sediments at a location upstream of this site were much lower (100 ng I-TEQ/kg d.m.), although this is still considerably higher than those concentrations found at background locations in the Arctic.

A3.3.3 Sea Sediments

Koistinen *et al.* (1995b) also analysed the concentrations of PCDD/PCDF in sediment samples from Bothnian Bay, from two reference locations and two locations near to a pulp mill. The concentrations in sediments from the Bothnian Bay were lower than those in the Kymijoki River described above, ranging from 38 to 63 ng I-TEQ/kg d.m. in the reference locations, and 270 to 350 in the area close to the pulp mill. Congener profiles were different from those found in the Kymijoki River. This study also considered levels of PCDE, which are also associated with chlorobleaching and Ky-5 contamination.

Koistinen *et al.* (1997b) analysed sediments obtained from the Finnish Institute of Marine Research, which had been collected in 1993 from the Gulf of Finland and Gotland Deep in the Baltic Proper. The top 30 mm from the sediment cores were divided into 10 mm slices for analysis of sedimentation over roughly the last 30 years. A pattern of reduction over time was found in the sediment, showing more clearly in the Baltic Proper sample (concentrations of 27, 36 and 53 ng I-TEQ/kg d.m. at increasing depths 0-1, 1-2 and 2-3 cm respectively) than in the Gulf of Finland (concentrations of 20, 24 and 24 ng I-TEQ/kg d.m. at increasing depths 0-1, 1-2 and 2-3 cm respectively). The high concentrations of Cl₈DF in sediments in the Gulf of Finland are typical of contamination by PCP or other chemical wastes (like Ky-5), and the dominance of Cl₈DD in the sediments of Gotland is typical of those influenced by atmospheric transport from combustion sources.

A3.4 WILDLIFE

Data are available for a large number of fish, and these are considered separately in the next main section of this report. There are also a few data available on other species of wildlife, which are detailed in Table A 11. There are data for the eggs of Black Guillemot, two types of seal and White-tailed sea eagles.

White-tailed sea eagles are predators at the top of the Baltic food chain, and organic contaminants of many types can be found in their tissues. Koistinen *et al.* (1995a) analysed the comparative toxic loading by a variety of contaminants, including PCDD/PCDF. Of particular interest to the study were polychlorinated diphenyl ethers (PCDE), as these had not been studied before in these birds. The eagles live on a very varied diet, and therefore the levels of contaminants varied considerably between samples.

The eggs of black guillemots were also studied. These are one trophic level below the eagles, but live on a much less varied diet, of just eelpouts. They show very constant levels of contamination across samples. The guillemot would therefore make a useful species for environmental monitoring (see Task 7 - Ecotoxicology).

Three white-tailed sea eagles were found dead in the Gulf of Bothnia between 1988 and 1991. The breast muscle was analysed. Eggs of black guillemots were found that had failed to hatch, and these were collected from nests. The I-TEQ concentrations varied from 830 to 66,000 pg I-TEQ/g fat in eagles, and 1,500 to 1,700 pg I-TEQ/g fat in black guillemots. The predominant congeners in guillemot were 1,2,3,7,8-Cl₅DD and 1,2,3,6,7,8-Cl₆DD, but in eagles 2,3,4,7,8-Cl₅DF dominated. The pattern in eagles was similar to that seen in Swedish osprey and in Baltic fish. Similar concentrations in guillemots have been reported in Sweden.

It should be noted, however, that PCB constituted by far the largest proportion of the toxic equivalent with 87-98% of the total I-TEQ.

Koistinen *et al.* (1997b) compared PCDE levels with PCDD/PCDF in seals and sediments to discover if there was a reason for high levels of mortality in ringed seals in the Gulf of Finland in 1991. Fourteen ringed seals and six grey seals were collected having been found dead in the Gulf of Finland, mostly in the winter of 1991-2. The seals varied widely in age and health status.

Detection limits for PCDD/PCDF were quite high in comparison with other studies, because of problems with instruments and sample size, but it was concluded they would be sufficient for this study. Concentrations in seals were dominated by 2,3,7,8-Cl₄DD, 1,2,3,7,8-Cl₅DD, 1,2,3,6,7,8-Cl₆DD, 2,3,7,8-Cl₄DF and 2,3,4,7,8-Cl₅DF.

Low concentrations were found in juveniles just weaned (with a mean of 12 pg I-TEQ/g f.w., and the highest concentration was found in starved old (18 yrs) female (150 pg I-TEQ/g f.w. These concentrations are similar to those found in Lake Saimaa by Koistinen *et al.* (1997b) where the median concentration of four ringed seals was 105 pg I-TEQ/g fat. The results also coincide with those of a Swedish study (Bergek *et al.* (1992). The study found that the ringed seals had been exposed to a greater toxicity of PCDD/PCDF than the grey seals but this was not enough to explain the greater mortality of the ringed seals in the Gulf of Finland. Differences between species in the Gulf of Finland were suggested to be a result of diet, but this has not been proven.

AMAP (1997) reports that information on levels of PCDD/PCDF in Arctic marine biota is limited. The greater complexity and higher cost of analysis of these compounds has limited the number of samples analysed to date. Concentrations in Arctic seals are lower than in animals from the Baltic and the North Sea, but higher than those found in Antarctic seals, presumably because of the generally higher levels of pollution in the Northern Hemisphere in comparison with the Southern Hemisphere.

A3.4.1 Fish

Various species of fish have been used as monitoring species, to study variations in concentrations of contaminants in the environment, and other data has been collected for the purpose of human exposure calculations. The latter data have been included here for completeness, but will also be considered in Task 4 - Human Exposure. Concentrations of PCDD/PCDF are given in N-TEQ, with fat adjusted concentrations in bold text, and fresh weight concentrations in normal text. The results are grouped by species of fish (Table A 12).

A3.4.1.1 Fish in the Arctic

Three subarctic headwater lakes in Lapland have been monitored as part of AMAP (Vartiainen *et al.* 1996). Fish were caught during the winters of 1993 and 1994. PCDD/F concentrations in Arctic Char were considered to be extremely low, on average 0.06 pg I-TEQ/g f.w. in both Lake Pahtajärvi and Lake 222. Lake Nitsijärvi had higher concentrations, with a mean of 0.1 pg I-TEQ/g f.w. Concentrations were about one tenth those found in Lake Valkjärvi (see below), and about two orders of magnitude lower than the highest concentrations in the Baltic Sea or the Gulf of Finland (see below). Concentrations in

burbot liver were also analysed, and the concentrations found were much lower than those found in the south of Finland (Korhonen *et al.* 1997), at 2.76 and 0.7 pg I-TEQ/g f.w. in comparison with 122 and 82.4 on the Kymijoki River. PCDD/PCDF concentrations are relatively high in Burbot liver compared to other fish tissues. This can be explained by the fact that burbot is a predatory bottom-feeding fish, and has a high fat content particularly in the liver. This makes this species suitable for monitoring lipophilic pollutants. In Scandinavia, TEQ due to co-planar and *mono-ortho*-substituted PCB were much greater than those due to PCDD/PCDF (AMAP 1997).

PCDD/PCDF in freshwater fish in the Arctic are low (typically < 1 pg/g) in comparison to concentrations in fish sampled near bleached kraft mills or to species in the Baltic Sea (AMAP 1997). For example, in a comparison between Baltic Salmon in the Arctic Tenjoki River and the Baltic Sea (Vuorinen *et al.* (1997a), the concentrations, on a fresh weight basis, in the Arctic were found to be statistically significantly lower ($p < 0.05$) than in the Baltic Sea. However, owing to the differences in size of the fish studied, the difference in concentration was not significant on a fat basis.

A3.4.1.2 The River Kymijoki and its Estuary

PCDD/PCDF concentrations in three pike samples from the Kymijoki River and five from Bothnian Bay (Koistinen *et al.* 1995b) showed typical congener patterns for chlorobleaching of pulp. Concentrations in the pike in the Bothnian Bay were low in comparison to other measured concentrations in Finland and Sweden. Estimated I-TEQ concentrations were 0.32-0.62 pg I-TEQ/g f.w., whereas concentrations in the Kymijoki River were 0.36-0.97 pg I-TEQ/g f.w. Concentrations in the pike caught upstream of the pulp mill at Kuusankoski were lower than those caught downstream, indicating the source of the contamination.

A further study of the fish in the Kymijoki River and its estuary was undertaken in 1996. Fish were caught by local fishermen in 19 localities along the water course (Korhonen *et al.* 1997, Verta *et al.* 1997). The fish collected were burbot, perch, pike, pike perch, salmon and bream. Fresh weight I-TEQ concentrations were low in all locations, even near to the previous pollution source, in all samples except salmon. Fat adjusted concentrations in salmon were similar to the other types of fish, but the fat content in salmon is high, at 7% compared with 0.3-1.3% in others, resulting in a higher fresh weight concentration. Concentrations were also high in liver of burbot and spawn of bream for similar reasons. The concentrations in the river were again found to be higher than in the estuary.

Thirty caged freshwater mussels were incubated for four weeks in the summer of 1995 in River Kymijoki and Lake Vanaja in south Finland, which are both polluted by industrial effluents such as from wood preservative production and paper and pulp mills (Koistinen *et al.* 1997a). Fifteen mussels were analysed for PCDD/PCDF in each location. The study was part of an ongoing monitoring of mussels by the Finnish Environment Institute, since 1986. Concentrations of PCDD/F were much higher in mussels from the River Kymijoki than from those in Lake Vanaja, with mean concentrations of 210 pg I-TEQ/g fat compared to 61 pg I-TEQ/g fat, respectively. The pattern of congeners in River Kymijoki was indicative of the wood preservative source, with very high concentrations of 1,2,3,4,6,7,8-Cl₇DF and Cl₈DF. Similar patterns had also been found in sediments. It is likely that the contamination had been released from the sediments.

A different congener profile was found in the mussels from Lake Vanaja and therefore the nature of the source is likely to be different.

A3.4.1.3 Simojoki River

Vourinen *et al.* (1997b) investigated the relationship between concentrations of several organochlorine compounds in the muscle of female Baltic salmon from the Simojoki River and reproductive defects and mortality (M74 syndrome). Sampling occurred every year from 1989 to 1993, with samples caught and muscle taken from 73 fish in total in this time. 15 PCDD/F congeners were analysed, but data are only available for 3 of these (2,3,7,8-Cl₄DF, 1,2,3,7,8-Cl₅DF and 2,3,4,7,8-Cl₅DF, concentrations of the latter being the highest). I-TEQ concentrations were resolved graphically and range from ~75 pg I-TEQ/g fat to ~200 pg I-TEQ/g fat. These compare well with the results found by Koistinen *et al.* (1995b) in the Kymijoki River. Concentrations for I-TEQ originating from PCB were considerably higher, and make up the large proportion of the total I-TEQ.

A3.4.1.4 Marine Fish

The Finnish Environmental Institute are undertaking a monitoring programme measuring concentrations of PCDD/PCDF in coastal waters. Part of this has been an analysis of concentrations of PCDD and PCDF in Baltic Herring and Northern Pike in Finnish coastal areas (Korhonen and Vartiainen 1997). The monitoring sites were mainly far from local pollution, but two were in the estuaries of the rivers Kymijoki and Kokemäenjoki, which are contaminated with agricultural, waste water and industrial pollution.

Baltic Herring were sampled in autumn 1990 and 1993 and pike were sampled in spring 1989 and 1992. Numerical data were not published, and they therefore do not appear in Table A 12. However, a graphical comparison of fresh weight concentrations shows concentrations ranging from ~0.25–3.6 pg I-TEQ/g f.w. for Baltic Herring, and ~0.05–0.8 pg I-TEQ/g f.w. for pike. Herring is a much fattier fish (5%) than pike (0.4%). The highest concentrations were found in the Gulf of Finland, and the lower concentrations were found further north in the Gulf of Bothnia and the Bay of Bothnia.

A3.4.1.5 Freshwater Lakes

The occurrence of PCDD/PCDF in fish in Lake Valkjärvi in Southern Finland near to Kärkölä have been analysed (Vartiainen *et al.* 1995). The lake has been found previously to be contaminated by chlorophenols from the 1970s (Lampi *et al.* 1990), and this study aimed to discover if current contamination in fish could be attributed to the same source. Sediment cores were also analysed (see section xx).

A variety of fish were caught in 1988 and 1994. Concentrations were found to be low, at 0.21-0.67 pg I-TEQ/g f.w., and at the same concentration as other fish from inland waters in Finland. The paper concludes that PCDD/PCDF concentrations were low in comparison to those expected if the contamination was linked with the chlorophenol pollution.

Koistinen *et al.* (1989) undertook a study to consider the relative influences of local leakages and atmospheric transport of contaminants on pollution in water systems in the Baltic and in Lake Kernaala. Lake Kernaala is contaminated by PCB and derivatives. PCDD/PCDF were

among a large group of compounds studied. Samples of salmon, pike, walleye and cod samples were taken in the period 1986-88 from the Gulf of Finland, Gulf of Bothnia and Baltic Proper. Pike and walleye were sampled from Lake Kernaala; and cod from the Arctic coast of Norway (cod livers analysed).

The analysis found that the occurrence of PCDD and PCDF was low or non-detectable (< 5 pg/g) in all samples. Concentrations of 2,3,7,8-Cl₄DF were detected only in the pike and walleye in Lake Kernaala, (Table A 12). The I-TEQ concentrations must be far below 12 pg/g, which is the value calculated if non-quantifiable congeners were assumed to be present at the limit of quantification (5 pg/g). This was very early analysis and, therefore, relatively insensitive, and PCDD/PCDF were only found in areas of contamination.

A3.5 VEGETATION

No data on concentrations of PCDD/PCDF in vegetation are available for background locations in Finland. One study has been undertaken in order to assess the effects of changing combustion processes at an industrial location. The needles of coniferous trees were collected in August 1996 from eight sites near to a forest industry plant on the coast of the Gulf of Bothnia (Sinkkonen *et al.* 1997). One location 44 km away was also sampled as a reference. Pulp and paper are produced at the plant, historically using chlorine bleaching, but now using oxygen and chlorine dioxide. Waste from the plant, and municipal waste is also burned. Individual I-TEQ concentration data are not available. However, in all nine samples the concentrations of 2,3,7,8-substituted PCDD congeners except for Cl₅DD were below the limit of determination (0.08 pg/g). Of the PCDF, Cl₆DF was the most dominant. An influence of the plant on the concentrations could not be identified.

A3.6 DRINKING WATER

High concentrations of total chlorophenols were found in drinking water and in ground water close to a sawmill in southern Finland. Exposure of the population to PCDD/PCDF could not be ruled out. However, no increased concentrations in mothers milk were found in the population who had used the contaminated water (Lampi *et al.* 1990).

Task 2 – Technical Annex

Table A 10: Finland - Sediments

Location	Type of location/Source of contamination	Layer of core (cm) (approx. date)	Date of Analysis	n	Mean Concentration (ng I-TEQ/g d.m.) Range in brackets	Ref.
Background and reference sites						
Lake 222, Lapland	Uninhabited area	Surface (1994)	1994	2-4 per lake	1.4	1
Lake Pahtajarvi, Lapland	Uninhabited area	Surface (1994)	1994	2-4 per lake	4.2	1
Lake Sierram, Lapland	Uninhabited area	Surface (1994)	1994	2-4 per lake	4.1	1
Lake Pahtajarvi, Lapland	Uninhabited area	1900 level	1994	2-4 per lake	0.7	1
Lake Sierram, Lapland	Uninhabited area	1900 level	1994	2-4 per lake	0.9	1
Freshwater, isolated from Bothnian Bay	Reference location	3-6 cm	1993	1	48	2
Bothnian Bay	Reference location	0-3 cm	1993	1	63	2
Bothnian Bay	Reference location	18-22 cm	1993	1	38	2
River Kymijoki, at Pilkanmaa	Reference location	surface	1993	1	100	2
Contaminated sites						
Lakes in Central Finland	Downstream of pulp mill	surface	1990?	18	3.4 (0-13) median 0.7 ^a	3
Lake Valkjarvi – estuary zone	Area of chlorophenol contamin.	14-18 cm (1960s)	1992	1	1.0	4
Lake Valkjarvi – deep zone	Area of chlorophenol contamin.	6 cm (1960s)	1992	1	1.1	4
Bothnian Bay	Close to pulp mill	0-3 cm	1993	1	350	2
Bothnian Bay	Close to pulp mill	9-12 cm	1993	1	270	2
Baltic Proper; near Gotland	Impact from combustion sources	0-1, 1-2 and 2-3	1993	3	39	5
Gulf of Finland	PCP or other chemical waste	0-1, 1-2 and 2-3	1993	3	23	5
River Kymijoki, at Korja	Downstream pulp mill, Ky-5 production site	surface	1993	1	59000	2
River Kymijoki, at Myllykoski	Downstream pulp mill, Ky-5 production site	surface	1993	1	6000	2
Lower course of the River Kymijoki	Historic inputs from pulp mill, chemical industry	0-3 and 0-6	1996	8	(500-3500)	6
Upper course of the River Kymijoki	Historic inputs from pulp mill, chemical industry	0-3 and 0-6	1996	6	(6000-80000)	6

n = number of samples; ^a = using the Nordic I-TEQ system;

References: 1. Vartiainen *et al.* (1997); 2. Koistinen *et al.* (1995b); 3. Koistinen *et al.* (1990); 4. Vartiainen *et al.* (1995); 5. Koistinen *et al.* (1997b); 6. Verta *et al.* (1997).

