Compilation of EU Dioxin Exposure and Health Data

Task 6 – Trends

Report produced for

European Commission DG Environment

UK Department of the Environment, Transport and the Regions (DETR)

October 1999

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Executive Summary

Many analyses have been carried out in European Union Member States to determine the concentrations of dioxin in the various environmental media and other matrices. This report draws on information collected in the course of a number of other Tasks within the project (Environmental Levels, Human Exposure, Human Tissue and Milk Levels) and presents an overview of the trends in dioxin concentrations observed in the environment, wildlife, foodstuff, human milk and blood. This report focuses on a number of individual studies which have specifically investigated trends in dioxin concentrations, while the more detailed Tasks consider the broader picture of dioxin concentrations and variations within Member States. Most trend data available within the EU relate to temporal variations, rather than spatial variations or congener patterns.

Although the information is sometimes somewhat contradictory, it can be concluded that the anthropogenic input of dioxins into the environment started around 1940. Earlier samples only contained very low concentrations of dioxins, which might originate from minor sources such as forest fires, domestic heating and smaller industrial activities. Since 1940 marked increases in concentration have been observed, for example in sediment cores from Arctic Finnish lakes, where the dioxin concentrations were very low, as well as in samples from more industrialised areas in Germany with higher concentrations. Concentrations generally peaked between the late 1950s to the 1970s and started to decline in more recent years, as a result of measures taken to reduce dioxin emissions. It was also found that the profiles changed: whereas the older congener profile is indicative of the production and use of chlorinated phenols, the more recent profile is indicative of combustion sources.

The overall finding from this Task is that trend analysis is a helpful tool to investigate the input and occurrence of dioxins in the environment and human food-chain. It helps to determine the effectiveness of measures taken by governments and agencies to reduce the release of dioxins into the environment. The data have shown clearly that there is a need for long-term follow-up of such data gathering, as between year variation can be significant and, thus, long time periods are required in order to establish trends. As there is still dynamic in many matrices and locations, it can be assumed that the dioxin concentrations in the Member States of the EU have not yet levelled of and, thus, there is a need to continue the analyses that have established the present trends.

As this report draws on information collected as part of other Tasks comprising this study, many of the key recommendations have been made in the relevant sections of those Task reports. However, a number of more general observations and recommendations are made here.

- As congener-specific dioxin analysis (very often using high resolution mass spectrometers) has only been used routinely for around 10 years, long time series of data are, clearly, not available. Thus, a number of current monitoring and research programmes should be extended for at least a decade, in order to establish adequate series of data to demonstrate trends in dioxin concentrations in the environment. A database of

trends in dioxin concentrations in European Union Member States would greatly assist the implementation of a number of important international agreements.

- In general, governmental agencies, research institutions and private laboratories have generated data on dioxin concentrations for specific locations or matrices. Assuming that these samples have been analysed using methods which are comparable with the high standard in dioxin analysis available today, these institutions should be encouraged to continue their programmes on a similar basis.

- A number of Tasks within this study have highlighted the fact that there remain considerable data gaps for a number of countries and, in particular, the Southern EU Member States. New monitoring and research programmes should be set up in these countries in such a way that the procedures are consistent, and the data comparable, with existing programmes in other Member States. Such information would help to establish whether geographical patterns of dioxin concentration exist in the various regions of the EU.

- New Member States of the European Union should be encouraged and assisted in establishing monitoring and research programmes to generate data which is consistent and comparable with that from the existing Member States. If necessary, this should involve support in achieving the highest standards of dioxin analysis.

Contents

1 Introduction

Worldwide many countries have performed a multitude of analyses in order to determine the concentrations of polychlorinated dibenzo-*p*-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF), often collectively termed 'dioxins', in environmental media and other matrices. Data provided by individual EU Member States and found in the published literature are summarised in Task 2-Environmental Levels, Task 4-Human Exposure and Task 5-Human Tissue and Milk Levels.

This report provides an overview of trends in dioxin concentrations, based on the information received during the course of this study. It focuses upon a number of individual studies which have specifically investigated trends in dioxin concentrations, while the more detailed Tasks, listed above, consider the broader picture of dioxin concentrations and variations within EU Member States. In most cases, the information compiled here was drawn from the published literature and personal communication with individuals actively involved in the relevant research.

Trends can be established if a series of data is available. A prerequisite for any trend analysis is that the samples are comparable. This means that the same analytes were quantified. The samples must have been collected using the same method and the analysis have been the same for all samples. As an example, very often dioxin results obtained with high resolution mass spectrometry (HRMS) in recent years cannot be compared with data from the late seventies or early eighties, when the analysis was less advanced.

Evaluation of these data can be temporal, with a fixed location and a variation in time. The time-line can be along several years or just a few days or weeks. Spatial variation is determined if at the same time, samples are taken from different locations.

The information in this report is grouped according to the environmental matrices considered. In most cases, the trend evaluation relates to changes over time. Spatial distribution could be given only in exceptional cases.

1.1 TIME TRENDS

Time trends can be established by analysing existing data, which have been generated in a laboratory or a country over several years. In a few cases, trends can be established using existing samples kept in storage until analysis can be carried out on all samples at the same time. The latter has the advantage that the conditions in the laboratory are the same for the whole sample set and, thus, the results are more suitable for comparison. One example of this is the samples stored at Rothamsted Experimental Station in the United Kingdom, where soil and herbal samples are available dating back to the middle of the nineteenth century.

Sometimes, the sample matrix itself allows for a trend analysis: this is the case when soil or sediment core samples are taken. Core sampling is appropriate only if there are undisturbed

matrices. Upon optical inspection and subsequent analysis the different layers in a core can be dated and the measured concentrations can be attributed to certain years.

Spruce needles also allow for a 3-4 years retrospective analysis. The different colours of the spruce needles on the twig allow analysis of 1, 2, 3 and even 4 years old needles.