Table A 11: Finland - Wildlife

Species	Location	Type of Location	Sampling method	Date	n	Mean Concentration (pg I-TEQ/g lipid)	Ref.
Black Guillemot egg	Quarken area of the Gulf of Bothnia	Background	eggs failed to hatch, collected from nests	1985	3	1600; 1500; 1700 mean: 1600	1
White-tailed sea eagle	Quarken area of the Gulf of Bothnia	Background	found dead	1988	1	6900	1
				1990	1	66000	1
				1991	1	830	1
Ringed Seal	Lake Saimaa	unknown	found dead	1988	4	142.5 (100-260) median 105	2
Grey Seal	Gulf of Finland	Potentially contaminated	“ ”	winter 1991/92	6	32 (12-61)	3
Ringed Seal	Gulf of Finland	“ ”	“ ”	winter 1991/92	14	70 (45-150)	3

n = number of samples

1. Koistinen *et al.* (1995a); 2. Koistinen *et al.* (1995b); 3. Koistinen *et al.* (1997b).

Note: The shown in this table are not true arithmetic means because some samples were grouped for reporting, and therefore the data shown are means of means.

Table A 12: Finland - Fish

Species, Location	Date	n	Mean Conc. (pg I-TEQ/g) (range)	Mean % fat	Comments	Ref.
Arctic Char						
Lake Pahtajarvi	1993-94	10	0.06		background location	1
Lake 222	1993-94	12	0.06		background location	1
Lake Nitsijarvi	1993-94	6	0.1		background location	1
Baltic Herring						
Unstated source	ns	10	0.94 (0.64-1.9) 30.2 N-TEQ			2
Baltic sea	ns	ns	9.32			3
Baltic Salmon						
Baltic Sea	1994	7	4.81, 35.9	10.9		4
Tenojoki River, Arctic	1994	5	0.35, 8	2.4		4
Kymijoki Estuary	1996	3	7.1 1	7		5
Bream						
Kymijoki Estuary	1996	2	0.4	0.3	muscle	5
		2	4.9	1.8	liver	5
		1	13.1	3.0	spawn	5
Kymijoki River	1996	1	1.0	0.3	muscle	5
		1	4.1	1.8	liver	5
Lake Valkjarvi	1988, 1994	5	(0.33-0.6)			6
Burbot						
Kymijoki Estuary	1996	10	0.4	0.3	muscle	5
		2	82.4	31.2	liver	
		2	16	7.6	spawn	
Kymijoki River	1996	5	0.7	0.3	muscle	5
		5	122	24.6	liver	
		2	22.8	10.1	spawn	
Lake Nitsijarvi	1993/4	5	2.76		liver; background location;	1
Lake Pahtajarvi	1993/4	4	0.7		liver; background location;	1
Perch						
Kymijoki Estuary	1996	1	0.4	0.4	muscle	5
		1	6.1	4	spawn	
Kymijoki River	1996	8	3.8	3.9	liver	5
			4.1	2.7	spawn	
Lake Valkjarvi	1994	3	(0.45-0.7)			6
Pike						
Bothnian Bay	1993	5	(0.32-0.62) (140-270)	0.23		7
Kymijoki River	1993	3	(0.36-0.97) (200-540)	0.18		7
Lake Kernaala in South Finland	1987	2	(5-9)		only 2378-Cl ₄ DF, PCB contamination	8
Kymijoki Estuary	1996	2	0.3	0.4	Muscle	5
		2	4.0	1.1	Liver	
		1	4.6	7.8	Spawn	
Kymijoki River	1996	4	0.9	0.4	Muscle	5
		2	5.1	4.1	Liver	

Species, Location	Date	n	Mean Conc. (pg I-TEQ/g) (range)	Mean % fat	Comments	Ref.
Lake Valkjarvi	1988 and 1994	1 3	6.4 (0.21-0.5)	1.1	Spawn	6
Pike perch						
Lake Valkjarvi	1988, 1994	4	(<0.01-0.2)			6
Rainbow trout						
Experimental situation	ns	1	0.88		Prior to exposure	3
Experimental situation	ns	1	9.63		Fed contaminated Baltic herring for 4 months	3
Unstated source	1991?	6	0.53 (0.23-1.5)		Fed on normal feed	2
Unstated source	1991?	1	2.1, 4.17^N , 33.4		Fed on Baltic herring for 3 months	2
Lake 222	1993-94	4	0.08		background location;	1
Ruff						
Lake Valkjarvi	1994	1	0.57			6
Mussels						
Lake Vanaja	1995	3	61 (54-71)		Industrial area	9
River Kymijoki	1995	3	210 (150-260)		Contamination – Ky-5?	9
Walleye						
Lake Kernaala in South Finland	1987	1	6		only 2378-Cl ₄ DF, PCB contamination	8

^N = Nordic TEQ; n = number of samples; ns = not stated in literature;

data for fresh weight concentrations, unless in **bold** signifying lipid adjusted concentrations

References: 1. Vartiainen *et al.* (1996); 2. Vartiainen and Hallikainen (1992); 3. Isosaari *et al.* (1998); 4. Vuorinen *et al.* (1997a); 5. Korhonen *et al.* (1997b); 6. Vartiainen *et al.* (1995); 7. Koistinen *et al.* (1995); 8. Koistinen *et al.* (1989); 9. Koistinen *et al.* (1997a).

A4 France

Recently, France has initiated monitoring programmes. Special emphasis is given to the PCDD/PCDF emissions from known sources, such as municipal solid waste incinerators and metallurgical plants.

A4.1 FOODSTUFFS

The Ministry of Agriculture, Fisheries and Food has a surveillance programme in place to control the levels of PCDD/PCDF in milk. In the year 1994 and 1995, the average concentrations in cow's milk were between 1 and 4.5 pg I-TEQ/g fat. None of the samples was above the guideline concentration of 6 pg I-TEQ/g fat as established by the Dutch Government. Presently, France applies the same guideline concentration for dairy products as Germany, namely 5 pg I-TEQ/g fat. Dairy products exceeding this concentration are not allowed to be placed on the market (see also Task 1).

A press release dated February 18, 1999, stated that cow's milk, as a sensitive biomonitor for atmospheric pollution, should be monitored once a municipal solid waste incinerator had stack emissions greater than 10 ng I-TEQ/m³ in the year 1997. Subsequently, in 1998, cow's milk was measured in the neighbourhood of municipal solid waste incinerators. A summary of the 65 samples analysed for PCD/PCDF from April through to November 1998 is shown in Table A 13. Most sample were from cows grazing 3-5 km from the MSWI. The lowest concentration was 0.32 pg I-TEQ/g fat and the highest was 8.37 pg I-TEQ/g fat. The median of all samples was 1.50 pg I-TEQ/g fat. One goat milk sample gave 0.68 pg I-TEQ/g fat. Three samples of cow's milk had concentrations above the guideline concentration.

Table A 13: France – Cow's milk produced in the neighbourhood of municipal solid waste incinerators. Concentrations in pg I-TEQ/g fat

	n	Range	Mean
April 1998	4	0.32-0.97	0.53
May 1998	2	0.61 1+ 0.74	0.68
June 1998	6	1.52-6.77	2.77
July 1998	22	0.54-8.37	2.34
August 1998	25	0.42-4.02	1.87
September 1998	5	0.35-4.23	1.44
November 1998	1	1.81	
Total	65		1.91

Within the milk surveillance programme, a few vegetables were also analysed. The results are summarised in **Table A 14**.

**Table A 14: France – Vegetables grown in the surroundings of MSWIs.
Concentrations in ng I-TEQ/kg d.m.**

Vegetable	Concentration
Cabbage (choux)	0.21
Rhubarb	0.54
Salad	1.10
Salad	0.17
Onions	0.44
Herbs	1.62

A5 Germany

A5.1 SOIL

A PCDD/PCDF database summarising 1,594 soil samples from Germany was published in 1992 and shown in Table A 15 (BLAG 1992). The soil levels from rural areas ranged between 1 and 5 ng I-TEQ/kg. Also, relatively high concentrations - up to 46 ng I-TEQ/kg - were detected in the organic top-soils of forests. A maximum concentration of 140 ng I-TEQ/kg d.m. was found in litter. In urban areas, typically, the PCDD/PCDF concentrations in soil were between 10 and 30 ng I-TEQ/kg d.m. whereas, in industrial areas, the concentrations were up to 100 ng I-TEQ/kg d.m. PCDD/PCDF soil levels of 30,000 ng I-TEQ/kg were detected near point-sources, such as copper smelting plants, or in chemical waste disposal sites. Other, more recent data from Bavaria, showed that 89 % of 120 soil samples from arable land had concentrations below 1 ng I-TEQ/kg (UBA-DB 1998). The highest concentration was found to be 25 ng I-TEQ/kg.

Table A 15: Germany – Soil, per State. Concentrations in ng TEQ/kg d.m. (BLAG 1992 with additions from Fiedler 1998)

Location/Region	Use Pattern	n	Concentration	Reference
Germany	Rural		1-5	BLAG 1992
	Urban		10-30	BLAG 1992
	Pasture		Median: 0.4-8	BLAG 1992
	Rural		Mean: 3.0	
	Urban		Mean: 3.8	
	Arable land			BLAG 1992
	Rural		Mean: 2.0	
	Urban		Mean: 2.8	
	Litter /Forest			BLAG 1992
	Rural		Mean: 35	
Urban		Mean: 48		
	Point sources		up to 30,000	BLAG 1992
Bavaria	Arable land	120	<1	Joneck <i>et al.</i> 1992
Hamburg	Southeast, highly impacted		Median: 20 Range: 1.7-684	Umweltbehörde Hamburg 1993
Hessia	House gardens		10	HLfU 1991

- Measurements from potentially contaminated sites were very often confirmed: 230 ng TEQ/kg d.m. were found at a former site with cable burning; 139 ng I-TEQ/kg were determined in litter close to an aluminium smelter (Joneck and Prinz 1994, LfU 1992).
- Impact from automobiles can only be detected close to the streets. Thus, traffic cannot be considered as an aerial source for soil contamination (LfU 1992).

The UBA Dioxin Database (UBA-DB 1998) lists a total of 789 results from soil samples. A summary of the minimum and maximum concentrations found for different use patterns of the

soil in dependence of the location is given in Table A 16. There is a large range being covered by the analyses and obviously there is not a clear differentiation between rural and urban environments (the median concentrations were very similar). The table also shows that so-called “industrial” areas do not have to be contaminated with dioxins and furans necessarily. The concentrations ranged from almost zero to more than 5,000 ng I-TEQ/kg d.m. (in total there were 26 analyses available from Saxony).

Table A 16: Germany - Soil per use pattern. Concentrations in ng TEQ/kg d.m. (UBA 1998)

Type	Rural	Rural and Conurbation	Conurbation Background	Conurbation/Contaminated	Urban	Contaminated
Forest	0.04-38				0-50	0.05-139
Forest/coppice wood			0.04-36.3			
Coniferous forest	0.004-112					
Deciduous forest	0.02-102					
Mixed forest	0.06-5.4		0.2-1856			
Arable	0.003-3.7		2.8-74			0.2-25
Meadow	0.004-29.5	0.3-8.9	0.7-235	1.0-4.6		
Parkland			3.6-4.9			
Pasture	0.002-5.6	1.4-6.5		3.5	0-18	0.2-24
Fallow	0.003-0.03	0.9-7.7	3.0-41		0.1-17	0.2-230
Garden/parkland	1-3.1				0-13	0.5-42
Industrial			0.05-5742			
No vegetation		1.0				
Close to nature				1.2-8.3		
Clearfelled area	0.008-0.2					
Area of lakes, ponds, river				0.3-0.9		
Sports site/campsite		3.1				
Playground			0.7	1.0-3.2		
Residential				0.5		
Min	0.002	0.3	0.04	0.3	0	0
Max	112	8.9	5741	8.3	50	230
Mean	6.1	3.2	68.6	1.9	4.8	9.5
Median	3.4	3.0	14.4	1.5	2.3	4.8

From the database of the Federal Environmental Agency (UBA-DB 1998), the PCDD/PCDF results from 442 samples taken in Bavaria in 1989/90 are compiled in Table A 17. All samples were composites and analysed with GC/MS; for all samples, data for PAH and PCB are available as well. The data from Bavaria nicely show that the Of horizon was the layer that contained the highest PCDD/PCDF concentrations on a dry mass basis. This finding is due to the high content in organic carbon; the mineral horizons generally had lower concentrations. There is no difference between concentrations of background soils between urban and rural sites. This finding implicates that there is no difference in deposition; a finding that is confirmed by ambient air measurements in Bavaria (see section A5.3).

Table A 17: Germany – Soils in Bavaria (data compiled from UBA-DB 1998; Original sources: GLA 1991, 1994)

Type of location	Type of Project	Horizon	Date	n	Min	Max	Mean	Median
Rural	Routine	Ap	1989/90	27	n.d.	3.7	0.41	0.12
		Ah	1989/90	46	n.d.	5.6	0.46	0.21
		Of	1989/90	20	n.d.	38	11.9	8.8
		Ah	1989/90	15	0.04	3.9	1.01	0.59
		Ah	1989/90	2	0.97	3.1		
Urban	Routine	Ap	1989/90	41	n.d.	5	0.7	0.24
		Ah	1989/90	27	n.d.	18	3.9	0.6
		Of	1989/90	32	n.d.	50	14.9	10
		Ah	1989/90	30	n.d.	17	2.6	0.77
		Ah	1989/90	30	n.d.	13	1.8	0.95
		Ah/Ap	1989/90	4	0.1	17	4.8	1.02
Contaminated	Potentially contaminated	Ap	1989/90	41	n.d.	25	2.69	0.2
		Ah	1989/90	38	n.d.	24	3.73	1.05
		Of	1989/90	7	21	139	51.3	37
		Ah	1989/90	6	0.05	29	5.9	0.8
		Ah	1989/90	18	n.d.	42	5.38	0.9
		Ah/Ap	1989/90	15	0.7	230	21.26	3.8
Contaminated, near waste plants		Ap	1989/90	11	0.2	7.35	1.26	0.68
		Ah	1989/90	12	0.2	2.3	0.78	0.41
		Of	1989/90	2	3.72	3.87		
		Ah	1989/90	3	0.69	1.44		
		Ah	1989/90	7	0.47	1.63	0.94	0.83
		Ah/Ap	1989/90	8	0.22	2.45	1.6	1.72

A5.2 SEDIMENT

Sediment concentrations were reported for high contaminations: up to 1,500 ng 2,3,7,8-Cl₄DD in the Hamburg harbour (Götz and Schumacher 1990, Götz *et al.* 1993). Also, river sediments may contain high concentrations, *e.g.* between 41 and 73 ng I-TEQ/kg d.m. were detected in the river Elbe (Götz *et al.* 1994).

The UBA database (UBA-DB 1998) lists the results of 28 sediment samples from two major rivers in Germany (Table A 18): in the State of Lower Saxony the concentrations in Elbe sediments ranged from 1.17 to 19.2 ng I-TEQ/kg d.m. (median = 2.79 ng I-TEQ/kg d.m.); these concentrations represent background concentrations. Higher concentrations were found in the Elbe sediments close to the mouth of the river: in Hamburg, after receiving many industrial inputs, concentrations in suspended particles range from 17.5 to 76 ng I-TEQ/kg d.m. (median = 27.6 ng I-TEQ/kg d.m.). The results from the river Rhine range from 11.6 to 37.2 ng I-TEQ/kg with medians from two different studies of 34.7 and 28.4 ng I-TEQ/kg d.m. The results of a sediment core from Lake Constance is reported in Task 6.

**Table A 18: Germany –Sediments. Concentrations in ng I-TEQ/kg d.m.
* as suspended matter in the river Elbe**

Type	State	Type of Project	Date	n	Min	Max	Mean	Median
River Elbe	Hamburg	(Con)Urban backgr.	8/28/95	3	17.5	76.0	40.4	27.6
River Rhine	NRW	Special haz. situation	1989-1996	11	15.8	103	47.4	34.7
River Rhine	NRW	Special haz. situation	1995-1996	3	11.6	37.2	25.7	28.4
River Elbe	Lower Saxony	Environmental surveillance	Sep. 94	11	1.17	19.2	5.12	2.79

A5.3 AIR

In Germany, ambient air measurement programs were initiated since the mid 1980s. For 1993, “typical“ ambient air concentrations are shown in Table A 19 (BGA/UBA 1993). However, later results showed downward trends, due to the application of better flue gas abatement technologies. As an example, results from Northrhine Westphalia are compared in Table A 20. Within six years, a decrease in the PCDD/PCDF concentrations in ambient air of between 46% and 69% were determined.

Table A 19: Germany – Ambient air and deposition before 1992 (BGA/UBA 1993)

Description of Location	Ambient Air Concentration (fg TEQ/m ³)	Deposition (pg TEQ/m ² .d)
Rural area	25-70	5-20
Urban area	70-350	10-85
Close to point source	350-1,600	up to 1,000

Table A 20: Germany – Ambient air in Northrhine Westphalia as determined in 1987/88 and 1993/94 (NRW 1995)

Location	Concentration (fg I-TEQ/m ³)		Reduction (%)
	1987/88	1993/94	
Köln-Riehl	130	40	69
Duisburg-Meiderich	332	124	63
Essen-Altendorf	204	76	63
Dortmund (city)	224	120	46

For Bavaria, the same trend could not be confirmed. Whereas in 1992, analyses from 36 locations in Bavaria gave mean ambient air concentrations of 23 and 31 fg I-TEQ/m³ for rural and urban areas, respectively (LfU 1992), recent measurements (winter 1993/94 until summer 1996) in the surroundings of Augsburg and Burgkirchen did not show this downward trend. In the latter study, mean concentrations of 27 and 52, respectively, were determined (Fiedler *et al.* 1997). The results are summarised in Table A 21.

Table A 21: Germany - Ambient air in Bavaria (Fiedler *et al.* 1997)
Sp = Spring; Su = Summer; Wi = Winter
Concentrations in fg I-TEQ/m³

	n	Sampling Time	Mean	Minimum	Maximum
Bavaria	36				
Rural areas		1992-1993	22.5	3.3	88.4
Impacted areas		1992-1993	31.2	3.0	85.3
Augsburg					
Before start of MWI	125	Su92-Wi92/93	49	14	120
After start of MWI		Su94-Sp96	52	7.6	206
Burgkirchen					
After start of MWI	98	Wi93/94-Su96	27	4.4	78

It has to be mentioned that strong seasonal trends for PCDD/PCDF were found in all networks in Germany with higher concentrations during the winter months and up to 10-fold lower concentrations during the summer months. In the Bavarian networks, the difference of the mean concentrations was 10-fold.

The UBA Dioxin Databases lists 849 ambient air samples from four States (the database does not contain the data discussed above). A summary of these data is shown in Table A 22. The concentrations in ambient air ranged from 2 fg I-TEQ/m³ to 812 fg I-TEQ/m³. Although the data are highly aggregated it can be seen that the concentrations in summer were lower than the concentrations in winter (with Hessia as an example). For more details, the original publications should be consulted. From the data it can also be concluded that at each location and independent of the season, there is at least one outlier with comparatively high concentrations. This finding was also detected in the Augsburg/Burgkirchen program (see Table A 21).

Table A 23 lists the results of the 387 deposition samples from the UBA Dioxin Database (UBA-DB 1998). All samples were taken according to VDI Richtlinie 2119, Blatt 2, the so-called Bergerhoff method. The PCDD/PCDF concentrations range from 0.5 to 464 pg I-TEQ/m²·d. Interestingly, the samples collected in July 1995 in the city of Hamburg, one of the most densely populated and highly industrialised areas in Germany, showed quite low PCDD/PCDF concentrations (median of 3 and 6 pg I-TEQ/m²·d, respectively).

Table A 22: Germany - Ambient air (UBA-DB 1998). Concentrations in fg I-TEQ/m³

State	Type	Sampling Period	n	Min	Max	Mean	Median
Bavaria	urban	1992/93	12	5	68	23	13
Bavaria	urban	1992/93	13	4	67	27	21
Bavaria	urban	1992/93	61	3	343	33	25
Bavaria	urban	1992/93	107	3	179	29	24
Bavaria	urban	Fall/winter 1992	6	5	36	19	20
Hessia	-	spring 90-95	61	4	812	30	14
Hessia	-	spring 90-95	77	2	705	59	36
Hessia	-	Summer 90-95	67	3	433	18	9
Hessia	-	Summer 90-95	87	6	232	40	25
Hessia	-	autumn 89-95	68	11	379	70	51
Hessia	-	autumn 89-95	88	27	454	112	85
Hessia	-	winter 90-95	56	11	216	68	50
Hessia	-	winter 90-95	73	30	464	118	99
NRW	High impact	1991/92	1	78			
Thuringia	urban	Oct 93 - Oct 97	48	9	231	71	56
Thuringia	urban	Oct 93 - Oct 94	12	15	126	61	52
Thuringia	urban	Oct 93 - Oct 94	12	18	210	92	83
Total			849				

Table A 23: Germany – Particulate deposition (UBA-DB 1998). Concentrations in pg I-TEQ/m²-d

Region	Type of location	Sampling Period	n	Min	Max	Mean	Median
Brandenburg	conurbation	Oct 1993	4	26	47	36	36
Hamburg	urban background	Jul 95	12	1	7	3	3
Hamburg	urban background	Jul 95	12	2	10	6	6
Hessia		1989-1995	72	2	181	21	11
Hessia		1989-1995	34	1	20	7	7
Hessia		1989-1995	73	0	32	5	4
Hessia		1989-1995	70	0	23	4	3
NRW	urban background	1993/94	13	7	35	20	19
NRW	contaminated	1991/92	1	17	17	17	17
Rheinland-Palatinate	urban	1993/94	24	0.5	24	9	8
Thuringia	urban	1993-1997	48	3	464	29	14
Thuringia	urban	1993/94	12	11	169	52	38
Thuringia	urban	1993/94	12	10	407	73	27
Total			387				

A5.4 VEGETATION

A5.4.1 Grass

Welsh Rye grass is typically exposed for four weeks during the summer (May through October). In Bavaria, typical ranges are 0.5-1 ng I-TEQ/kg d.m., the maximum concentration was 2.1 ng I-TEQ/kg d.m.

The UBA Dioxin Database lists PCDD/PCDF concentrations in the grass samples from 1.3 to 7.7 ng I-TEQ/kg dm; in a situation of a known dioxin contamination, 10.9 ng I-TEQ/kg dm were found. These numbers are higher than those found in Bavaria in Welsh rye grass.

A5.4.2 Kale

According to the German guideline for biomonitoring, kale is exposed for eight weeks starting after the last exposure of the Rye grass (typically October to early December). The ranges of PCDD/PCDF concentrations found in Bavaria are relatively narrow: 1.0-1.7 ng I-TEQ/kg d.m. However, close to combustion sources, 3.7-12.6 ng I-TEQ/kg d.m. were detected whereas, in remote areas, concentrations of 0.5 ng I-TEQ/kg d.m. was measured (Köhler 1994). The concentrations found in Kale Northrhine Westphalia (1990-1992) are summarised in Table A 24. It should be noted that the denomination “baseline” refers to a highly industrialised and densely populated area in Germany and to a time when dioxin air pollution abatement in many combustion related sectors, such as waste incineration, iron and steel as well as non-ferrous metal industries, was just going to start.

Table A 24: Germany – Kale from Northrhine Westphalia, years 1990-1992 (MURL 1993)

	Loaction	Year	n	Median	Min.	Max.	Remarks
Kale (baseline)	c	1990	15	0.58	0.13	2.08	
Kale	b	1990	8	0.85	0.58	1.27	
Kale	a	1990	10	2.10	1.35	8.68	Bioindicator
Kale	c	1990	4	0.78	0.57	1.67	Bioindicator
Kale	b	1990	4	1.64	1.46	2.21	Bioindicator
Kale	c	1991	4	1.20	1.00	1.50	Reference
Kale	c	1992	5	2.30	0.48	33.60	1)
Kale	c	1992	3	1.26	1.18	1.65	2)

a = Urban center

b = Edge of urban center

c = Rural area

n = Number of samples

1) = Fire of a plastic storage

2) = Close to cement kiln

Median, Min., Max. = Concentrations in ng I-TEQ/kg d.m.

Additional data are available from a biomonitoring program in Hestia, which reported PCDD/PCDF concentrations of 28 samples ranging from 0.75 to 1.89 ng BGA-TEQ/kg d.m. (Table A 25). The study also investigated if heavy air traffic will increase the PCDD/PCDF concentrations in air. As can be seen from Table A 25, the PCDD/PCDF concentrations in the

nine biomonitors exposed at the Frankfurt airport did not have higher levels than the remainder samples in the state of Hesse. For completeness, data from Hamburg are included as well: in this much more urban environment, the concentrations in kale are somewhat higher than in Hesse.

Table A 25: Germany – Kale in Hesse and Hamburg (ng BGA-TEQ/kg d.m.)

	Hesse	Frankfurt Airport	Hamburg
Number of Samples	28	9	22
Mean	1.09	1.18	2.32
Median	1.05	1.14	2.35
Minimum	0.75	0.84	0.69
Maximum	1.89	1.75	4.78

A5.4.3 Spruce Needles

First studies reported PCDD/PCDF concentrations in spruce needles 1.15 and 4.47 ng I-TEQ/kg d.m. (Table A 26) (Reischl *et al.* 1991). Later data from Bavaria and Hesse in Germany gave mean PCDD/PCDF concentrations in pine needle extracts ranging from 0.53 to 1.64 pg I-TEQ/g d.m. (Table A 27) (Köhler *et al.* 1994, Fiedler *et al.* 1995).

Table A 26: Germany - Spruce needles before 1990 (Reischl *et al.* 1991) (ng BGA-TEQ/kg d.m.)

	Nürnberg	Passau	Hof	Schwandorf
Concentration	4.47	1.17	1.15	1.83

Table A 27: Germany - Spruce needles (ng BGA-TEQ/kg d.m.) (Köhler *et al.* 1994)

	Number of Samples	Minimum	Maximum	Mean	Median
October 1992	26	0.18	1.20	0.53	0.43
April 1993	26	0.27	3.45	1.12	0.84

A5.5 WILDLIFE

The UBA Dioxin Database contains 40 results classified as “wildlife” (Table A 28). Of these, the majority was on fish and shellfish; the remainder being grass (11). The samples from Bavaria and Northrhine Westphalia listed in the section A5.4 were not included. Except for one case, composite samples were analysed. Most investigations were done in special situations where PCDD/PCDF contamination was suspected.

The median concentrations in birds were between 1.9 and 2.8 pg I-TEQ/g fresh weight. Two perch samples from Brandenburg gave 40 and 51 pg I-TEQ/g fat. Mussels showed lower concentrations than fish, with a median of 0.81 pg I-TEQ/g fw.

Table A 28: Germany – Wildlife. All concentrations in ng I-TEQ/kg (either fresh weight – f.w. or dry matter – d.m., see column Unit) (UBA 1998)

Type	State	Type of Location	Date	n	Min	Max	Mean	Median	Unit	Ref
Herb/grass	Brandenburg	Conurbation backgr.	Nov 93	2	1.3	1.5	1.4	1.4	d.m.	1
Herb/grass	Brandenburg	Point source nearby	Nov 93	8	2.5	7.7	4.4	3.7	d.m.	1
Grass	NRW	Contaminated	Jan 94	1	10.9	10.9	10.9	10.9	d.m.	3
Brace	Brandenburg	-	Apr 94	2	1.8	1.9	1.9	1.9	fw	
Brace	Hamburg	-	Jul 94	6	0.48	4.1	2.1	2.0	fw	1
Brace	Lower Saxony	-	Jul 94	3	0.68	13	5.5	2.8	fw	1
Brace	Saxony	-	Apr 94	2	0.83	4.2	2.5	2.5	fw	1
Brace	Schleswig-Holstein	-	Apr 94	1	2.4	2.4	2.4	2.4	fw	1
Perch/fish	Brandenburg	Point source nearby	Nov 93	2	42	60	51	51	d.m.	1
Mussels	Lower Saxony	Rural backgr.	Aug 94	13	0.56	0.96	0.77	0.81	fw	2

- 1 F&E Biomonitoring of the river Elbe, connection with investigations on sediment in longitudinal profile of the river Elbe, 1994
- 2 Abschlußbericht der ERGO Forschungsgesellsch. mbH , i.A. des Niedersächsischen Umweltministeriums: Untersuchung der Belastung von Böden, Miesmuscheln und Sedimenten aus dem Bereich Wilhelmshaven, Abschlußbericht, Mai 1995
- 3 Determination of substance flow, entering of deposition rates in soil, determination of deposition rates to value in soil

A5.6 SEWAGE SLUDGE AND COMPOST

A5.6.1 Sewage Sludge

PCDD/PCDF analyses of German sewage sludges indicate a trend for decreasing concentrations during the last years. In 1987, the first survey on PCDD/PCDF concentrations in German sewage sludges, Hagenmaier determined an average concentration of 202 ng I-TEQ/kg d.m. (range: 28-1,560 ng BGA-TEQ/kg d.m.) (Hagenmaier 1988). In 1990, the average concentration from more than 300 samples of sewage sludge, which were used in agriculture, was 50-60 g I-TEQ/kg d.m. (Mach and Butzkamm-Erker 1990). Only in a few cases, 2,3,7,8-Cl₄DD could be detected. About 80 % of all samples had concentrations below the (legal) limit concentration of 100 ng TEQ/kg d.m.

Analytical data on PCDD/PCDF concentrations in sewage sludges obtained from 30 and 28 sewer plants, respectively, in the area of Frankfurt, a densely populated and industrialised area in Germany have been published (Table A 29). In most cases PCDD concentrations were about twice the PCDF concentrations. In 1991 two out of 30 sewage sludge samples showed concentrations above the limit value of 100 ng I-TEQ/kg d.m. set by the German Sewage Sludge Ordinance for use of sewage sludge in agriculture; in 1992 no concentrations above the limit value was reported (Fiedler *et al.* 1996).

Table A 29: Germany - Sewage sludges from greater Frankfurt area, data from two subsequent years, same sewer plants (Sachs-Paulus 1992)

	August 1991		March 1992	
Number of samples	30		28	
Minimum	18	ng TEQ/kg d.m.	9	ng TEQ/kg d.m.
Maximum	144 (620)	ng TEQ/kg d.m. ng TEQ/kg d.m.)	63	ng TEQ/kg d.m.
Mean	39	ng TEQ/kg d.m. ¹	24	ng TEQ/kg d.m.
Median	32	ng TEQ/kg d.m. ¹	23	ng TEQ/kg d.m.
Samples TEQ > 100 ng/kg d.m.	2		0	
Samples TEQ > 50 ng/kg d.m.	8		1	

¹ = results without the extreme value of one sewage sludge plant

The presence of PCDD/PCDF in sewage sludge is not a recent finding. Lamparski *et al.* (1984) detected dioxins and furans in sludge samples from the year 1933. The authors concluded that chlorination of drinking water might be the cause. Since this finding, further evidence was gained to explain the presence of PCDD/PCDF in sewage sludge with no obvious industrial inputs. It was found that waste waters from washing machines are the major contributor to the dioxin and furan load in sewage sludge. The results from Germany did not attribute a high percentage to run-offs from streets and roofs. Some other sources are detergents, human and animal feces (Horstmann 1994, Horstmann and McLachlan 1995a).

A5.6.2 Compost

Composting of bio- and kitchen waste is a common practice in Germany, firstly to obtain a fertiliser to be used in house gardens and horticulture and secondly to minimise wastes to be disposed of. As can be seen from Table A30, composting of the total organic fraction from municipal solid waste results in an unacceptable dioxin and furan contamination¹. Concentrations below the guideline limit of 17 ng I-TEQ/kg d.m. can be obtained if only kitchen wastes or so-called green wastes, which are the greens from house gardens, parks, *etc.* are composted.

Table A30: Germany - Compost, typical ranges

Municipal solid waste compost	38 ± 22	ng I-TEQ/kg d.m.
Biowaste compost	14 ± 9	ng I-TEQ/kg d.m.
Green compost	11 ± 8	ng I-TEQ/kg d.m.

To obtain an overview on the present levels of dioxin contamination in bio-compost, 101 analyses have been evaluated with the following result: most samples had concentrations between 7.49 and 17.5 ng I-TEQ/kg with a median of 11.3 ng I-TEQ/kg.

¹ The guideline concentration of PCDD/PCDF in compost is 17 ng I-TEQ/kg d.m. (for reference, see Task 1)

Green and bio composts have nearly the same homolog distribution for PCDD and PCDF; in other words, the profiles are very similar (Figure 1). The most abundant homolog (and congener) is Cl₈DD followed by Cl₇DD (Fricke 1994).

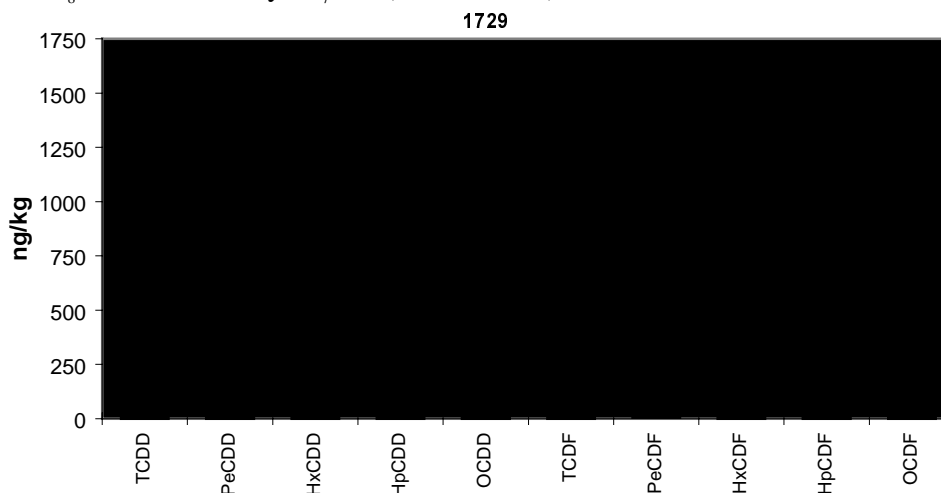


Figure 1: Homolog concentrations in biocomposts (n=62); concentrations in ng I-TEQ/kg normalised to 30 % original substance per kg dry matter (Fricke 1993)

During the last years, there was hardly a change observed: also in the city of Hamburg it was found that the dioxin concentrations in biocompost were in a very narrow range; a downward trend could not be established (Table A 31).

Table A 31: Germany - Organic fraction from private households in Hamburg (biocompost) (Hutzinger *et al.* 1995)

Location	Year	N	PCDD/PCDF (ng I-TEQ/kg d.m.)
Bergedorf	1992	2	4.8; 9.8
Harburg	1992	2	6.1; 10.4
<i>Mean from Bergedorf and Harburg</i>	<i>1992</i>	<i>4</i>	<i>7.8</i>
Bergedorf	1993	2	11.2; 15.7
Bergedorf	1994	1	21.1
Bergedorf	1995	1	7.0
<i>Mean Bergedorf</i>	<i>1993-1995</i>	<i>4</i>	<i>13.8</i>
Harburg	1993	1	10.6
Harburg	1994	1	10.4
Harburg	1995	1	12.1
<i>Mean Harburg</i>	<i>1993-1995</i>	<i>3</i>	<i>11.1</i>
<i>Mean Bergedorf and Harburg</i>	<i>1993-1995</i>	<i>7</i>	<i>12.6</i>

A5.7 COMMODITY GOODS

The presence of PCDD/PCDF in commodity goods was proven many times. Some examples identified in Germany are shown in the following sections.