Examples for temporal trends are given below for soil, sediments, air, sewage sludge, biomonitors, foodstuffs, human milk and blood.

1.2 SPATIAL TRENDS

Spatial trends can provide important information on the contamination profile. Successful examples are the application of trajectories through countries. It is possible to identify sources of contamination when correlating, for example, air concentrations with major wind directions.

Other spatial variation is obtained when measuring, for example, soil concentration at different distances and directions from a known point source.

Examples for spatial trends are given below for soils.

1.3 CHANGING PATTERNS

With distance or time, the relative composition of dioxin congeners may change. The first effect would be attributable to a chromatographic effect, which means that the lower chlorinated congeners are more volatile than the higher chlorinated, and the second effect would be attributable to transformation processes, with photochemical degradation or metabolism as two examples.

For PCDD and PCDF, there are no conclusive data which show significant changes in congener patterns. So far, most researchers have concluded that there is hardly any loss of dioxins from, for example, soil samples over several years. In addition, due to the high value of the organic carbon/water partition coefficient (K_{OC}) for PCDD and PCDF (see Task 3 – Environmental Fate and Transport), there is hardly any transport in soils with depth and thus, a chromatographic effect in soils has not been established.

A chromatographic effect was established for the composition of dioxin congeners in Arctic animals, which showed that, for example, 2,3,7,8-TCDF is found at higher concentrations in Arctic Seals than in animals from more southern latitudes.

2 Soil

Kjeller *et al.* (1991) reported the analysis of archived soil samples dating back to 1846, which were stored at the Rothamsted Experimental Station in southern England. These soils came from a control plot, which had never received applications of fertiliser, soil amendments or pesticides. All the samples were taken from the 0-23 cm layer, except the first sample in 1846, which was taken at a time when the plough layer was shallower, at 0-12 cm. The results of the dioxin analysis are shown in Table 1. The data show that dioxins were detected in all samples and there has been a progressive increase in concentrations since the beginning of this century. The average year-on-year increase from 1893 to 1986 was around 1.2%.

It can also be concluded from these data that, once a contamination level is reached, dioxins are not lost from the soil. Consequently, photodegradation does not play a role in decreasing dioxin contamination in soils.

Table 1: UK - Temporal trends in soil concentrations

Year	Σ PCDD/PCDF (ng/kg)
1846	61
1846	54
1856	31
1893	31
1914	42
1944	62
1944	57
1956	74
1966	89
1980	81
1986	95
1986	88
1986	92

Data from various studies carried out in Germany on the concentration of dioxins in soil allow an evaluation of concentrations according to horizons or sampling depth. The individual data from the various studies are given in the Annex to this report and more detail, together with the appropriate references, can be found in the Technical Annex to Task 2 – Environmental Levels. The summary data for 442 samples from Bavaria are given in Table 2, below, and show that, in the uncontaminated rural locations, the mean and maximum concentrations decreased with increasing depth into the soil.

Table 2: Germany – Trends in soil concentrations in Bavaria according to the horizons analysed (n=442) ng I-TEQ/kg d.m.

Horizon	Data	Type of Project	
		Potentially contaminated	Rural - Routine
Of	Minimum	3.72	ND
	Maximum	139	50
Ah	Minimum	ND	ND
	Maximum	42	18
Ah/Ap	Minimum	0.22	0.1
	Maximum	230	17
Ap	Minimum	ND	ND
	Maximum	25	5
Total Minimum		ND	ND
Total Maximum		230	50

ND = Not detected

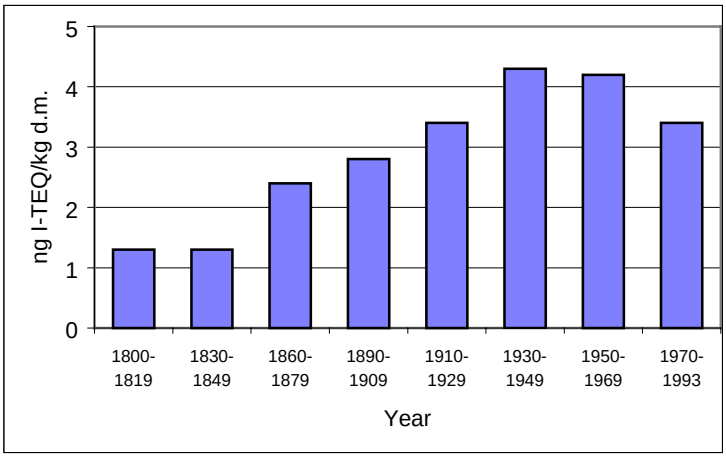
She and Hagenmaier (1996) analysed dioxins in soil around the Rastatt metal reclamation plant in Germany. It was supposed that the dioxins in the soil originated in the stack emissions from the plant (shut-down in 1986). A total of 154 soil samples were taken within 500 m of the source, at 0-30 cm depth. The choice of sampling sites was based on distance to the stack and direction of the prevailing winds. Of the 154 samples, there were 77 samples with one or more 2,3,7,8-substituted congeners below the limit of quantification; most of these were far from the source. For the overall evaluation, these samples were excluded. The remaining 77 samples gave the following findings:

- the concentrations ranged from 290 to 208,000 ng PCDD/kg d.m. and 400 to 447,000 ng PCDF/kg d.m. The minimum concentration expressed as I-TEQ was 12 ng I-TEQ/kg d.m. and the maximum was 14,500 ng I-TEQ/kg d.m;
- the geographic distribution of the dioxin followed an exponential decrease with distance from the source. The decrease for PCDF was faster than for PCDD.