A5.7.1 Textiles and Wool

For textiles, the occurrence of dioxins and furans in the final products can be due to:

- Use of chlorinated chemicals, *esp.* PCP, to protect the raw material (cotton, wool or other fibers, leather, *etc.*)
- Use of dioxin-contaminated dye-stuffs
- Formation of PCDD/PCDF during finishing.

In twelve out of 13 samples of raw textiles, Horstmann found concentrations below 1 ng I-TEQ/kg (mean: 0.16 ± 0.24 ng I-TEQ/kg). However, the 13th sample (bleached polyester) contained 244 ng I-TEQ/kg (Horstmann 1994, Horstmann and McLachlan 1995b). Based on the analyses of 16 samples, it was concluded that – at least for the finishing processes used in the German textile industry – the dioxin concentration will not increase significantly: mean concentrations found in finished cotton were at 0.21 ± 0.10 ng I-TEQ/kg with a median of 0.20 ng I-TEQ/kg (Horstmann 1994). These results were confirmed by random sample analyses of raw and pre-treated cotton arriving at the Hamburg harbour, which contained 0.03-0.2 ng I-TEQ/kg (Hutzinger *et al.* 1995, FHH 1995).

The results of PCDD/PCDF analyses of textiles found in department stores in Germany are summarised in Table A 32. Of the samples 17, 21 and 23, which were also analysed for PCP, only T-shirt sample 21 contained detectable concentrations of 2,000 ng PCP/g. Thus, no correlation between PCP and PCDD/PCDF could be established (such result was confirmed in other cases as well) what is reasonable as PCP is water soluble and will be removed with this finishing process whereas the PCDD/PCDF adsorb to the fibre.

A screening exercise of more than 140 textiles showed that 90% were not contaminated (Σ PCDD/PCDF < 1,000 pg/g, I-TEQ < 10 pg/g). On the other hand, sometimes extremely high contamination was determined as well: Highest concentrations were up to 300,000 pg/g Σ PCDD/PCDF or 370 pg I-TEQ/g (Horstmann and McLachlan 1995b, Klasmeier and McLachlan 1998). The homologue profiles of all these samples was dominated by the higher chlorinated PCDD and PCDF (Cl₇ and Cl₈) indicating that either pentachlorophenol or dyestuffs are the source of the contamination.

Table A 32: Germany – Textiles. Concentrations in ng/kg.
PA = Polyamide, PAN = Polyacryl nitrile, PE = Polyethylene

	Type	Colour	Fibers (%)	Σ PCDD/PCDF	I-TEQ
1	Socks	White, red rings	64 PA, 36 PAN	20	0.24
2	Socks	White, blue rings	81 Cotton, 19 PE	21	0.17
3	T-Shirt	White	100 Cotton	5.2	0.07
4	Bedsheet	White	100 Cotton	131	1.31
5	Body	White-grey	100 Cotton	6.9	0.01
6	Legging	White	95 Cotton, 5 Elasthan	23	0.11
7	T-Shirt	Dark blue	100 Cotton	99	0.27
8	T-Shirt	Light blue	100 Cotton	3210	3.64
9	Towel	Violet	100 Cotton	144	0.22
10	Bedsheet (jersey)	Amber	100 Cotton	174	0.54
11	Underwaer (men)	White	100 Cotton	23	0.74
12	Underwear (women)	White	100 Cotton	9.6	0.05
13	Shirt (men)	Light/dark blue stripes	65 PE, 35 Cotton	356	8.42
14	Jeans	Blue	100 Cotton	93	0.21
15	T-Shirt	Orange	100 Cotton	15	0.59
16	Socks	Blue	100 Cotton	37	0.22
17	T-Shirt	Light blue	100 Cotton	1240	2.11
18	T-Shirt	Rosa	100 Cotton	259	0.88
19	T-Shirt	Blue	100 Cotton	667	1.51
20	T-Shirt	Dark blue	100 Cotton	24	0.14
21	T-Shirt	Green	100 Cotton	16330	26.82
22	T-Shirt	Green	100 Cotton	920	1.96
23	T-Shirt	Violet	100 Cotton	295440	369

Mayer (1998) analysed 30 wool samples and wool products including untreated two samples of sheep wool and four samples of lanoline for PCDD/PCDF. 25 of the samples had very low PCDD/PCDF concentrations ($< 0,2 \mu\text{g } \Sigma\text{PCDD/PCDF/kg}$). Elevated concentrations were found in three samples of socks (1-1.4 $\mu\text{g } \Sigma\text{PCDD/PCDF/kg}$); the maximum was at 10.6 $\mu\text{g } \Sigma\text{PCDD/PCDF/kg}$. The concentrations expressed as I-TEQ were less than 1 ng I-TEQ/kg in low contaminated wool samples. The highest concentration was 86 ng I-TEQ/kg. Besides the 2,3,7,8-substituted congeners, all samples contained high concentrations of non-2,3,7,8-substituted congeners, indicating that the body burden of the animals (which should exclusively contain 2,3,7,8-substituted congeners) is not the only source of the PCDD/PCDF in the wool samples. Atmospheric deposition may be a source of some of these congeners. However, a considerable similarity between the patterns found in the wool and pentachlorophenol suggested that the chemical might have been used during production or storage of the wool. In another sample, very high concentrations of Cl_6DF have been detected, where the source could not be identified.

A5.7.2 Leather

Leather was found to be contaminated with PCDD/PCDF, too; mainly due to the practice to use PCP in many countries. Since the ban of PCP in Germany from the year 1989², PCDD/PCDF concentrations in leather goods decline. In contrast to textiles, PCP once applied on leather is not so easily removed by washing processes. Malisch (1994b) found up to 430 ng I-TEQ/kg in leather “breast-wallets”, in leather shoes, the concentrations were up to 6,400 ng I-TEQ/kg. Klasmeier and McLachlan (1997) analysed 49 different leather samples and found that ten contained PCDD/PCDF above the legally binding limits (see Task 1). Highest concentrations were at 2,100 or 3,000 ng I-TEQ/kg in shoes (bought in 1991). However, in shoes from the year 1996, highly elevated concentrations continued to exist. For leather goods, the PCP concentrations correlate with PCDD/PCDF concentrations at least qualitatively. The homologue and congener profiles and patterns strongly indicate that PCP is the source of the dioxin contamination.

A5.7.3 Wood and Cork

Analyses found a wide range of dioxin concentrations in PCP-treated wood: 1,500-19,000 ng I-TEQ/kg at the surface (0-1 mm) (Ehmann *et al.* 1993). Untreated wood typically has approximately 5 ng I-TEQ/kg; such contamination was confirmed by Fiedler *et al.* (1993a) who detected 4.6 ng I-TEQ/kg in a roof construction. More recent investigations show a decreasing trend in dioxin contamination what may be due to the fact that less wood is treated with PCP.

Cork is widely used to seal wine and Champaign bottles but also in the living environment, *e.g.* wall and floor coverings. Whereas in the wine industry only natural cork is used, the latter applications ask for bleached materials (very often with chlorine) and for biocidal treatment (often with PCP). Both facts may lead to dioxin contaminations in the product. According to Frommberger (1991) corks for wine were in the range 0.18-2.6 ng BGA-TEQ/kg³; cork in wall coverings were contaminated with 12.6 ng BGA-TEQ/kg. Whereas the wine cork exhibited a pattern typical for chlorine bleaching (1,2,7,8-Cl₄DF dominating), the homologue pattern of the coverings was dominated by the higher chlorinated PCDD and PCDF indicating PCP as the dioxin source.

A5.7.4 Candles and Raw Materials for Candle Production

Lau *et al.* (1997) analysed the emissions of three candles without any finish (beeswax, stearin, and paraffin, respectively), and 13 paraffin candles with different colours and surface finish. The concentrations of PCDD/PCDF in waxes, wicks, colours, and lacquers are shown in Table A 33. The raw materials for the production of candles – paraffin and stearin – showed very low dioxin contaminations (0.6 and 1.6 ng I-TEQ/kg). The natural product bee wax

² Which sets a maximum concentration of 5 mg PCP/kg in the final product

³ BGA-TEF = TEF according to the older TEF scheme as established by the former German Health Agency and the Federal Environmental Agency. In these samples, the difference between the BGA-TEF and the I-TEF are negligible.

contained 11 ng I-TEQ/kg. Wicks contained 0.08-0.18 ng I-TEQ/kg (Lau *et al.* 1997). Only composite samples were analysed and therefore no information on the composition of specific colours or lacquers is available. Levels of PCDD and PCDF in both samples are low and do not indicate dioxin contamination. As a general statement it can be concluded that levels of organic pollutants found in raw materials for candle manufacturing are low.

Table A 33: Germany - Waxes and candle materials. Concentrations in ng I-TEQ/kg

Type of Wax Concentration	Paraffin Wax 0.59	Stearin Wax 1.62	Beeswax 10.99
Wicks Used for Concentration	Paraffin Candles 0.18	Stearin Candles 0.12	Beeswax Candles 0.08
Candle colours Concentration	Composite of 6 Colours 0.5		Composite of 3 Lacquers 6.5

Candles coloured with Pigment Violet 23 or Direct Blue can be contaminated with PCDD/PCDF. When analysing 30 candles, Malisch (1994c) found concentrations up to 1.8 ng I-TEQ/kg in the coloured layer; the mean was at 0.33 ng I-TEQ/kg, the median at 0.18 ng I-TEQ/kg. The Environmental Agency in Hamburger detected similar ranges: 1.2-1.4 ng I-TEQ/kg (Umweltbehörde Hamburg 1995).

A5.7.5 Pulp and Paper

A research project was performed in 1993 to evaluate the German pulp and paper products for PCDD/PCDF (FhG-ILV 1993). The major results can be summarised as follows:

- All samples contained Cl₈DD; 17 of 19 samples of primary pulps contained 2,3,7,8-Cl₄DF in quantifiable concentrations. 2,3,7,8-Cl₄DD was positive in only four samples. The highest concentrations were in a newspaper with 1.65 ng I-TEQ/kg. In general, Swedish and Canadian paper products had contaminations below 1 ng I-TEQ/kg.
- The lowest PCDD/PCDF contaminations were present in samples using the Organocell⁴ or the ASAM⁵ processes (< 1 ng I-TEQ/kg).
- Papers, cardboards and cartons produced from recycling paper (year 1991) had dioxin concentrations in the range from 0.83 and 11.53 ng I-TEQ/kg d.m. (see Table A 34). De-inked products had lower contaminations, especially hygiene papers, newspapers and graphical recycling papers. All secondary products had more PCDD than PCDF.

⁴ Bleaching sequence: Oxygen in alkaline medium added with hydrogen peroxide followed by peroxide

⁵ Uses an alkaline sulfite step with anthraquinone, oxygen and ozone bleaching

**Table A 34 Germany - Products made of recycling paper (FhG-ILV 1993).
Concentrations in ng I-TEQ/kg**

Type of Paper	Number of Samples	Range
Hygiene papers	4	0.83-2.57
Newspaper	3	1.48-2.73
Graphical recycling paper	3	2.83-3.24
Carton for box	3	4.54-10.15
Testliner II	4	7.40-11.53
Wellenstoff	4	6.93-11.12
Grey carton	3	7.09-11.42
Wrapping paper	1	9.13

Only three of the 26 samples contained 2,3,7,8-Cl₄DD above the detection limit of 0.02 ng/kg. The relatively high concentration of 4.0 ng 2,3,7,8-Cl₄DD was detected in packaging paper. Most samples contained higher concentrations of the higher chlorinated PCDD; the congener with the highest concentration was Cl₈DD (up to 23,000 ng/kg). In general, unbleached products had lower concentrations of PCDD/PCDF than bleached products. Recycling products had higher concentrations than products made from primary fiber. The typical chlorine bleach pattern with a dominance of Cl₄DD and Cl₄DF was no longer found.

A5.8 WASTES AND RESIDUES

A5.8.1 Wastes and Residues from Waste Incineration

Presently, there exist approximately 50 municipal solid waste incinerators (MSWI) in Germany. Since December 1, 1996, all MSWI have to comply with the stringent limit values set in the 17th Ordinance on Incineration of Waste (17th BImSchV 1990). Whereas there exists a large database on PCDD/PCDF stack emissions, the concentrations of PCDD/PCDF in the solid residues are less frequently analysed. PCDD/PCDF concentrations in slags and bottom ashes are orders of magnitude lower than concentrations in fly ashes. In its report on the status of waste incineration, published in 1990, the Environment Agency found an average of 50 ng I-TEQ/kg for bottom ashes and 13,000 ng I-TEQ/kg for fly ashes from municipal solid waste incinerators.

Concentrations of PCDD/PCDF in bottom ashes and fly ashes from Bavarian MSWI detected higher concentrations in slags but also much lower contamination – below 1,000 ng I-TEQ/kg – in fly ashes. It is assumed that the modern MSWI will have better burn-out of the ashes and thus, lower concentrations of PCDD/PCDF than the former generation of MSWIs. As an example: the new MSWI in Hamburg has mean dioxin concentrations of 19 ng I-TEQ/kg in the bottom ash and 1,100 ng I-TEQ/kg in the fly ash.

A5.8.2 Other Ashes and Sludges

Ashes from fire places in private homes were analysed for PCDD/PCF as well. Dumler-Gradl *et al.* (1995) determined concentrations up to 42,000 ng I-TEQ/kg in fireplaces, chimneys, *etc.* fuelled with wood or coal (n=118). The mean concentrations were in the range of several thousand nanograms TEQ per kilogram (range: 4.0-42,048 ng I-TEQ/kg). No difference in concentrations was found between heating system fed with coal or wood.

The Dioxin Database of the German Environment Agency contains a few data reporting PCDD/PCDF concentrations in sludges and ashes from metallurgical treatment plants. All samples are from States in the New Laender (former GDR).

Table A 35 Germany - Sludges and ashes (UBA 1998). Concentrations in ng I-TEQ/kg

Type of sludge	Treated?	State	Sample Date	n	Min	Max	Mean	Median
industrial sludge: plants to roast, melt or sinter of ores	Dust from 2 ESPs	Brandenburg	Nov 93	2	623	9127	4875	4875
Municipal sewage sludge	Biological sewage purification	Brandenburg	Aug 93-Apr 94	16	27.6	1207	274	158
Stack ash from plant for melting and refining copper	-	Saxony-Anhalt	-	1	311	311	311	311
Shaft furnace ash (copper works), plant for melting and refining copper	-	Saxony-Anhalt	-	1	23779	23779	23779	23779
Dust from anode casting systems, plant for melting and refining copper	-	Saxony-Anhalt	-	3	1225	18660	7425	2392
Ash from plant for melting and refining copper	-	Saxony-Anhalt	-	2	311	23779	12045	12045
Ash from copper melting / refining plant	filter dust	Saxony-Anhalt	-	3	1225	18660	7425	2392
Municipal sewage sludge	biological sewage purification	Thuringia	1993-95	51	0.7	170	14	2.1

A5.8.3 Residues from Dry Cleaning

The distillation residues from dry cleaning shops can contain dioxin concentrations in the range of several ppb TEQ. It was found that the perchloroethylene used for dry cleaning was not the source of the PCDD/PCDF contamination. Also, the process itself does not generate PCDD nor PCDF. The major source are the textiles to be cleaned in the shops. It was found that the residues from shops that exclusively cleaned cloths had lower PCDD/PCDF concentrations than shops that cleaned new textiles (2-3 µg I-TEQ/kg). The textiles were identified as the major source rather than adsorption of dust particles on the cloth nor body fluids (sweat) of the persons wearing these cloths

A6 Greece

Information on the concentrations of PCDD/PCDF in Greece is extremely scarce. Information has only been found relating concentrations in soil and sand in Crete (Martens *et al.* 1998, Schramm a. No information on the concentrations in other media seems to be available for Greece.

A6.1 SOIL

The concentrations of PCDD/PCDF were measured in soil/sand samples in the vicinity of a site of uncontrolled solid waste combustion in the Kouroupitos ravine, Crete, Greece. The samples were collected in October 1996. The reference site is likely to be typical of agricultural soils in Greece; all others sites were directly affected by the ash from the open burning of municipal refuse or material from the landfill may have eroded down the ravine to the beach. Characteristics of the sampling sites together with the concentrations are shown in Table A 36. The background concentration for Greece is 2 ng I-TEQ/kg

Table A 36: Greece - Soil (ng I-TEQ /kg d.m.)

Site	Distance from waste site (km)	Concentration (ng I-TEQ/kg)
Area of the waste disposal site inside the ravine	0	1144
Area of waste dumping	0.06	34
Arable land as reference	15	2
Sand sample from the beach at the exit of the ravine to the sea	0.1	45

A7 Ireland

From Ireland, there are only a few data available. In addition, no research or monitoring programmes to analyse PCDD/PCDF have been identified (see Task 1). The data generated in the past addressed analyses of soil and cow's milk. In general, the PCDD/PCDF concentrations found in a variety of locations are low.

A7.1 SOIL

Five studies were identified which reported concentrations in soils; all were small scale surveys. The results are shown in Table A 37. Maloney & Associates (1997) reported on a baseline study in the vicinity of the Avondale plant at Rathdrum, Co Wicklow. This plant produces a variety of pharmaceutical products and intermediates. The study analysed PCDD/PCDF concentrations in soils and cows' milk. Soil was analysed in six rural areas and one forested site. Eight cores were taken at each site and mixed into a composite sample for analysis. Very few congeners were detected in the soil. The analysis had quite low sensitivity, with detection limits between 1 and 17 ng/kg, and therefore there is considerable uncertainty over the results. A further study was undertaken in 1998 to improve the data quality (Maloney & Associates 1998). The same procedure was used, including one new sample location. Lower levels were reported, as a result of lower detection limits and, therefore, fewer results based on assumed levels at the limit of detection

The Forbait (1994) study also considered both soil and cows' milk. This was undertaken on behalf of Cork County Council to establish levels of dioxins in various types of location, both rural and urban, across the County. Fourteen samples were taken from four locations, but the actual distribution of these samples is not known. Levels were, again, low and were associated with traffic and industrial combustion sources.

Two further sources of data were from small studies relating to specific sites. The first is a soil survey at the site of Merck Sharpe and Dohme, Ballydine, County Tipperary (AES 1997). The second was part of a wider study of Animal Health Problems at Askeaton, County Limerick (Environmental Protection Agency 1995). These studies again showed low PCDD/PCDF concentrations.

A7.2 COW'S MILK

Cows' milk has been sampled as an indicator of environmental levels of dioxins and will therefore, be included in this report, rather than in the human exposure task report. No estimates have been made of human exposure to dioxins in Irish cows' milk. Results of the analyses of cow's milk are shown in Table A 38.

The main data source for this section was the Irish EPA report on a national survey (Concannon 1996). This is the only national scale dioxins survey in Ireland to date. A total of thirty-two samples were taken from regional creameries, reflecting background levels, and

from individual road tankers, chosen to represent individual potential impact areas close to sources of contamination. Concentrations were low, and stated to be consistently lower than that reported for other European countries.

In addition, the data from Maloney & Associates (1997, 1998) were available. Limits of detection in the first study were relatively high, and no congeners were detected in milk. Levels given were based on the assumption that non-detects were present at the level of half of the detection limit. Levels estimated were low. These numbers have been recalculated from whole milk results, assuming 4% fat. The second survey in 1998, used analysis with lower detection limits. The concentrations in milk were lower in this survey, as was the case for the second soil survey.

Lastly, milk samples were included in the Cork Dioxin Survey (Forbait 1994), but, again, no congeners were detected. Concentrations were assumed to be equal to the limit of detection for each congener in each individual test. The concentrations given are therefore over estimates.

Table A 37: Ireland - Soil

Location	Land use type	Date	n	Median Conc. (ng I-TEQ/kg d.m.) (Ranges)	Ref.
Rathdrum, County Wicklow	Pasture, rural areas	1997	6	7.5 (2-13.3)	1
Rathdrum, County Wicklow	Pasture, rural areas	1998	7	1.3 (0.8-3)	2
Rathdrum, County Wicklow	Forest clearing, rural area	1997	1	4.8	1
Rathdrum, County Wicklow	Rural area	1998	1	1.3	2
County Cork	Remote open land	1993	a	1.7	3
County Cork	Open land, country area	1993	a	2.1-3.0	3
County Cork	Open land, city suburb	1993	a	4.5-4.7	3
County Cork	Open land, city centre	1993	a	8.6	3
Ballydine, Co Tipperary	Unknown	1997	8	0.3 (0.15-0.80)	4
Askeaton, Co. Limerick	Unknown	1995	6	1.1 (0.6-1.5)	5

n = Number of samples

a 14 samples were taken from four locations, but the actual location of the individual samples is not known

1: Moloney & Associates (1997); 2: Maloney & Associates (1998); 3: Forbait (1994); 4: AEA (1997); 5: Environmental Protection Agency (1995)

Table A 38: Ireland - Cow's milk

Location	Source of milk	Date	n	Median Conc. (pg I-TEQ/g fat) (ranges in brackets)	Ref.
National Survey	Regional creameries, background	1995	20	0.14-0.5 (0.21)	1
National Survey	Road tankers, vicinity of potential dioxin source	1995	12	0.13-0.51 (0.23)	1
Rathdrum, County Wicklow	Milking parlours	1997	3	0.48-0.49 (calculated from whole milk data)	2
County Cork	Farms across the county	1994	14	<1.3-<1.5	3

n = Number of samples

1: Concannon (1996); 2: Moloney & Associates (1997); 3: Forbait (1994)

A8 Italy

PCDD/PCDF concentrations were measured in the various environmental compartments in the early 1990s in Italy. Results were reported for urban air, soils, and foodstuffs (Berlincioni *et al.* 1995, Berlincioni *et al.* 1993, Berlincioni *et al.* 1992, Turrio-Baldassari *et al.* 1994).

A8.1 SOIL

No pesticide application or sludge spreading is known to have occurred in the past. All samples were from single pick-ups (no composite samples) (Berlincioni *et al.* 1995, di Domenico 1993). The 69 soil samples were randomly collected from a farming area (7 cm topsoil) neighbouring a large urban area. Only homolog concentrations were available, I-TEQ were calculated according to di Domenico *et al.* 1990. From the data obtained it was concluded that the TEQ-concentrations were well below the pertinent MTL⁶-level of 10 ng EPA-TEQ/kg d.m.

Table A 39: Italy - Soil (Berlincioni *et al.* 1995, di Domenico 1993). Concentrations in ng TEQ/kg d.m.

Type	State/City/Region	Type of location	Soil History	Date	n	Min	Max	Mean
Caves not visited by the general public	Latium, Abruzzi	Background		1992	6	0.057	0.12	
Pasture, from sea level up to 1300 m	Latium, Abruzzi, Piedmont, Tuscany	Background	no pesticide use or sludge spreading	1992	21	0.1	43	
Farming area	Tuscany	Neighbouring large urban areas	no pesticide use or sludge spreading	1993	69	1.9	3.1	2.4

A8.2 SEDIMENT

Sediment cores (10-30 cm) were taken from the lagoon of Venice. The sampling program was set up in connection with the search for potentially contaminated sites. A total of 31 samples were taken from different areas of the lagoon. Concentrations ranged from below 1 ng I-TEQ/kg d.m. in the open sea area and mixed urban areas up to 570 ng I-TEQ/kg d.m. close to the industrial point sources.

⁶ MTL = Maximum Tolerable Limit (normalised to EPA-TEQ as established in 1987) for risk assessment

**Table A 40: Italy – Sediments in the lagoon of Venice (di Domenico *et al.* 1998)
Concentrations in ng TEQ/kg d.m.**

Location	Type	Date	N	Min	Max
Venice, lagoon	Industrial point source	1992, 1995, 1997	8	12	570
	Urban		5	4.8	23
	Mixed industrial and urban		6	0.48	8.5
	Open fishing areas		6	0.35	1.8
	General environment, open sea		6	0.073	10

A8.3 AIR

So far, ambient air data are available only from two cities and from winter measurements. A total of 18 samples were analysed from Florence (Berlincioni *et al.* 1995, 1993, 1992) and 10 from Rome (Turrio-Baldassari *et al.* 1994). All samples were taken with a combination of glass-fiber filters with either XAD or PUF. The ambient air concentrations were comparable in the two cities with minimum concentrations of 48 and 72 fg I-TEQ/m³, respectively and maximum concentrations of 200 and 277 fg I-TEQ/m³, respectively. For Rome, a mean concentration of 85 fg I-TEQ/m³ and a median concentration of 62 fg I-TEQ/m³ was calculated.

Table A 41: Italy - Urban air (Berlincioni *et al.* 1995, 1993, 1992, Turrio-Baldassari *et al.* 1994). Concentrations in fg TEQ/m³

Sampling Location	Type of location	Sampling Period	n	Min	Max	Mean	Median
Florence	Urban	Jan.-Feb.1993	18	72	200		
Rome	Urban	Nov. 1990-Apr. 1991	10	48	277	85	62

A9 Luxembourg

Information regarding the concentrations of PCDD and PCDF in Luxembourg is relatively scarce. It has however been possible to gain information from Administration De L'Environnement in Luxembourg relating to some media. Concentrations of PCDD/PCDF have been obtained in ambient air, soil, sediments, spruce needles, moss and kale. No information was found on fresh water, sewage sludge, wildlife and waste.

A9.1 SOIL

A soil program, established by the Administration de l'Environnement in Luxembourg, investigated the concentrations of PCDD/PCDF at 69 locations throughout the country. The locations varied between rural agricultural locations to urban traffic affected sites. Sampling depths were 30 cm. The highest concentration measured was 20.4 ng TEQ/kg (Table A 42).

Table A 42: Luxembourg - Soil (ng I-TEQ/kg d.m.)

Site Type Description	PCDD/PCDF (ng I-TEQ/kg d.m.)
Rural Arable	1.40
Rural Pasture	1.40
Rural wood	5.95
Industrial/urban Garden/lawn	6.39
Urban Garden/Lawn/Park	4.26
Urban Traffic location	1.84
Rural Garden/lawn	3.59
Industrial/urban Pasture	2.85

A9.2 SEDIMENT

PCDD/PCDF concentrations in sediment were assessed in 1993 at four locations in Luxembourg. The samples were taken from the river Alzette near Schifflange in the south west, the river Alzette between Beggen and Walferdange in central southern Luxembourg, the river Sûre near Erpeldange in central Luxembourg and the river Moselle between Grevenmacher and Wasserbillig. The average concentration of PCDD/PCDF in sediment in the sampling sites studied is 9.4 ng I-TEQ/kg.

Table A 43: Luxembourg - Sediments

River and location	Concentration (ng I-TEQ/kg)
Alzette near Schifflange	10.1
Alzette between Beggen and Walferdange	2.4
Sûre near Erpeldange	9.3
Moselle between Grevenmacher and Wasserbillig	15.7

A9.3 AIR

Information has been gathered on ambient air in selected locations in Luxembourg (Environment Agency Luxembourg 1998). The sampling sites with data available are Esch/Alzette which is a city and is characterised by a steel producing industry, Luxembourg which is a city with urban and traffic zone, Dippach which is a rural location west of Luxembourg city and Vianden which is a rural location with no traffic or industry located in the north east of Luxembourg. The average concentration of PCDD/PCDF in the rural locations is 47 fg I-TEQ/m³.

Table A 44: Luxembourg - Air (fg I-TEQ/m³)

Location	Average 1992-94 (fg I-TEQ/m ³)
Esch/Alzette	77
Luxembourg City	54
Dippach	64
Vianden	30

A9.4 VEGETATION

There are results available for spruce needles, moss and kale. One year old spruce needles were collected at eight sites and analysed in 1993/94.

Table A 45: Luxembourg – Vegetation, here: spruce needles (ng I-TEQ/kg d.m.)

Area and description	Concentrations (ng I-TEQ/kg d.m.)
Esch/Alzette (City and steel producing industry)	3.5
Schiffange (City and steel producing industry)	7.9
at the west of frontier in the south of Luxembourg	2.0
at the west of frontier in the middle of Luxembourg	0.8
at the west of frontier in the north of Luxembourg	1.6
at the east of frontier in the south of Luxembourg	4.8
at the east of frontier in the middle of Luxembourg	2.3
at the east of frontier in the north of Luxembourg	3.1

In addition to the above study on spruce needles a study of the concentrations of PCDD/PCDF in moss and kale was carried out. The sites that were covered in the study were quite varied

- steel producing industry,
- waste incinerators,
- industrial zones,
- rural areas,
- urban areas and
- rural areas.

For the study relating to moss the moss samples were collected from natural positions. For each site, a volume of 4 litres of moss material was collected and prepared for chemical analysis. For the kale study the plants raised in a standard soil under greenhouse conditions. Selected plants were placed into flowerpots and transferred to the sites of investigation and exposed for two months. The concentration of PCDD/PCDF in natural moss in the background zones is 1.0 ng I-TEQ/kg and the average of the two rural sites is 0.7 ng I-TEQ/kg. The concentration of PCDD/PCDF in kale in the background zones is 0.7 ng I-TEQ/kg and the average of the two rural sites is 0.45 ng I-TEQ/kg (Table A 46).

Table A 46: Luxembourg - Vegetation (ng I-TEQ/kg d.m.)

Sampling Site	Type of Location	Mean in Moss (ng I-TEQ/kg)	Mean in Kale (ng I-TEQ/kg)
Differdange	Steel producing industry	2.2	1.1
Esch/Alzette Ramerich	Steel producing industry	2.4	2.3
Esch/Alzette Centre	Steel producing industry	2.6	106
Schifflange	Steel producing industry	4.5	9.7
Clemency	Background zone near steel producing basin	1.0	0.7
Leudelange	Waste incinerator	2.0	0.8
Bettembourg	Industrial zone	1.2	0.9
Wiltz	Industrial zone	0.9	0.4
Kockelscheuer	Near urban zone	1.3	1.5
Beckerich	Rural area	0.6	0.5
Osweiler	Rural area	0.8	0.4

A10 The Netherlands

Extensive research and monitoring has been done in the Netherlands concerning dioxins and other organic pollutants, and there is therefore a considerable amount of data available. A key source of data for this section of the report has been the recent doctoral thesis by Liem and Theelen (1997) of RIVM, which provides an overview of PCDD/PCDF concentrations in the Dutch environment.

A10.1 SOIL

Analysis of PCDD/PCDF in soil has been divided into two main areas: potential sources, such as MSWIs and background exposures. Three areas of local contamination near MSWIs, were studied in 1990 to 1991: the Lickebaert area, Zaanstad and Leeuwarden (De Jong *et al.* 1990 1991; Matthijsen *et al.* 1991). The Lickebaert area is in the vicinity of the largest waste incinerator in Rotterdam. Analysis in Lickebaert and Zaanstad was done in bulk samples representing different layers of the soil. In Leeuwarden ten sites were chosen at increasing distance from the source, with samples collected only from the top 0-5 cm layer. The results of all three surveys are shown Table A 47

It was found that concentrations of dioxins decreased with depth in the soil. This corresponds with the fact that dioxins are virtually immobile in soil and they, therefore, accumulate in the upper layers. Higher levels were found in the Zaanstad area than in Lickebaert. The very high concentrations were explained by irregularities in the process management of the Zaanstad plant.

Data for three further areas of contamination are available: a floodplain contaminated by river sediment (Hendriks *et al.* 1996), the Volgemeerpolder hazardous waste site (Heida *et al.* 1995), and scrap wire and scrap car incinerator sites near Amsterdam (Van Wijnen *et al.* 1992). The results of these analyses are also shown in Table A 47. Reference background contaminations were analysed at Bergambacht for comparison with those samples from the Lickebaert, Zaanstad and Leeuwarden areas of contamination. Significantly lower concentrations were found at the reference site. A wider survey of background concentrations across the Netherlands was also carried out in 1991 (Van den Berg *et al.* 1994). Thirty-two grassland sites were sampled at locations where there had been no tillage for 20 years. Concentrations were found to be similar to the lowest levels found in the above mentioned surveys of contaminated areas. An estimate for general background concentrations in Dutch soils was given at between 2-10 ng I-TEQ/kg d.m., with a maximum of 20 ng I-TEQ/kg d.m. (Van den Berg *et al.* 1994).

Table A 47: The Netherlands - Soil

Location	Type of location	Depth cm	Date	n	Median ng I-TEQ/kg d.m.	Ref.
Contaminated sites:						
Zaanstad	Near MSWI	0-2	1990-1991	5	13-252	1
		2-10	1990-1991	5	12-46	
		10-50	1990-1991	2	2-5	
Lickebaert	Near MSWI	0-1	1990-1991	5	18-51	1
		1-2	1990-1991	5	13-55	
		2-10	1990-1991	5	10-26	
		10-25	1990-1991	2	nd nd	
Leeuwarden	Near MSWI	0 – 5	1990-1991	10	2.6 – 22.6	2
??	Floodplain contaminated by river sludge	0-5	1993	3	17-78	3
7 Sites close to Amsterdam	Scrap wire and scrap car incinerator sites		1988	20	60 – 98000 (3750)	4
Volgemeer-Polder	Hazardous waste site	??	??	??	??	5
Background sites:						
Bergambacht	Rural	0-1	1990-1991	2	5-9	1
32 Sites across the country	Rural	0-5	1991	26	2.2-16.4 (4)	6

n = Number of samples

1: de Jong *et al.* (1991); 2: Matthijssen *et al.* (1991); 3: Hendricks *et al.* (1996); 4: Van Wijnen *et al.* (1992); 5: Heida *et al.* (1995); 6: van den Berg *et al.* 1994

A10.2 SEDIMENTS

Results of PCDD/PCDF measurements in sediments are available from Dutch rivers, canals, estuaries, seas, lakes and stagnant waters. Transboundary pollution is an important issue in the Netherlands, resulting from polluted water entering the country in rivers. Many measurements are, therefore, taken at frontier locations.

In a summary report, van Zorge (1998) gave typically concentrations for the various parts of the aquatic system as follows: sediments in rivers, lakes and canals usually fall in the range 1-10 ng I-TEQ/kg d.m, but concentrations up to 420 ng I-TEQ/kg d.m. have been detected. Contaminated stagnant waters have 650 ng I-TEQ/kg d.m, harbours up to 4,000 ng I-TEQ/kg d.m. Estuaries and sea sediments have ranges 8-21 ng I-TEQ/kg d.m. Concentrations in lakes and stagnant water, sediments in background locations, are considered to be approximately 1-10 ng I-TEQ/kg d.m. (Liem and Theelen 1997).

Based on chemical analysis of suspended loads in rivers in 1994, Evers *et al.* (1996) calculated annual transboundary loads of 15.3 g I-TEQ/a for the River Meuse at Eijsden, 65.7 g I-TEQ/a for the River Rhine, at Lobith, and of 11.9 g I-TEQ/a for the River Scheldt, at Schaar van Ouden Doel. These data can partly explain variations in levels of PCDD/PCDF in sediments in these rivers in the Netherlands. Concentrations in sediments in the River Rhine were generally higher than those found in sediments from the Rivers Meuse and Scheldt

(Evers *et al.* 1988; Turkstra and Pols 1989). A summary of the Dutch sediment concentrations is given in Table A 48.