3 Sediment

Sediment cores from Loch Corie nan Arr, a remote lake in Scotland, were analysed in order to determine changes in atmospheric deposition of dioxins through time (Rose, 1996). The most recent sediment sample was dated as 1970-1993, in which the concentration was 3.4 ng I-TEQ/kg d.m. This was the lowest contamination found in contemporary UK lake sediments. The time trend analysis showed that low concentrations of dioxin were present in the lake in the early 1800s, but started to increase towards the end of the 19th century. The peak concentration, when expressed as I-TEQ, was reached in the 1930-40s, whereas the peak was in the 1950s and 1960s when based on total dioxin concentration. Since then, concentrations have decreased to the present day. The temporal trend is shown in Figure 1. The homologue profiles of the sediments have remained broadly constant through time, dominated by the higher chlorinated PCDD. This pattern is consistent with the remote nature of the lake catchment, with all contamination arriving through atmospheric transport and deposition.

Figure 1: Time trends in concentrations in sediments in Loch Corie nan Arr, Scotland

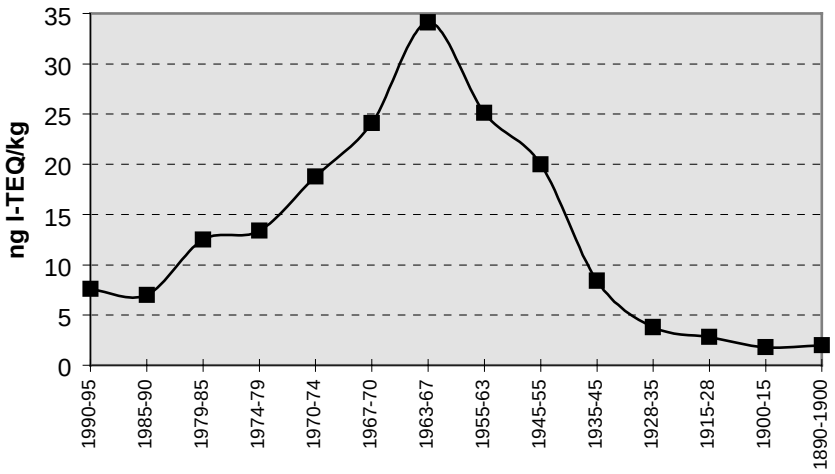


The Arctic Monitoring and Assessment Programme report (AMAP 1998) presents historical profiles of PCDD/PCDF (expressed as totals) and OCDD for three Finnish lakes. The graphical presentation is shown in Figure 2. Two of these cores from Arctic lakes in northern Finland show recent declines in dioxin deposition following a major increase since the 1940s. In the Lake Pahtajärvi, a subsurface maximum for dioxins is discernible in slices dated to the mid 1970s along with an elevated concentration in the surface slice. The historic records for OCDD differ from the results reported by Kjeller and Rappe (1995) for cores in the Baltic Sea, which had highest concentrations in the more recent slices (1970s-1980s).

Figure 2: **Concentration profiles for OCDD and ΣPCDD/PCDF in dated sediment cores from Arctic lakes in Finland and Canada. The median age of the slices is displayed on the y-axis (AMAP 1998)**

Hagenmaier and Walczok (1996) analysed sediment cores from Lake Constance, Germany. They were able to document a time trend for dioxin concentrations for 100 years. A graphical presentation is shown in Figure 3. 2,3,7,8-Substituted congeners could be detected in every layer. Older layers contained more PCDF than PCDD, a result which was also found by Kjeller and Rappe (1995) in their analyses of sediment cores. From approximately 1940, the PCDD concentrations increased faster than the PCDF concentrations and thus, since 1982, there was more PCDD present than PCDF. As can be seen from Figure 3, the concentrations, expressed as I-TEQ/kg d.m. increased after about 1940 and had a maximum in the 1960s. A pattern analysis shows that the more recent samples exhibit the typical combustion pattern whereas the older layers (ca. 1940-1970) reflect the "chlorine" pattern dominated by OCDD. It is interesting to note that I-TEQ concentrations in Loch Corie nan Arr and Lake Constance were similar over the period to around 1935, and could be regarded as 'background' levels, subsequently concentrations in Lake Constance reflect the impact of more localised sources.

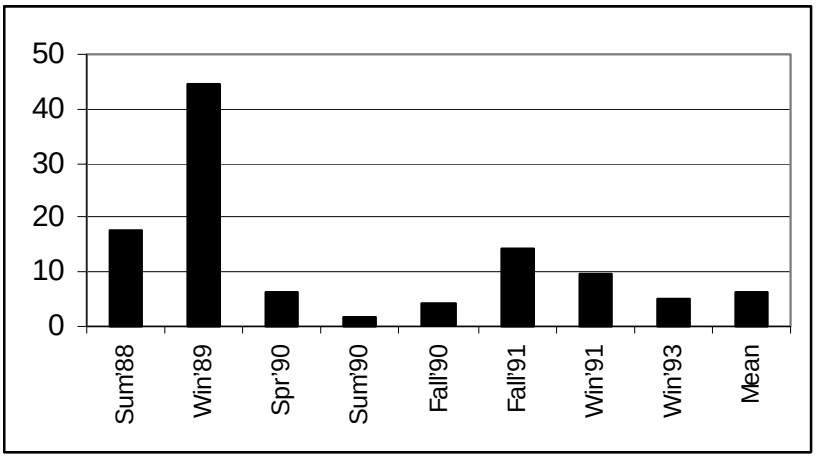
Figure 3: Germany – Sediment cores from Lake Constance (concentrations in ng I-TEQ/kg d.m.) (Hagenmaier and Walzcok 1996)



4 Air

From Sweden, there are some results available for ambient air concentrations starting from summer 1988 and ending in winter 1993. The concentrations are shown in Figure 4. It can be seen that the overall trend is downward.

Figure 4: Sweden – Ambient air concentrations (means) grouped into seasons and years. Concentrations in fg N-TEQ/m³



The same trend in a comparable time period was obtained in Germany in the State of Northrhine Westphalia, where reductions between 46 and 69 % were observed when comparing the concentrations from 1987/88 with 1993/94. The concentrations in Germany, however, were on much higher levels than those in Sweden (see Table A20 in the Technical Annex to Task 2). The higher concentrations in Northrhine Westphalia reflect the much more densely populated and highly industrialised character of this area.

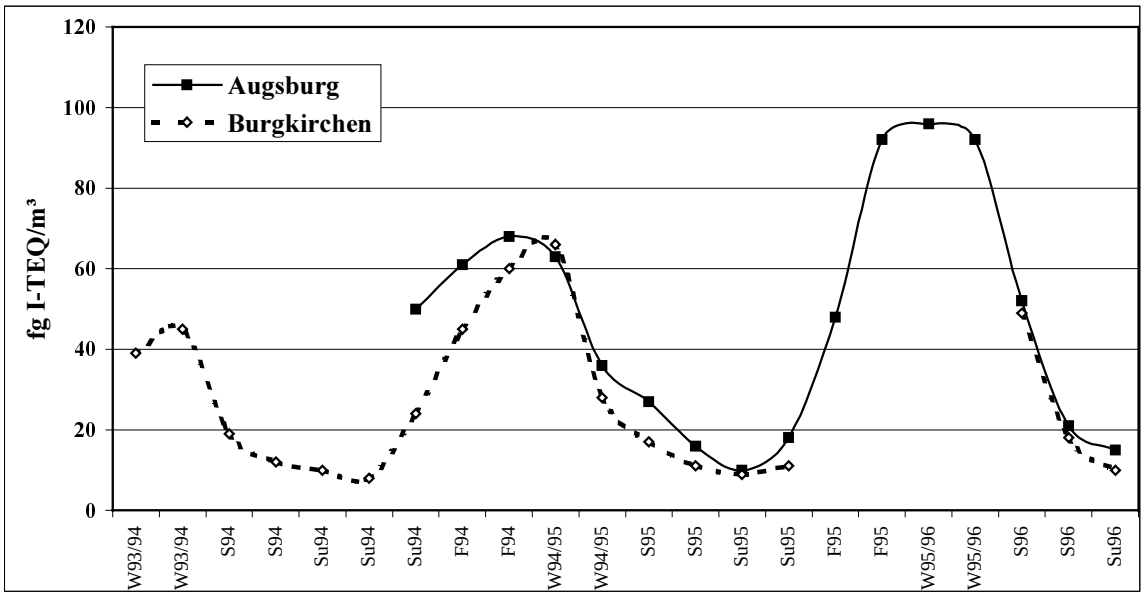
During a 2½ year monitoring programme (winter 1993/94 until summer 1996), 223 ambient air samples were obtained from two sampling areas in Bavaria, Germany. The sampling sites were located in the impact area around two municipal solid waste incinerators in Augsburg and in Burgkirchen. In addition, one remote sampling site was included in the monitoring programme for comparison. As there was no difference found between the concentrations obtained within and outside the impact areas of the two MSWIs, the results from the two remote locations were included in the overall evaluation. The results are summarised in Table 3 and a graphical presentation is shown in Figure 5. As can be seen, a strong seasonal trend for dioxins was found in both locations with higher concentrations during the winter months and up to 10-fold lower concentrations during the summer months.

Table 3: Seasonal trend of ambient air concentrations in Southern Bavaria. The values represent median concentrations from 8 sampling locations in the Augsburg network and five sampling locations in the Burgkirchen network (Augsburg: n=125 and Burgkirchen: n=98) (Fiedler *et al.* 1997)

	Year	Winter		Spring		Summer			Fall	
Augsburg	1993/94							50	61	68
Burgkirchen	1993/94	39	45	19	12	10	8	24	45	60
Augsburg	1994/95	63	36	27	16	10	18		48	92
Burgkirchen	1994/95	66	28	17	11	9	11			
Augsburg	1995/96	96	92	52	21	15				
Burgkirchen	1995/96			49	18	10				

From the data shown in Table 3 and in Figure 5, and the results reported for Bavaria, Germany, in Task 2, the downward trend as shown for ambient air concentrations in Northrhine Westphalia during the last years could not be confirmed in southern Bavaria. Combining the Bavarian data obtained since 1992, it can be concluded that the ambient air concentrations in Bavaria have not changed during the last five years and remained at a low level.

Figure 5: Ambient air concentrations of dioxins in southern Germany. Median concentrations obtained from the networks around the MSWIs at Augsburg and Burgkirchen



5 Sewage Sludge

Archived sewage sludge samples are available from the United Kingdom (Sewart *et al.* 1995). These were collected from the Isleworth sewage treatment works (STW) in West London between 1942 and 1960, and have been kept as part of the long-term agricultural experiments at Rothamsted Experimental Station. The results of the time-trend analysis are presented in Table 4. These archived samples show an increase in dioxin concentrations with time from 1942 to 1956, followed by a steady decrease to 1960. The congener profiles in the archived sampled also showed evidence of contamination by PCP, towards the more recent years