Particularly high concentrations have been found in industrial harbours, such as Chemieharbour in Rotterdam, where there is a local vinylchloride monomer factory, and St Laurens Harbour, which has domestic and chemical waste incinerator nearby (see Table A 48). Further contamination has been found in a canal thought to have been polluted by an accident at a nearby herbicide production plant (Turkstra and Pols 1989).

Table A 48: The Netherlands - Sediments recorded 1980-1990. All concentrations in ng I-TEQ/kg d.m.

Location	Conc.	Location	Conc.
Rhine Basin		Harbours and stagnant waters	
Tolkamer	78	Eerste Petroleumhaven	250
Lobith	22	Derde Petroleumhaven	23
Lobith	4–180	Chemieharbour	4000
Tiel	120	St Laurens Harbour	2600
Dalem	220	Lake Ijsselmeer	91
Hollands Diep	12–17	Lake Veluwemeer	10
New Meuse	26-78	Ijmeerput	120
Nieuwe Waterweg	43–48	Haringvliet	120
		Lake Ketelmeer	72
Other Rivers and Canals		Estuaries and Sea	
Nordzeekanaal	10–420	Western Scheldt	15–15
Apeldornskanaal	260	Wadden Sea	8-21
River Meuse at Eisjen (frontier)	12	Ems Dollard	10
River Meuse at Roermond	82		
River Meuse at Grave	11		
Hollands Diep at Moerdijk	25		
Hollandse Ijssel	42–170		
Western Scheldt	28		
N.-Holl.kanaal	12		

Reference: as compiled by Liem and Theelen 1997; original sources: Turkstra and Pols (1986, 1987, 1989), Evers *et al.* (1988, 1993) and Beurskens *et al.* (1993)

A10.3 AIR

The Dutch ambient air sampling campaign included analysis of samples collected in the period May 1991 to July 1993, at four locations across the country. The characteristics of the sampling locations were as follows: Vlaardingen (industrial area 10 km north-east of two MSWIs), Witteveen (a rural area), Wijnandstrade (urban area near to the German and Belgian border), and Zegveld (in a large urban area). A total of 45 samples were taken. The results are presented Table A 49. An evaluation of the influence of wind direction showed that the

highest concentrations were measured in the proximity of the MSWI, with prevailing south west winds. The congener profile of PCDD/PCDF in air was often dominated by 1,2,3,4,6,7,8-Cl₇DD/Cl₇DF and Cl₈DD/Cl₈DF.

Table A 49: The Netherlands - Ambient air. Concentrations in fg I-TEQ/m³

Location	Type of Location	Sampling Time	n	Min	Max	Mean
Vlaardingen	Industrial, close to MSWI	May-Sep'91	10	6	140	62
Witteveen	Rural	Sep-Nov'92	12	9	63	31
Wijnandsrade	Urban	Mar-May'93	12	26	99	55
Zegveld	Urban	Jun-Jul'93	11	4	59	18

n = Number of samples

References: Bolt-Moekoet and De Jong (1992 1993a, 1993b and 1994); De Jong *et al.* (1996) cited in Liem and Theelen (1997).

A10.4 VEGETATION AND WILDLIFE

No data have been found for vegetation or wildlife.

A11 Portugal

No data were available from Portugal.

A12 Spain

Information on the concentrations of PCDD/Fs in soil, vegetation, and sewage sludge are available from several studies in Spain. We were unable to identify sources of information on PCDD/PCDF concentrations in air, sediment, water or waste. The information available is summarised below.

A12.1 SOIL

There have been several studies into the concentrations of PCDD/PCDF in soil around the waste incinerators in Spain. However except for background samples taken as part of such studies we have been unable to identify surveys of background concentrations in soils away from potential point sources.

Baseline concentrations of PCDD/PCDF were studied in detail around a proposed hazardous waste incinerator at Constanti in Catalonia before it was constructed (Schuhmacher *et al.* 1997). Samples were taken at forty sites around the proposed site, thirty of these sampling positions were in rural locations and the remaining ten were considered to be within urban areas. The sampling took place within a 7 km radius of the proposed site. At each sampling sites soil was taken from the upper 5 cm of soil. Litter and other interfering materials were removed prior to collection. The results are shown in Table A 50.

Table A 50: Spain – Soils close to a proposed incinerator (ng TEQ/kg d.m.)

Type	Mean	Standard Deviation	Median	Max.	Min
Rural	0.84	1.47	0.54	8.40	0.08
Urban	4.40	7.51	1.56	24.20	0.63

In another study, soil samples were analysed in an area around a clinical waste incinerator in Madrid (Jimenez *et al.* 1996) . The clinical waste incinerator is located 15 km east of Madrid (Spain). In this area is also located the rubbish dump of Madrid, scrap recovery industries and a busy road network. In total 16 surface (0-5 cm) soil samples were taken at various distances from the incinerator in 1993. Two of these samples were background samples, located 4.5 km away from the clinical waste incinerator. The concentrations at these two sites were 0.71 and 0.69 ng I-TEQ/kg d.m.

- The PCDD/PCDF concentrations in Spanish soils vary between 0.08 ng/kg I-TEQ in some rural sites to 24.20 ng/kg I-TEQ in some urban sites.
- In Catalonia the average PCDD/PCDF concentration in rural areas is 0.84 ng/kg I-TEQ and in urban areas in the same region the average is 4.40 ng/kg I-TEQ.
- The PCDD and PCDF concentrations around Madrid area are less representative as only two samples (possibly both rural) were taken the average of these was 0.70 ng/kg I-TEQ.

A12.2 VEGETATION

The studies of vegetation were performed in areas around waste incinerators. The majority does not give representative figures for background concentrations. There has however been a study similar to that of soil (Schuhmacher *et al.* 1997) around an incinerator under construction which related to vegetation.

The study into the concentrations of PCDD/PCDF in vegetation was carried out whilst an incinerator was under construction in the Catalonia region of Spain (Schuhmacher *et al.* 1998). The study consisted of 40 sampling sites of which 30 were rural and the remaining ten were urban. The forty samples of *Bouteloua gracilis* were taken within a 7 km radius of the incinerator that was under construction. The results are shown in Table A 51. The average for all sites is 0.61 ng/kg I-TEQ, the average concentrations of PCDD/PCDF in the rural and urban areas are 0.53 and 0.86 ng I-TEQ/kg, respectively. The difference between the urban and rural concentrations is statistically significant ($p < 0.01$).

Table A 51: Spain - Vegetation (ng I-TEQ/kg d.m.)

Concentration	Mean	Median	Max.	Min
All samples	0.61	0.53	1.22	0.24
Mean for urban areas	0.86			
Mean for rural areas	0.53			

A12.3 SEWAGE SLUDGE AND SEDIMENTS

There is limited information into the concentrations of PCDD/PCDF in sewage sludge in Spain. A study reported concentrations of PCDD/PCDF in sewage sludge in Catalonia (Eljarrat *et al.* 1998). Samples were taken from ten rural and urban waste water treatment plants. The average PCDD/PCDF concentrations in contemporary sewage sludge was 64.3 ng/kg I-TEQ dry weight. The PCDD/PCDF concentrations in sediments were between 0.21 and 57.04 ng I-TEQ/kg d.m. at the sites studied. The average concentrations at the sites studied are shown in Table A 52.

Table A 52: Spain - Sediments (ng TEQ/kg d.m.)

* The Besós river corresponds to the sewage sludge dumping site

Site	ng I-TEQ/kg
Llobregat river	3.66
Ebro river	2.08
Venice lagoon	9.74
Orbetello lagoon	2.43
Besós river*	57.04
Sediment – Mean without dumping site	4.48

A13 Sweden

The Swedish EPA maintains the Dioxin database, which contains the results of several hundred PCDD/PCDF analyses. This was the first dioxin database set up. The samples date back to 1970 but unfortunately there is no newer data than from 1993 included. This major drawback limits the usefulness of the data. This section deals with the evaluation of the data contained in the Swedish database. However, we are aware that there are more and more recent results of PCDD/PCDF analyses available from the Swedish environment. Within this project, these data could not be evaluated individually. Task 9 contains some results obtained from the published literature, which deal with time trends.

A13.1 SOIL

There are the PCDD/PCDF concentrations from 20 soil samples; all samples are dated in 1990. All samples were related to three special cases where accidents might have led to soil pollution. At a chemical manufacturing site, where in the past chloralkali electrolysis was performed, the concentrations ranged from 5.3 to 11,446 ng N-TEQ/kg d.m. This range clearly indicates that the operation of the plant caused a major pollution problem. The remainder two groups of samples were directed to the impact on soils after PVC fires. In both cases, no impact on the soils could be determined. As can be seen from Table A 53, the minimum and maximum concentrations were in a very narrow range. The soil concentrations were all below 1 ng N-TEQ/kg d.m.

Table A 53: Sweden – Soil. All concentrations in pg N-TEQ/g d.m.

	Minimum	Maximum	Mean	Median
PVC fire, Ronne	0.12	0.79	0.55	0.55
Background site, Ronne	0.47			
PVC fire, Valdemarsvik	0.11	0.33	0.17	0.17
Chloralkali plant	5.3	11,446		

A13.2 SEDIMENT

The Swedish database contains approximately 100 results for sediments, which includes bottom sediments of rivers, lakes, a canal, the Baltic Sea, and the Arctic Sea as well as sediment cores from polluted and unpolluted sites (Table A 54). Lastly, seston samples were generated as well.

In general, the concentrations in the sea sediments were lower than in the lake and river sediments: concentrations above 200 ng TEQ/kg d.m. (207 pg N-TEQ/kg d.m.) were detected in the free port of the Göta river. Other hotspots identified were a site affected by a chloroalkali plant which had PCDD/PCDF concentrations up to a maximum of 1,692 ng N-

TEQ/kg d.m. Along the Dalsland canal the PCDD/PCDF concentrations ranged from almost non-detectable to 828 ng N-TEQ/kg d.m.

Table A 54: Sweden – Sediments. All concentrations in pg N-TEQ/g d.m.

	Minimum	Maximum	Mean	Median
Sea Sediments	0.8	82.2	10.4	6.4
River/Lake Sediments	1.0	207.5	19.5	9.9

The concentrations found in a core from the Baltic Sea is shown in Figure 2. It can be seen that the PCDD/PCDF concentrations varied with time indicating that some years ago, the input of dioxins and furans into this location was higher than in 1988 when the core was taken. Much earlier in time, the input was very low indicated by the low concentrations. Over the years the concentrations of PCDD/PCDF increased up to 58 ng N-TEQ/kg d.m. Although the concentrations started to decrease to almost 50% of the highest concentrations the low concentrations as found in the deeper layers have not been reached.

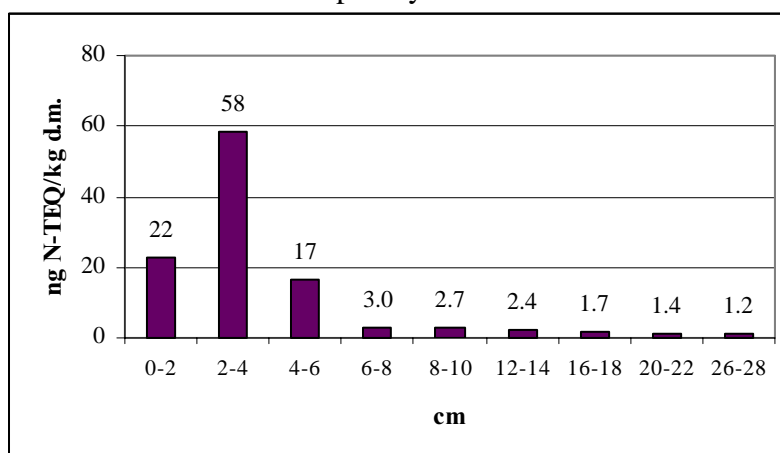


Figure 2 Sweden – Sediment core from the Baltic Sea. Concentrations in ng N-TEQ/kg d.m.

A13.3 AIR

There were 33 ambient air data available from high volume samplers. All samples were analysed for particles and gaseous phase; most of these were composites of a 48 hours sampling period. The time covered was June 1988 until February 1993. Except for a few unspecified samples, all sampling locations were urban. Concentrations ranged from 0.16 to 53.7 fg N-TEQ/m³; the mean of all samples was 10.4 fg N-TEQ/m³. The minimum, maximum and mean concentrations are summarised in Table A 55.

Table A 55: Sweden – Ambient air samples. All concentrations in fg N-TEQ/m³

Sampling Date		Minimum	Maximum	Mean
Summer 88	Urban	14.9	19.9	17.7
Winter 89	Not specified	27.6	53.7	44.5
Spring 90	Urban	0.16	29.2	6.5
Summer 90	Urban	0.43	4.5	1.8
Fall 90	Urban	0.87	10.0	4.3
Fall 91	Not specified	5.4	23.3	14.4
Winter 91	Urban	1.0	22.1	9.8
Winter 93	Urban	5.1		
Urban		0.16	29.2	6.5
Not specified		5.4	53.7	32.4

A13.4 WILDLIFE

A13.4.1 Fish, Shellfish, and Aquatic Mammals

Within the Swedish database, by far, aquatic organisms were the most abundant samples analysed for PCDD/PCDF. Overall, the database contains 481 results of which 347 are from muscles, 47 from blubber, 43 from liver, and 20 from hepatopancreas. 20 Samples were undefined tissue and 4 were water samples.

A summary of all fish data contained in the Swedish EPA database is shown in Table A 56. It can be seen that there is a wide range of species analysed. Consequently, the range of PCDD/PCDF concentrations was wide, too. Most samples were from herring (n=184), which had a mean concentration of 3.9 pg N-TQE/kg f.w. Astonishingly high concentrations were found in liver of burbot (maximum = 158 pg N-TEQ/kg f.w.) and pike (maximum = 154.4 pg N-TEQ/kg f.w.).

An overview of all shellfish samples is shown in Table A 57 and on all aquatic mammals in Table A 58. Generally, the concentrations on a fresh weight basis are much higher in hepatopancreas than in muscle. Maximum concentrations higher than 300 pg N-TEQ/kg f.w. were determined in crab and lobster. For the aquatic mammals, the highest tissue concentrations were in blubber with a maximum of a ringed seal with 216 pg N-TEQ/kg f.w.; mean concentrations were normally a few pg N-TEQ/kg f.w.

The database contains one sample with a concentration of 0.07 pg N-TEQ/kg f.w., which is not further specified.

Table A 56: Sweden – Fish. All concentrations in pg N-TEQ/g f.w.

Sample-type	Type	n	Minimum	Maximum	Mean
Arctic char	Muscle	9	0.01	7.8	4.8
	Undefined	1	11.1	0.01	
Burbot	Liver	23	3.8	158.1	77.7
	Muscle	6	0.05	1.7	0.9
Cod	Liver	3	30.3	45.3	40.3
	Muscle	3	0.34	0.38	0.4
	Undefined	1	14.0		
Herring	Muscle	184	0.07	20.1	3.9
Mackerel	Muscle	1	2.8		
Pike	Liver	15	7.9	154.4	40.8
	Muscle	112	0	2.8	0.5
Plaice	Muscle	3	0.22	0.45	0.35
Salmon	Muscle	2	2.8	3.2	3.0
	Undefined	6	8.2	67.7	26.9
Trout	Muscle	11	0.23	11.8	6.3
Whitefish	Muscle	5	0.09	8.4	4.4
Whitefish-bleak	Muscle	2	1.4	2.1	1.7

Table A 57: Sweden – Shellfish. All concentrations in pg N-TEQ/g fresh weight

Sample-type	Tissue	n	Minimum	Maximum	Mean
Crab	Hepatopancreas	13	1.9	336.2	67.6
Cray fish	Hepatopancreas	3	2.9	8.2	5.1
	Muscle	1	0.12		
Crustaceans, misc.	Undefined	2	9.8	10.1	9.9
Lobster	Hepatopancreas	2	17.2	380.1	198.6
Mussel	Undefined	5	0.09	0.58	0.24
	Water	4	0.3	0.51	0.41

Table A 58: Sweden – Aquatic mammals. All concentrations in pg N-TEQ/g fresh weight

Sample-type	Tissue	N	Minimum	Maximum	Mean
Common Porpoise	Blubber	18	0.78	5.8	2.0
Common Seal	Blubber	8	5.6	25.7	11.5
Crabeater Seal	Undefined	4	0.78	2.0	1.1
Grey Seal	Blubber	7	3.1	22.8	12.9
	Liver	1	6.5		
Harp Seal	Blubber	1	5.5		
Otter	Muscle	6	0.04	0.68	0.4
Ring Seal	Blubber	13	6.3	216.9	66.0
	Liver	1	65		

A more specific study shows the geographic distribution of herring: fifty homogenates of herring (pooled samples) from different collection sites along the Swedish coast of the Baltic Sea were analysed. Most of these were taken close to the coast, one was taken in the middle of the Bothnian Sea. The PCDD/PCDF concentrations were very similar from north to south. There seemed to be a slight trend towards higher concentrations along the coast of Norrland where numerous pulp mills are located. Pulp mills are potential sources of PCDD/PCDF and the contamination in herring was supported by the finding that these samples contained higher relative concentrations of Cl₄DD and Cl₄DF. The sample from the middle of the Bothnian Sea contained 8.7 N-TEQ/g f.w. The herring collected from Kattegatt and Skagerrack had much lower concentrations. A summary is given in Table A 59

Table A 59: Sweden - Herring

Area	Pg N-TEQ/g f.w.		Pg N-TEQ/g fat	
	Average	Range	Average	Range
Entire Baltic Sea	9.5	2.1-20	150	27-420
Norrlands Coast	12	5.4-20	160	88-260
Kattegatt / Skagerrack	1.9	1.8-2.1	24	9.1-52

A13.4.2 Terrestrial Animals

A summary of analytical results from terrestrial wildlife stored in the Swedish Dioxin Database is shown in Table A 60; all concentrations are in N-TEQ/kg on a fresh weight basis. There were many different tissues analysed from a variety of animals. However, the value of the data is somewhat limited as most data were generated in the 1980s when a) the analytical capabilities were limited and b) some data seem to target high exposure scenarios. The cow's milk results with a mean concentration of 22 pg N-TEQ/g f.w. may serve as an example for the latter statement. Although somewhat lower, the PCDD/PCDF concentrations in cream with a mean of 5 pg N-TEQ/g are high in comparison the results from other EU countries. On the other hand, the results for butter with a mean of 0.28 pg n-TEQ/g are low and reflect the more rural character of Sweden having a less densely population than other Member States if the EU.