Table 4: Concentrations in sludge from Isleworth STW, West London

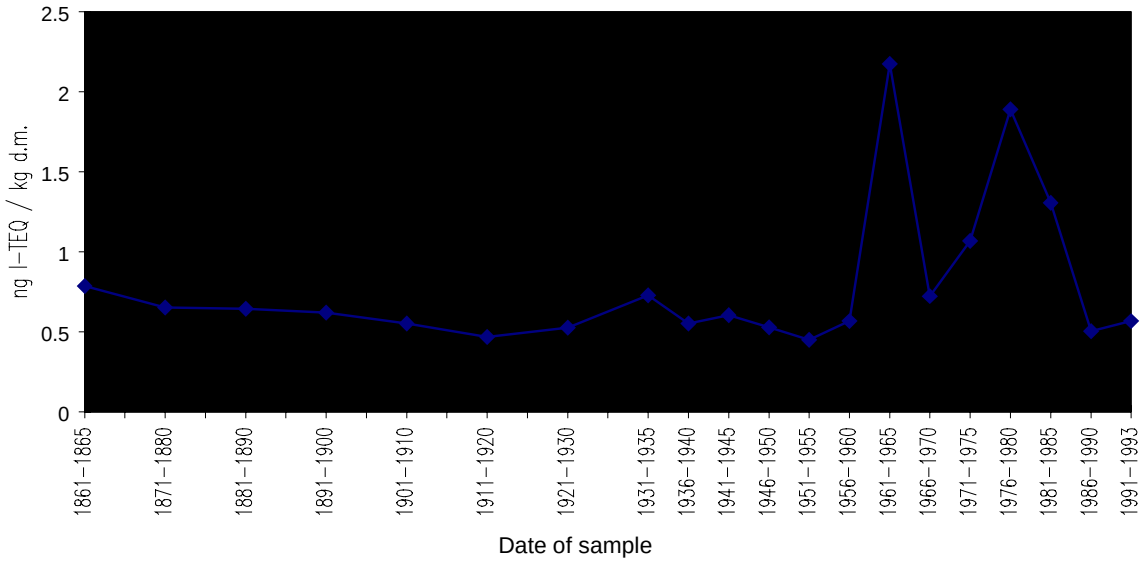
Sample Date	Concentration (ng I-TEQ /kg)
1942	18
1944	36
1949	61
1953	127
1956	402
1958	229
1960	166

6 Biomonitors

6.1 VEGETATION

Kjeller *et al.* (1996) described a study of an archive of bulked grass samples from the Rothamsted Experimental Station in the Southeast of England. The grass came from an undisturbed plot, which had never received applications of fertilisers, soil amendments or pesticides. Twenty composite samples were used, dating from 1861 to 1991, covering varying intervals of 3, 5 and 10 years. The graph in Figure 6 shows that concentrations remained roughly constant between 1861 and 1956, after which there was a steep increase to a peak in 1961-1965, possibly relating to peak usage of chlorinated pesticides. There was a second peak in 1976-1980, possibly linked to an increase in waste incineration. Since then there has been a four-fold decline in I–TEQ back to the pre-1960 levels. It is interesting to note that the most toxic congener, 2,3,7,8–TCDD, has not shown a decline and, therefore, the I–TEQ concentrations have not fallen as fast as the total dioxin concentrations. The most recent samples contained 0.57 ng I–TEQ/kg d.m, which is a typical present day concentration for UK vegetation (Startin *et al.* 1989). There is a continuing debate about the source of contamination in pre-1946 samples, which is most likely to be from the combustion of wood, coal and metal smelting, but may also represent post-collection contamination (Kjeller *et al.* 1996).

Figure 6: UK – Dioxin in archived grass samples from Rothamsted

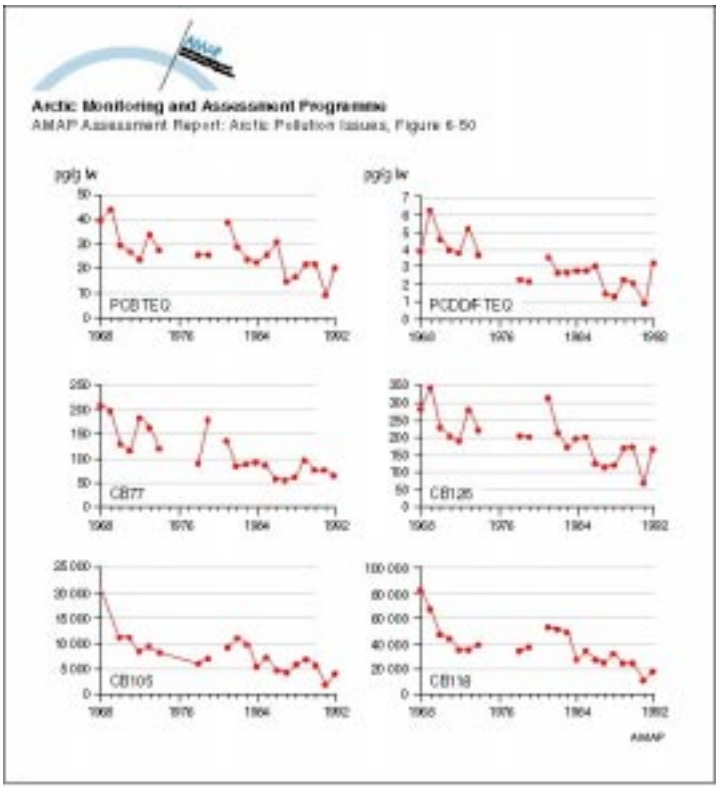


6.2 WILDLIFE

In 1968 a Swedish program was initiated to monitor pollutants in the Baltic environment. Samples were taken annually to evaluate temporal trends of various aquatic species. Target locations were areas with little or no known local discharges. Each annual sample at a site was represented by 10-25 specimens, thus, within year variation could be estimated. The selection of samples maintained consistency in sex, age, size and sampling season.

Lake Storvindeln is a forest lake near the Swedish Alps, an Arctic sampling site. Muscle samples from 20 pike collected in spring were analysed each year since 1968. Figure 7 shows the results for PCB and PCDD/PCDF reported in the AMAP report (AMAP 1998) for these samples. The Figure demonstrates the correlation between the various congeners, but also the between-year variation of concentrations. The overall decline for PCDD/PCDDF (in TEQ) is smaller and more variable than the decline for PCB.

Figure 7: Concentrations of PCDD/PCDF (pg TEQ/g fat) and PCB congeners in Lake Storvindeln, Sweden, pike muscle between 1968 and 1992 (AMAP 1998)

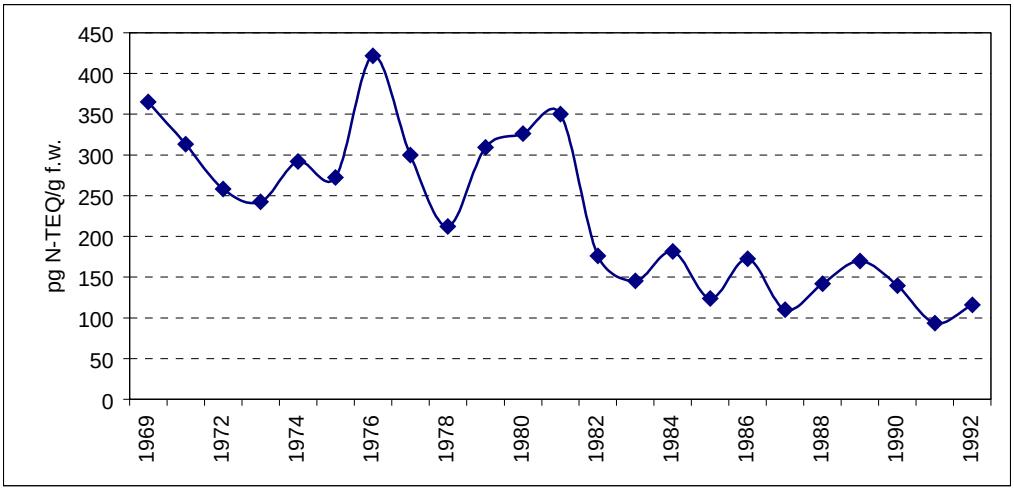


With the Environmental Specimen Bank (ESB) at the Swedish Museum of Natural History, Stockholm, Sweden has a valuable source of information for spatial and trend monitoring of contaminants. Since the 1960s, tissue samples from more than 150,000 living organisms have been collected from different groups of animals, habitats and types of landscape.

Figure 8 shows the time trend of dioxin concentrations in guillemots for the period 1969 to 1992. This shows an overall decline in TEQ concentrations, with short-term peaks around

1975 and 1978-80. The guillemot is a piscivorous bird that breeds in a few locations in the Baltic Sea. The island of Stora Karlsö is the most prominent breeding habitat for the species in the central Baltic Sea, comprising approximately 7,500 breeding pairs, according to the 1985 estimate. Eggs of guillemot have shown to be a very useful matrix for studies of environmental contaminants.

Figure 8: Sweden: Time trend of dioxin contamination in guillemot eggs from Stora Karlsö in the Baltic Proper



7 Foodstuffs

A total of 222 butter samples were analysed for dioxin in Baden-Württemberg, Germany. The sampling periods were from 1993 through 1996. As can be seen from Table 5, the mean concentrations show a decreasing trend. None of the samples exceeded the guideline concentration of 3 pg I-TEQ/g fat, and none of the samples had to be removed from sale (exceeding the limit concentration of 5 pg I-TQ/g fat) (Malisch 1999).

Table 5: Germany – Butter samples (pg I-TEQ/g fat)

Year	n	Mean	Minimum	Maximum
1993	27	0.83	0.19	1.52
1994	37	0.68	0.46	1.38
1995	92	0.64	0.27	2.00
1996	66	0.55	0.19	0.87

A summary of the dioxin concentrations detected in 667 cows' milk samples collected in Germany between 1993 and 1998 are shown in Table 6. The concentrations in the samples did not show the same downward trend as the butter samples, although from the same geographic area. From Table 6, it can be seen that, towards the end of 1997, the dioxin concentrations in cows' milk had started to increase. The reason for this was that in Baden-Württemberg the first evidence was detected of the Europe-wide contamination of dairy products, due to high concentrations of dioxins in citrus pellets, which were added to the cattle feed.

Table 6: Germany – Milk samples (pg I-TEQ/g fat)

Year	n	Median	Mean	Min	Max
1993	97	0.63	0.68		
1994	222	0.67	0.79		
1995	104	0.62	0.69		
1996	115	0.59	0.60		
1997	112	0.62	0.71		
01/97-08/97	76	0.59	0.62	0.36	1.02
09/97-12/97	36	0.82	0.89	0.35	1.92
01/98-02/98	27	1.06	1.41	0.46	7.86

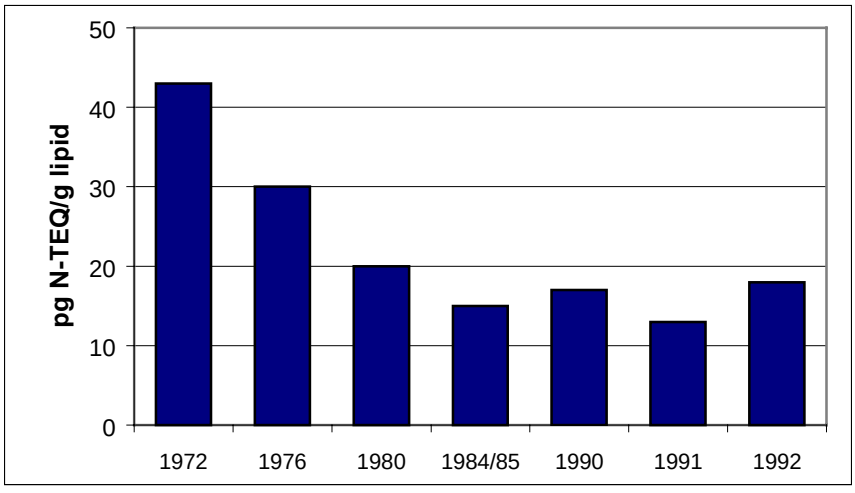
8 Humans

8.1 BREAST MILK

A number of EU Member States have contributed to the WHO-coordinated assessment of dioxin concentrations in breast milk (Austria, Belgium, Denmark, Finland, Germany, the Netherlands, Spain, Sweden and the United Kingdom), the first round of which was conducted during the period 1986-1988 and the second during 1992-1993. For both rounds, analyses were performed on pooled milk samples, composed of varying numbers of individual samples from at least 10 nursing mothers. Donors had to be nursing their first child (primaparae) and breastfeeding only one infant. At least two different groups from each country were included in the studies, for example, expected high exposure and low exposure groups, and samples were collected from exactly the same locations for each round of the study. These assessments have proved to be the main source of comparable data for the majority of Member States.