Table A 60: Sweden – Terrestrial. All concentrations in pg N-TEQ/g fresh weight

Matrix	Tissue	n	Year	Concentration	Overall Mean
Badger	Adipose	1	1989	2.08	1.8
		1	1990	1.45	
Chicken	Egg	7	1993	0.08	0.32
	Fat	4	1989	0.32	
Cow	Adipose	3	1989	0.36	0.23
		2	unknown	0.04	
	Kidney	2	1989	0.1	0.07
	Liver	2	1989	0.08	
	Milk	10	unknown	0.06	
	Muscle	2	1989	0.01	
Dairy	Butter	5	1989	0.28	5.1
	Cream	1	unknown	5.1	
Flycatcher	Muscle	1	1987	1.4	0.075
Moose	Muscle	1	1986	0.13	
		1	1987	0.02	
Osprey	Tallow	1	1983	2.2	
	Egg	1	1982	11.5	
		2	1982	18	
	Muscle	3	1987	6	
	1	1988	5.1		
3	1991	13.8			
Pig	Adipose	5	1989	0.26	0.06
Rabbit	Muscle	10	1992	0.06	
Red Fox	Muscle	1	1988	0.06	0.06
		1	1989	0.06	
Reindeer	Tallow	1	1983	0.33	1.1
		1	1984	0.41	
		4	1987	2.1	
		3	1988	0.86	
Sheep	Adipose	2	1989	0.37	143.6
White-tailed Eagle	Egg	2	1985	330	
		6	1989	81.1	
Wolf	Muscle	3	1986	0.47	

A13.5 SEWAGE SLUDGE

The Swedish Dioxin Database lists the results of 55 results for sludge analyses. All samples have been collected between 1989 and 1993. The municipal sewage sludges were all from urban areas. The results for the dry cleaning shops refer to the concentrations found in the stillbottoms and thus, have to be categorised as industrial residues or wastes. The category of “industrial” samples contained several samples from slaughterhouses. From the data displayed in Table A 61 it can be seen that most samples contained PCDD/PCDF concentrations around 20 ng N-TEQ/kg d.m. The concentrations in the pulp sludges were not

higher than those samples with now point source nearby. More than 100 ng N-TEQ/kg d.m. have been found in sedimentation basins from the metal (ferrous and non-ferrous) industry as well as from the textile industry. The stillbottoms from dry cleanings and from chlorine production with graphite electrodes generated high concentrations of PCDD/PCDF as well. The highest concentrations were reported for a sludge sample from mercury distillation, which gave 1,777 ng N-TEQ/kg d.m., and from dry cleaning with 974 ng N-TEQ/kg d.m.

Table A 61: Sweden – Sludges. All concentrations in ng N-TEQ/kg d.m.
* sedimentation basin, ** stillbottoms or crusts

Type	N	Minimum	Maximum	Mean
Municipal	12	1.44	115	19.8
Domestic	2	5.71	11.8	8.74
Farm	1	0.02		0.02
Pulp mill	5	2.4	53.8	26.6
Chemical industry	3	2.52	5.42	3.58
Mercury	1		1,777	1,777
Coal	1	0.33		0.33
Industrial inputs	4	0.16	12.2	4.83
Metal *	4	2.72	440	116
Textile industry *	5	16.8	207	84.9
Graphite electrodes**	4	29.0	376	238
Dry cleaning **	13	0.12	974	146
Total	55	0.02	1,777	108

A13.6 WATER AND EFFLUENTS

73 Data from various water samples are presently compiled in the Swedish Dioxin Database. The results are summarised according to major categories in Table A 62. It should be noted that several samples contained particles, which adsorb the PCDD/PCDF from the water phase. For all categories minimum concentrations were close to the detection limit or far below 0.1 pg N-TEQ/L. The high concentration of 221 pg N-TEQ/L is from the cable industry and the 133 pg N-TEQ/L is from a recycling paper mill. In the effluents from another recycling paper mill, however, 0.34 pg N-TEQ/L were determined. More than 10 pg N-TEQ/L were detected in the run-off from the chloralkali industry. Leachates from dumps were from below 1 pg TEQ/L to approximately 10 pg TEQ/L. In the effluents from laundry and dishwashers, the concentrations were in a close range around 1 pg N-TEQ/L; the highest concentration was 3 pg N-TEQ/L.

Table A 62: Sweden – Water and effluents. Concentrations in pg N-TEQ/L

Type	n	Minimum	Maximum	Mean
Effluent	6	0.01	27.1	10.7
Waste water treatment plant	18	0.04	89.5	14.4
Leachate	9	0.28	20.9	6.10
Pulp mill	18	0.11	133	19.1
Miscellaneous	22	0.08	221	23.5
Total	73	0.01	221	16.9

A14 United Kingdom

A14.1 SOIL

Data on PCDD/PCDF concentrations in UK soils are dominated by localised surveys in the vicinity of sources of contamination. There are few data on background locations.

An early soil survey of the UK was undertaken by Her Majesty's Inspectorate of Pollution (HMIP, now part of the Environment Agency) on a 50 km grid across the UK between 1985 and 1989 (HMIP, 1989; Creaser *et al.* 1989). 78 samples of rural soil were collected on this basis and a further 13 samples were collected from urban locations in London and Birmingham. The analysis of these soils was not isomer specific and hence I-TEQ values were not calculated. A re-examination of the chromatograms was undertaken at a later date, using 11 rural samples and 5 urban samples (HMIP, 1995). The results of this re-analysis, shown in Table A 63, indicated a clear difference between the concentrations in rural and urban soils, with the mean concentration in urban soils being over five times higher than that for rural soils.

A similar survey was undertaken in Northern Ireland (Caulfield and Ledgerwood, 1989) but, again, congener-specific analysis was not performed and so I-TEQ data are not available. Comparison of the detailed congener results shows that the concentrations were lower than those in the HMIP survey, reflecting the more rural nature of Northern Ireland in comparison to the UK mainland.

Table A 63: UK - Soils (ng I-TEQ/kg)

Survey	Type of soil / source of contamination	n	Min	Max	Mean	Median	Reference
HMIP UK survey	rural locations	11	0.78	17	5.2	-	HMIP (1995)
HMIP UK survey	urban locations	5	4.9	87	28	-	HMIP (1995)
Kirk Sandall, Doncaster	Baseline study for proposed chemical waste incinerator	12	3	20	8	7	Stenhouse and Badsha (1990)
Walsall	Vicinity of secondary non-ferrous refiners	103	1.0	209	35	19	Fernandes <i>et al.</i> (1994)
Hampshire	Vicinity of 4 MSW incinerators	51	2.4	160	19	11	Abbott <i>et al.</i> (1997)
Panteg, South Wales	sites close to hazardous waste incinerator	16	13.3	1,585	230*	-	Ball <i>et al.</i> (1993)
Panteg, South Wales	sites further from incinerator	26	2.5	14	5.8*	-	Ball <i>et al.</i> (1993)
Bolsover	Vicinity of Coalite Works	46	3.3	680	29	9.5	Environment Agency (1997b)
Bolsover	Vicinity of Coalite Works	9	5.8	90	32	22	MAFF (1992)

* mean of mean concentrations for various groups of samples given in reference

Table A 63 also shows the results of the various localised soil surveys in the UK. The Kirk Sandall survey was undertaken as a baseline study at the proposed location for a chemical waste incinerator (Stenhouse and Badsha 1990). The site is mixed urban and industrial. The results show similar concentrations to those in the UK rural survey, with lower values than expected for this type of location. These were explained by the low organic content and sandy nature of the soil.

The Walsall survey (Fernandes *et al.* 1994) was initiated because a relatively high PCDD/PCDF contamination was found in the soil in the UK wide survey. The local council commissioned a survey of the soils across the borough to consider the impact of the three non-ferrous refiners. The result showed a wide range of concentrations, and a considerable difference between the mean and median values. This indicated that a few high values were causing the mean to be higher. The refiners are in the west of the borough, and the levels in this region were, on average, a factor of two higher than in the east. The highest concentrations were twice as high as found in the UK urban survey.

The Hampshire soil survey was also commissioned as a follow-on study after relatively high levels were found there in the UK survey (Abbott *et al.* 1997). The survey was concentrated around the four municipal waste incinerators in the county. The results showed that the concentrations in the vicinity of the MSW incinerators were above those expected in rural locations, but are mostly comparable to urban locations. There were some outliers found, with a maximum concentration at 160 ng I-TEQ which was twice the urban maximum. Again there was considerable difference between the mean and median results, showing the skew in the distribution of concentrations.

The Panteg monitoring project (Lovett *et al.* 1998) involved the analysis of soil samples taken from sites surrounding the Rechem hazardous waste incineration plant in Pontypool, South Wales (Ball *et al.* 1993). The highest concentrations were found at two locations close to the plant. These were very high, with a maximum of 1,585 ng I-TEQ/kg. At the sampling locations further from the incinerator the concentrations were similar to those found in the HMIP survey.

The Environment Agency survey around the Coalite Works in Bolsover in 1992 (Environment Agency 1997b) has already been described in relation to the air samples taken. The soil survey showed that there were elevated concentrations very close to the works, but PCDD/PCDF concentrations decreased to background levels at increasing distance from the Works. All the samples in the survey were from locations within 5 km of the Works. The Ministry of Agriculture Fisheries and Food (MAFF) analysed soil samples taken from farms surrounding the Bolsover plant in 1992 (MAFF 1992). The concentrations are also shown in Table A 63. These concentrations were similar to those found by the Environment Agency, but with a much lower maximum value. Further analysis has been undertaken recently at the Bolsover site (Holmes *et al.* 1998). Soils were sampled in 1997 at some of the same sample locations as used in the 1992 Environment Agency survey. Generally there was no change found in the concentrations, confirming the persistence of these contaminants in soil.

Time trends of soil concentrations are discussed in Task 6 – Time Trends.

A14.2 SEDIMENTS AND FRESHWATER SYSTEMS

There have been relatively few studies of concentrations of PCDD/PCDF in sediments in the UK. An important survey of water and sediments in England and Wales was undertaken in 1992 (Rose *et al.* 1994a), funded by the National Rivers Authority (NRA), which is now part of the Environment Agency. There have also been a number of studies of sediments in lakes (Brzuzy and Hites 1995; Rose and McKay 1996; Rose *et al.* 1997). Concentrations in sediments are generally higher than those in soils.

A14.2.1 Freshwater and River Sediments

In the NRA study, freshwater and sediment samples were collected to determine the distribution of PCDD/PCDF in surface freshwater in England and Wales (Rose *et al.* 1994a; 1994b). Water was sampled at 40 sites, and sediment at 36. The concentrations of PCDD/PCDF in water were very low, at less than 0.08 ng I-TEQ/l. In all samples the concentrations were close to or below the analytical limits of detection. Concentrations in the sediments ranged from 2-123 ng I-TEQ/kg d.m., with a median concentration of 16.7 ng I-TEQ/kg d.m.. The results are given in Table A 64.

Table A 64: UK - Sediments in Rivers in England and Wales. Concentrations in ng I-TEQ/kg d.m.

Sampling location	Conc.	Sampling location	Conc.
Background' locations		Near to point sources	
New River, Wicken	73.01	Mag Brook, Honley (below)	51.11
River Loxley, near Sheffield	14.95	Pressbrook, Ogston	9.07
River Thames, Cricklade	5.84	River Aire, Beal (above)	23.33
River Derwent, Froggatt	5.61	River Aire, Beal (below)	35.36
River Dane, Hugbridge	4.21	River Aire, Fleet Weir (above)	24.2
River Lathkill, Alport	2.76	River Aire, Fleet Weir (below)	13.5
		River Amber, Ambergate (above)	8.96
		River Amber, Ambergate (below)	80
Agricultural Catchments		River Calder, Mirfield (above)	44.21
River Granta, Hildersham	7.82	River Calder, Mirfield (below)	6.45
River Lugg, Hereford	1.99	River Cam, Hauxton (above)	26
		River Cam, Hauxton (below)	7.78
Urban catchments		River Derwent, Belper (above)	19.77
R Alt, Altmouth	122.81	River Derwent, Belper (below)	13.74
River Mersey, Howley Wier	101.51	River Derwent, Church Wilne (above)	70.11
River Weaver, Acton	18.4	River Derwent, Church Wilne (below)	16.54
River Thames, Caversham Weir	13.23	River Don, Rotherham (above)	50.93
River Thames, Tedington	12.28	River Don, Rotherham (below)	25.65
		River Erewash, Eastwood (above)	8.39
		River Erewash, Eastwood (below)	11.99
		River Spenn, Dewsbury (below)	48.74
		River Tame, Lea Marston (below)	16.77
		R. Tame, Lea Marston (above)	32.1

The sampling locations were chosen to represent a variety of catchments, with samples taken in rural ‘background’ locations, agricultural catchments, urban areas and in industrial areas. In general, concentrations of PCDD/PCDF were higher in industrial and urban areas (range of 6.5-122.8 ng I-TEQ/kg d.m.) than at the ‘background’ sites (2.8-73 ng I-TEQ/kg d.m.), although one background location showed higher concentrations than some of the industrial and urban sites. Excluding two of the higher concentrations recorded at the ‘background’ locations, the study showed that the general background concentration of PCDD/PCDF in UK river sediments is < 6 ng I-TEQ/kg d.m. Agricultural sites had low concentrations (2-7.8 ng I-TEQ/kg d.m.). In several cases, samples taken above discharge points appeared to be higher than those downstream, indicating that unidentified sources may be present, or that sample sites were not ideally located.

Data is also quoted in the NRA report for locations on the River Doe Lea, into which flows effluent from the biological treatment works for the waste water from the Bolsover Coalite works. The concentration in sediments immediately below the works was 45,300 ng I-TEQ/kg d.m., and at about 1.5 km downstream was 7,410 ng I-TEQ/kg d.m. (as quoted from NRA Yorkshire region by Rose *et al.* (1994a)).

A14.2.2 Lake Sediments

Rose and McKay (1996) describe a study to investigate the fate of airborne PCDD/PCDF deposited onto freshwater systems. The study involved the analysis of sediment and fish samples from four UK lakes: Eleven Acre Lake and Marsworth Reservoir in central England, Lough Neagh in Northern Ireland and Dry Loch in Scotland. The lakes are in background locations away from major atmospheric and industrial sources of PCDD/PCDF, except for a sewage treatment works effluent entering Lough Neagh. Surface sediments of 3 cm in depth were taken from each lake, and 1 m cores were collected from Eleven Acre Lake and Lough Neagh.

The concentrations in the sediments ranged from 5-100 ng I-TEQ/kg d.m. (Table A 65), which are within the same range as those found in the river sediments above (Rose *et al.* 1994a). The homologue profiles were dominated by the higher chlorinated PCDD, as usually occurs where the main source is atmospheric. However, there was a dominance by Cl₈DF in Dry Loch, which could be the result of atmospheric transport from regional industrial pollution. The variations in concentrations between the lakes are likely to be as a result of the differences in lake and catchment size. The higher concentration in the Marsworth Reservoir was not explained, but it is likely that a local source was contributing to the contamination.

Table A 65: UK – Sediments in lakes. Concentrations in ng I-TEQ/kg d.w.

Lake	Concentration	Reference
Eleven Acre Lake	6	Rose and McKay (1996)
Lough Neagh, Northern Ireland	21	Rose and McKay (1996)
Dry Loch, Scotland	42	Rose and McKay (1996)
Marsworth Reservoir	92	Rose and McKay (1996)
Loch Coire nan Arr, Scotland	3.4	Rose (1996)

A14.3 AIR

The UK Department of the Environment, Transport and the Regions maintains a monitoring network to determine atmospheric concentrations of toxic organic micropollutants (TOMPs). These include polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans, polychlorinated biphenyls and polycyclic aromatic hydrocarbons. Data have been collected from five urban sites and three rural sites between 1991 and 1997 (NETCEN 1998).

Initially, the number of sampling sites was limited to four, at which TOMPS were measured in ambient air and deposition. These were to be the first long-term measurements of the atmospheric concentrations of PCDD/PCDF undertaken in the UK. As the concentrations of PCDD and PCDF were not known, but were expected to be extremely low, it was decided that sampling should take place near to the known sources which were predominantly urban industries. Three urban sites were selected to be representative of large conurbations in different parts of the UK (London, Manchester and Cardiff), with a fourth site in a town with light industry (Stevenage). After the initial measurement programme demonstrated that the pollutants were detectable at Stevenage, and that it was not an acceptable low concentration site, a semi-rural site was established at Hazelrigg, near Lancaster. The Stevenage site ceased operation and was replaced by a further urban site in Middlesbrough. Measurements later ceased also at the Cardiff site. Two new rural sites were commissioned in 1997 at Stoke Ferry and High Muffles, to extend the number of measurements made. All the urban samplers were located on the roofs of buildings in, or near, the urban centres. Samples are presently collected over fourteen day periods throughout the year.