Over the five-year period from 1988 to 1993 the average dioxin concentration in breast milk in European Member States decreased by around 35% (8.3% per year), with a slightly higher decrease in rural areas and slightly lower in industrial areas. A continuous series of measurements made in the urban area of Stockholm, Sweden, have shown a steady decrease in average dioxin concentrations of around 65% over the period 1972 to 1984/85 (8.4% per year), followed by only slight fluctuations to 1992 (Lundén and Norén, 1998).

Figure 9: Sweden: Dioxin in breast milk (Norén and Meironyté, 1998):



8.2 BLOOD

Päpke *et al.* (1992) examined the whole blood of workers engaged in potentially dioxin-contaminated areas due to their working environment. The cohorts included the following working environments:

- Production of trichlorophenol (TCP)
- Production of pentachlorophenol (PCP)
- Metal reclamation
- Production of herbicides.

A total of 166 individuals were examined and the results compared with 102 concentrations considered as German background. The analyses were performed in the year 1988-1992. Here the results are presented on a TEQ basis. However, analysis of the patterns revealed that the composition of the 2,3,7,8-substituted congeners in the blood exhibited some typical behaviour and reflected the impact from the working environment. A few examples:

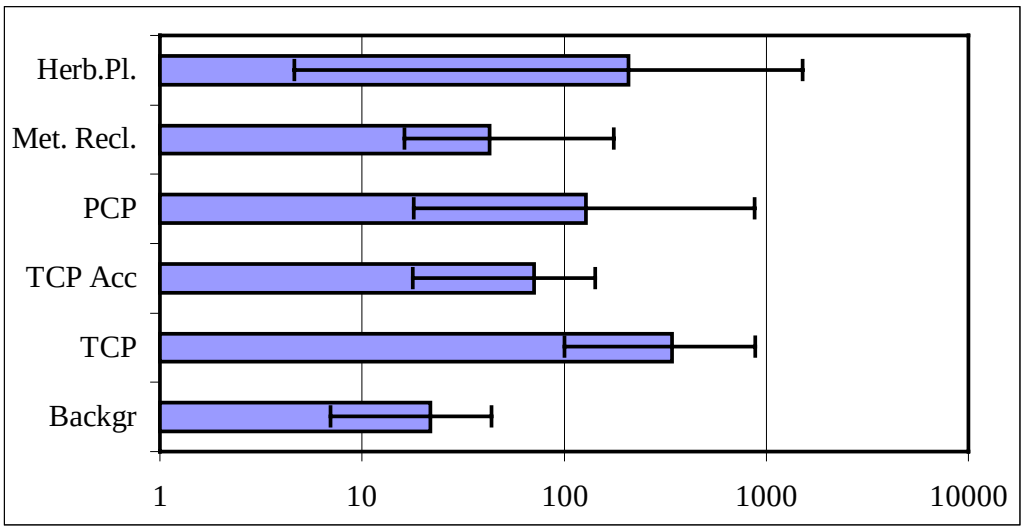
- for the trichlorophenol workers, only 2,3,7,8-Cl_4DD was elevated in comparison to the normal background pattern. Also, in the blood of workers exposed to elevated concentrations of trichlorophenol due to an accident 336 years earlier, concentrations of this congener were still above normal;
- the pentachlorophenol workers had elevated levels of almost all 2,3,7,8-substituted congeners but especially of Cl_7DD and Cl_8DD (up to 300,00 pg/g fat);
- the workers at the metal reclamation plant showed higher concentrations for the Cl_5DF, Cl_6DF and Cl_7DF, typical for their working environment.

Table 7 shows elevated dioxin concentrations in the blood of many of the individuals (on a TEQ-basis). Although low concentrations have also been found, the median concentrations were several times higher than the for the background population. The cohort of workers at the metal reclamation plant only showed a shift in the congener composition (pattern) but, in general, did not show higher concentrations from the work-place when evaluated using the median concentration. The mean, minimum and maximum concentrations are shown graphically in Figure 10.

Table 7: Germany – Blood concentrations as a function of the working place (pg BGA-TEQ/g fat)

	n	Minimum	Maximum	Median	Mean
Background Germany	102	7	43.9	20.8	21.7
Trichlorophenol	12	101	885	239	342
TCP Accident	17	17.9	142	91.7	71
Pentachlorophenol	20	18	877	56.1	128
Metal Reclamation	32	16.2	175	29.7	42.8
Herbicide Plant	85	4.6	1508	102	208

Figure 10: **Blood concentrations from humans in different working environments, means and minimum and maximum concentrations, respectively (pg BGA-TEQ/g fat) (Päpke *et al.* 1992)**



9 Conclusions

This Task draws upon a particular aspect of the information and data collected within other Tasks comprising this study: namely Environmental Levels, Human Exposure, Human Tissue and Milk Levels. Consideration has been given to the extent to which trends in dioxin concentrations have been observed and measured in the various environmental media. Most trend data available from European Union Member States relate to temporal variations, rather than spatial variations or congener patterns.

Although the information is sometimes somewhat contradictory it can be concluded that the anthropogenic input of dioxins into the environment started around 1940. Earlier samples only contained very low concentrations of dioxins, which might originate from minor sources such as forest fires, domestic heating and smaller industrial activities. Since 1940 marked increases in concentration have been observed, for example in sediment cores from Arctic Finnish lakes, where the dioxin concentrations were very low, as well as in samples from more industrialised areas in Germany with higher concentrations. Concentrations generally peaked between the late 1950s and 1970s and started to decline in more recent years, as a result of measures taken to reduce dioxin emissions.

In ambient air seasonal variations have been observed, with higher concentrations in winter and lower concentrations in summer. Variations in air concentrations can be strongly influenced by local sources. Depending upon when emission reduction measures were introduced, ongoing downward trends have been observed (*e.g.* Northrhine Westphalia in Germany) or the concentrations remained at a relatively low level, reflecting the baseline concentration due to inputs from long-range transport (*e.g.* Bavaria in Germany).

Shorter time series are available for foodstuffs and concentrations in humans. These cover periods of approximately ten years; and the biotic matrices , particularly, show strong declines in contamination.

The overall finding from this Task is that trend analysis is a helpful tool to investigate the input and occurrence of dioxins in the environment and human food-chain. It helps to determine the effectiveness of measures taken by governments and agencies to reduce the release of dioxins into the environment. The data have shown clearly that there is a need for long-term follow-up of such data gathering, as between year variation can be significant and, thus, long time periods will be required in order to establish trends. As there is still dynamic in many matrices and locations, it can be assumed that the dioxin concentrations in the Member States of the EU have not yet levelled of and, thus, there is a need to continue the analyses that have established the present trends.

10 Recommendations

As this report draws on information collected as part of other Tasks comprising this study, many of the key recommendations have been made in the relevant sections of those Task reports. However, a number of more general observations and recommendations are made below.

- As congener-specific dioxin analysis (very often using high resolution mass spectrometers) has only been used routinely for around 10 years, long time series of data are, clearly, not available. Thus, a number of current monitoring and research programmes should be extended for at least a decade, in order to establish adequate series of data to demonstrate trends in dioxin concentrations in the environment. This would support the 5th Action Plan of the EU, which states that dioxin emissions should be reduced by 90% when comparing the emissions in 1985 with emissions in the year 2005. Other international agreements, such as the UN-ECE Long-Range Transport and Assessment Programme, also set target dates for the minimisation of dioxin emissions. Similarly, the future POPs Convention will aim to reduce dioxins in the environment. A database of trends in dioxin concentrations in European Union Member States would greatly assist the implementation of these international agreements.

- In general, governmental agencies, research institutions and private laboratories have generated data on dioxin concentrations for specific locations or matrices. Assuming that these samples have been analysed using methods which are comparable with the high standard in dioxin analysis available today, these institutions should be encouraged to continue their programmes on a similar basis.

- A number of Tasks within this study have highlighted the fact that there remain considerable data gaps for a number of countries and, in particular, the Southern EU Member States. New monitoring and research programmes should be set up in these countries in such a way that the procedures are consistent, and the data comparable, with existing programmes in other Member States. Such information would help to establish whether geographical patterns of dioxin concentration exist in the various regions of the EU.

- New Member States of the European Union should be encouraged and assisted in establishing monitoring and research programmes to generate data which is consistent and comparable with that from the existing Member States. If necessary, this should involve support in achieving the highest standards of dioxin analysis.

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Annex

Table A 1: Germany – Soil concentrations according to horizons (ng I-TEQ/kg d.m.)

Type	Horizon	Date	n	Min	Max	Mean	Median
Bavaria							
Rural, routine	Ap	1989/90	27	ND	3.7	0.41	0.12
Rural, routine	Ah	1989/90	46	ND	5.6	0.46	0.21
Rural, routine	Of	1989/90	20	ND	38	11.9	8.8
Rural, routine	Ah	1989/90	15	0.04	3.9	1.01	0.59
Rural, routine	Ah	1989/90	2	0.97	3.1		
Rural, routine	Ap	1989/90	41	ND	5	0.7	0.24
Rural, routine	Ah	1989/90	27	ND	18	3.9	0.6
Rural, routine	Of	1989/90	32	ND	50	14.9	10
Rural, routine	Ah	1989/90	30	ND	17	2.6	0.77
Rural, routine	Ah	1989/90	30	ND	13	1.8	0.95
Rural, routine	Ah/Ap	1989/90	4	0.1	17	4.8	1.02
Potentially contaminated	Ap	1989/90	41	ND	25	2.69	0.2
Potentially contaminated	Ah	1989/90	38	ND	24	3.73	1.05
Potentially contaminated	Of	1989/90	7	21	139	51.3	37
Potentially contaminated	Ah	1989/90	6	0.05	29	5.9	0.8
Potentially contaminated	Ah	1989/90	18	ND	42	5.38	0.9
Potentially contaminated	Ah/Ap	1989/90	15	0.7	230	21.26	3.8
Potentially contaminated	Ap	1989/90	11	0.2	7.35	1.26	0.68
Potentially contaminated	Ah	1989/90	12	0.2	2.3	0.78	0.41
Potentially contaminated	Of	1989/90	2	3.72	3.87		
Potentially contaminated	Ah	1989/90	3	0.69	1.44		
Potentially contaminated	Ah	1989/90	7	0.47	1.63	0.94	0.83
Potentially contaminated	Ah/Ap	1989/90	8	0.22	2.45	1.6	1.72
Brandenburg							
Rural, env. Surveillance	2*Ah	Oct.92	6	0.002	0.17	0.05	0.03
Hamburg							
Conurbation, env. Surveillance	Ap	Nov.92	4	3.57	4.90	4.46	4.69
Saxony							
Background, env. Surveillance	Ap	Mar.95	1	10.2	10.2	10.2	10.2
Rural, env. Surveillance	Ap	Mar.95	1	0.09	0.09	0.09	0.09
Rural, env. Surveillance	Oh	Mar.95	1	2.48	2.48	2.48	2.48
Rural, env. Surveillance	1*H	-	7	0.22	29.5	8.55	7.05

ND Not detected