Results are shown in Table A 66 below, for all locations. The ambient air data shows a clear difference between the urban and rural sites, with median values ranging from 17-103 fg I-TEQ/m³ in urban areas, and 6-12 fg I-TEQ/m³ in rural areas. Maximum values were highest in Stevenage and Cardiff, in samples taken in 1992. These higher concentrations may be evidence of a time trend rather than a geographical, or of uncertainty in early analysis techniques. The limits of detection in 1992/93 were relatively high, as shown by the difference between the estimated concentrations where non-detects were assumed equal to the detection limit and zero. The deposition data in this study showed no pattern.

During the period over which these measurements were taken, the UK's Environmental Protection Act (1990) was implemented. This led to increasingly stringent controls on industrial emissions. As a result, all of the older municipal solid waste incinerators, which were significant emitters of PCDD/PCDF, closed by the end of 1996. Three of the measurement sites; in Manchester, London and Middlesbrough, are within a few miles of municipal waste incinerators.

Table A 66: UK - Ambient air and deposition (NETCEN 1998)

	Location	Sampling period	n	Min	Max	Mean	Median	Upper Quartile	Lower Quartile
Ambient air (fg I-TEQ/m³)									
Rural	High Muffles	Q1 1996 -	2	1	10	6	6	8	4
		Q3 1997		(2)	(12)	(7)	(7)	(9)	(5)
	Hazelrigg	Q1 1996 -	4	2	24	12	10	18	4 (5)
		Q3 1997		(3)	(34)	(15)	(11)	(21)	
	Stoke Ferry	Q1 1996 -	2	2	21	12	12	17	7
		Q3 1997		(3)	(25)	(14)	(14)	(20)	(9)
Urban	Cardiff	1992	22	10	404	135	103	146	77
				(115)	(410)	(207)	(118)	(231)	(160)
	London	Q1 1996 -	4	7	37	19	17	22	14
		Q3 1997		(8)	(99)	(41)	(29)	(54)	(16)
	Manchester	Q1 1996 -	4	52	97	74	72	87	59
		Q3 1997		(55)	(112)	(82)	(81)	(103)	(60)
	Middlesbrough	Q1 1996 -	4	13	60	34	32	44	21
		Q3 1997			(89)	(42)	(34)	(54)	(22)
	Stevenage	start 1992 -	7	0	810	256	70	425	30
		9/4/92		(70)	(830)	(301)	(120)	(450)	(95)
Deposition (pg I-TEQ/m² day)									
Rural	Hazelrigg	Jan - October	9	0	517	81	2	82	0.2
		1993		(57)	(537)	(137)	(75)	(98)	(75)
Urban	Cardiff	Jan - August	6	10	182	62	43	81	11
		1992		(87)		(118)	(107)	(133)	(88)
	London	Jan - July	5	1	33	8	1	5	1
		1993		(68)	(89)	(78)	(77)	(80)	(76)
	Manchester	Jan - June	6	11	59	28	25	27	23
		1993		(99)	(275)	(153)	(123)	(179)	(105)
	Middlesbrough	Jan - June	6	6 (79)	12	52	45	78	18
		1993			(174)	(114)	(101)	(136)	(88)
	Stevenage	Jan - April	3	0.4	312	105	3	157	1
		1992		(26)	(334)	(130)	(31)	(183)	(29)

n = number of samples

Data given using the assumption that non-detects=0, the value in brackets was calculated using non-detects=detection limit.

Further data for a rural location is provided by Lee *et al.* (1998). This study reported data from two separate sampling series from the Hazelrigg site in the north-west of England outside Lancaster. The concentrations in the autumn of 1995 ranged from 3.0 to 54 fg I-TEQ/m³ with an average of 14 fg I-TEQ/m³ and a median of 11 fg I-TEQ/m³. However, the high values of 47 and 54 fg I-TEQ/m³ were unusual, as they occurred on 6 November 1995, coinciding with Bonfire Night celebrations, when it is customary to light fireworks and bonfires. The mean and median concentrations excluding these two results are 8 and 6 fg I-TEQ/m³ respectively. The mean and median of concentrations in the summer of 1996 were 6 and 5 fg I-TEQ/m³ respectively. These concentrations compare well with the TOMPS results (NETCEN 1998).

Analysis of concentrations of PCDD/PCDF in ambient air have also been undertaken in a number of localised areas around particular sources of pollution. The results of an air and deposition monitoring programme of the area surrounding the Coalite works are given in one

of two reports concerned with this area (Environment Agency 1997a). The second report covers the soil survey (Environment Agency 1997b). The Coalite Works is an industrial complex in Bolsover in the north of England. The Works consists of a smokeless fuel plant and a chemical plant producing, amongst other substances, 2,4,6–TCP. The monitoring programme took place in late 1992 to 1993, after the closure of the chemical waste incinerator at the chemical works. The aim of the programme was to assess the environmental concentrations of dioxins in the surrounding environment and to assess whether the measured concentrations could be related to known emissions from the Works. Seven sampling locations were chosen. One was upwind of the works in the nearby village of Scarcliffe, to give an indication of ‘background’ concentrations. The results are shown in Table A 67.

Table A 67: UK - Ambient air and deposition in the Bolsover area

Location	Sampling period	Number of samples	Min	Max	Mean	Median
Ambient air (fg I–TEQ/m³)						
‘Background’ (1 site)	Oct 92-Oct 93	3	225	489	371	399
Industrial (6 sites)	Oct 92-Oct 93	79	33	547	214	192
Deposition (pg I–TEQ/m² day)						
‘Background’ (1 site)	Oct 92-Oct 93	2	15	89	52	-
Industrial (6 sites)	Oct 92-Oct 93	36	2	118	25	18

The study found that concentrations of PCDD/PCDF in the air at the industrial site were generally comparable to concentrations reported in urban areas of the UK. The concentrations were similar to those reported by NETCEN (1998) for Cardiff and Stevenage in 1992/1993. The concentrations for the ‘background’ site were not representative of a rural location, and were actually, on average, higher than the industrial sites. This may be a result of unusual wind conditions during the time of sampling, and possible further contamination by local domestic coal burning (Environment Agency 1997a). The Environment Agency reports some evidence of an intermittent source of 2,3,7,8-Cl₄DD and 2,3,7,8-Cl₄DF and clear evidence of a continuous source of other Cl₄DD, Cl₅DD, and Cl₄DF, which were consistent with the source ‘fingerprint’ for the works. The survey showed a broad spatial distribution of elevated concentrations across the area.

The deposition data showed wide variations in deposition rates. This may in part have been a reflection of the uncertainty in the sampling method used. Deposition rates ranged from 2-118 pg I-TEQ/m² day, with PCDF making the greater contribution to the I–TEQ. Again, the ‘background’ site did not show lower concentrations than the other sites. Vegetation samples were also analysed to further consider deposition of PCDD/PCDF (see below).

The conclusion of the study states that, in all media (air, deposition and vegetation), the sampling site closest downwind of the Works (site D) showed consistently higher concentrations of PCDD/PCDF than at other sites. The congener pattern at this site was constant, whereas the pattern at the other sites varied independently. This indicates a common source of contamination at site D. It was hypothesised that the 2,4,6–TCP production was responsible for the generation of the classic fingerprint pattern found in these samples, with high Cl₄DD, Cl₅DD, and Cl₄DF. This could result from one or more of three possible sources: emissions during the production of 2,4,6–TCP; fugitive emissions from the process or emissions from the wastes generated in the process.

Ball *et al.* (1993) analysed the area surrounding the Rechem hazardous waste incineration plant in the Panteg area of Pontypool in South Wales. The concentrations of PCDD/PCDF in ambient air and deposition are shown in Table A 68.

Table A 68: UK - Ambient air and deposition data for the Panteg Region of South Wales

Location	Type of location	Sampling period	n	Min	Max	Mean	Median
Ambient air (fg I-TEQ/m³)							
Llandegfedd Reservoir	semi-background	31/3/92 - 5/5/92	6	20	680	200	100
Pontyfelin House	industrial / urban	31/3/92 - 5/5/93	5	1,600	14,800	4,600	2,100
Deposition (pg I-TEQ/m²·d)							
Llandegfedd Reservoir	semi-background	24/3/92 - 20/5/92	3	32	44	36	32
Lucas - Girling	industrial / urban	24/3/92 - 20/5/92	3	41	44	42	41
Pontyfelin House	industrial / urban	24/3/92 - 20/5/92	3	21	141	76	65
Ty-coch Farm	Industrial / urban	24/3/92 - 20/5/92	3	33	172	106	113

The data shows that the Pontyfelin House site, which is very close (~150 m) to the Rechem plant, had concentrations of PCDD/PCDF in the air an order of magnitude higher than the semi-background site at Llandegfedd Reservoir, with ranges of 1,600-14,800 and 20-680 fg I-TEQ/m³, respectively. The deposition figures showed less dramatic differences, but the mean concentrations in the industrial sites (42, 76 and 106 pg I-TEQ/m²·d) were all higher than the semi-background site (36 pg I-TEQ/m²·d). The high air concentrations measured in this region were, therefore, higher than those concentrations recorded in other urban areas in the UK, but the deposition rates were broadly consistent with those reported by NETCEN (1998).

A14.4 VEGETATION

There has been a national survey of concentrations of PCDD/PCDF in vegetation in the UK, but this was carried out before the use of toxic equivalent factors, and therefore there are few comparable data available. However, there has recently been a study of time trends in concentrations in vegetation dating back to the 1840s and further studies have been carried out in areas of suspected contamination, notably Bolsover in Derbyshire and Panteg in South Wales.

The UK wide vegetation survey involved 67 vegetation samples collected from background locations throughout the UK (Startin *et al.* 1989). These produced a median concentration of total PCDD/PCDF of 45 ng/kg d.m. with a range of 9.6-455 ng/kg d.m. However, there has not been a more recent national survey for comparison of I-TEQ values.

A study of the environment surrounding the Bolsover Coalite Works (Environment Agency 1997a) included analysis of PCDD/PCDF concentrations in herbage. Twenty four samples of grass were collected from six sites surrounding the Works. A summary of the results is shown in Table A 69. Concentrations in the vegetation were generally <5 ng I-TEQ/kg d.m., but two samples from site D (closest down-wind site to the Works) contained 14.6 ng I-TEQ/kg. The pattern of congeners was similar to that found in the air and deposition samples, and was linked to 2,4,6-TCP production, with high Cl₄DD, Cl₄DF, and Cl₅DF.

Table A 69: UK -Vegetation at Bolsover (ng I–TEQ/kg d.m.)

Location	Sampling period	n	Min	Max	Mean	Median
Bolsover Coalite Works	March 93-Dec 93	24	0.3	14.6	3.9	3.7

The Panteg monitoring project involved the collection of 11 grass samples from sites surrounding the Rechem hazardous waste incinerator in Pontypool, South Wales (Ball et al, 1995). The sampling locations were divided into three regions, with increasing distance from the Rechem plant. The results of the analysis are given in Table A 70.

Table A 70: UK - Grass samples in the Panteg monitoring project (ng I–TEQ/kg d.m.)

Region	n	Min	Max	Median
Pontyfelin House; <150m	4	5.5	24	12
Tycoch Farm and Pontyrhossa; 150-400m	5	3.1	5.3	3.6
Race Farm, Ysguborneydd Farm, Maes-mawr Farm; >2800m	3	3.1	4.2	4.0

The highest values were recorded in the region closest to the Rechem works, and they were significantly higher than the concentrations found in the other regions. The concentrations found in the two outer regions were comparable with those in the Bolsover area, and roughly 6-7 times higher than those recorded in the recent background samples from Rothamsted. Vegetation samples from Rothamsted are discussed in Task 6 – Time Trends.

A14.5 SEWAGE SLUDGE

In the UK, 1.1 million tonnes (dry weight) of sewage sludge is generated each year, of which about 50% is applied to agricultural land and 8% is sent to landfill (Duarte-Davidson *et al.* 1997). The typical PCDD/PCDF concentration in sewage sludge is 61 ng I–TEQ/kg d.m. (Sewart *et al.*, 1995; DoE 1993). Inputs of PCDD/Fs from sewage sludge applied to agricultural land in the UK have been estimated to be currently about 25 g I–TEQ/year (Jones and Sewart 1995). The I–TEQ input is only about 1.8% of the estimated input derived from atmospheric deposition.

The UK Department of the Environment undertook a survey of sludges from 16 sewage treatment works, situated in a variety of catchment types (DoE, 1993). The results of this survey are summarised in Table A 71. The results show that the amounts of PCDD/PCDF in sewage sludges increase with increasing urbanisation and industrialisation of catchment types.

Table A 71: UK – Sewage sludges (from Jackson and Eduljee, 1994) Concentrations in ng I-TEQ/kg d.m.

	Mean	Range
Rural	23.3	9-73
Mixed industrial / rural	42.5	29-67
Light industrial / domestic	42.3	21-105
Industrial / domestic	52.8	8-192

In a separate survey, digested sewage sludges from urban and industrial waste water treatment plants were sampled in 1992 and analysed for PCDD/PCDF (Sewart *et al.* 1995). Concentrations given as I-TEQ values are available for eight of these samples. The data are provided in Table A 72.

Table A 72: UK - Sewage sludge from various locations in the north west of England

Source of sludge and type of treatment	Concentration (ng I-TEQ /kg)
Industrial / urban waste water treatment	
Activated sludge treatment and filter units	30
Primary sedimentation and activated sludge treatment	90
Activated sludge treatment	85
Activated sludge treatment	206
Average:	82
Urban waste water treatment	
Filter unitse	51
Activated sludge treatment	47
Filter units	46
Primary sedimentation only	19
Average:	33

These results are within a similar range to those in the DoE survey. The congener profiles showed that there was a general increase in concentration with increasing congener chlorination. The results in the table above show that, on average, the sludge from treatment works with industrial effluent as well as urban had higher concentrations of PCDD/PCDF. The pattern of contamination is similar to that of PCP, and this is suggested as the source of the higher contamination.

The results of archived sewage sludge samples are discussed in Task 6.

A14.6 WILDLIFE

Limited data on PCDD/PCDF concentrations in fish are available, but no other data on wildlife in the UK have been found. Rose and McKay (1996) describe a study to investigate the fate of airborne PCDD/PCDF deposited onto freshwater systems and to identify if there is a link between concentrations of dioxins in sediment and concentrations in fish. Single

samples were taken of pike and perch from Eleven Acre Lake, roach from Marsworth Reservoir, roach and eels from Lough Neagh and brown trout from Dry Loch. Samples of sediments were also taken from the four lakes above. Homogenised samples of the whole fish were analysed in each case, rather than just the fillet. The concentrations found were in the range 16-400 ng I-TEQ/kg fresh weight. The data are shown in Table A 73.

The wide ranges in concentrations were partly explained by the different behaviours of the fish, being at different concentrations in the food chain. Roach is a benthic feeder, whereas pike feeds solely on other fish. The eel is a fatty fish and spends the winter in sediments at the bottom of the lake. This may explain the relatively high fresh weight concentration. There were also variations in concentrations within the same species *i.e.* the roach, explained by differences in size and age. The study concluded that there was some correlation between sediment and fish concentrations but was not significant.

Table A 73: UK - Fish from four lakes

Type of fish	Location	Date	Concentration (ng I-TEQ/kg fw)	Concentration (ng I-TEQ/kg fat)
Pike	Eleven Acre Lake	1993	1.4	340
Perch	Eleven Acre Lake	1993	8.2	400
Roach	Marsworth Reservoir	1993	0.9	16
Roach	Lough Neagh, Northern Ireland	1993	22	700
Eel	Lough Neagh, Northern Ireland	1993	14	140
Brown trout	Dry Loch, Scotland	1993	4.5	44

A14.7 WASTE

An analysis of domestic waste has been undertaken in the UK, to assess the concentration of contamination by PCDD/PCDF, among other organic pollutants (Collings and Van Santen, 1996). Three waste samples were analysed from different locations, representing different regional and socio-economic characteristics. In each case approximately 5 tonnes of dustbin waste were collected in 1994, and these were sorted into categories of waste to produce composite samples by type of waste, such as putrescible, plastic, paper and fines. The PCDD/PCDF concentrations of each of these samples were then analysed. The overall concentration in the waste was found to range from 3.1-13 ng I-TEQ/kg dry matter, which is lower than other data quoted in the literature, which indicated a range of 10-250 ng I-TEQ/kg d.m. The highest concentrations found by Collings and Van Santen (1996) were in the miscellaneous combustible category (8.8-11 ng I-TEQ/kg d.m.) and in the fines (5.6-70 ng I-TEQ/kg d.m.), which consisted partly of coal ash.

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A16 Glossary

AMAP	Arctic Monitoring and Assessment Programme
BGA	Bundesgesundheitsamt (=former German Health Agency)
DG	Direction Générale
d.m.	Dry matter
EPA	Environmental Protection Agency (various countries)
EU	European Union
MSWI	Municipal solid waste incinerator
MTL	Maximum Tolerable Limit
PA	Polyacryl
PAN	Polyacryl nitrile
PE	Polyethylene
PCDD	Polychlorinated dibenzo- <i>p</i> -dioxins
PCDF	Polychlorinated dibenzofurans
PCP	Pentachlorophenol
POPs	Persistent Organic Pollutants
PUF	Polyurethane foam
SE	Southeast
SW	Southwest
UNEP	United Nations Environment Programme
TCP	Trichlorophenol
TeCP	Tetrachlorophenol
TEQ	Toxicity equivalent (I = International; N= Nordic)
W	West

Country Codes

A	Austria	I	Italy
B	Belgium	IRE	Ireland
DK	Denmark	L	Luxembourg
D	Germany	NL	The Netherlands
E	Spain	P	Portugal
F	France	S	Sweden
FIN	Finland	UK	The United Kingdom
GR	Greece		

Units

mg	milligram	10^{-3} g
µg	microgram	10^{-6} g
ng	nanogram	10^{-9} g
pg	picogram	10^{-12} g
fg	femtogram	10^{-15} g