

Science dossier

# Chloroform in the environment

Occurrence, sources, sinks and effects

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**Summary**

This dossier was prepared at the request of Euro Chlor and compiled from the open, peer reviewed literature. It has permitted the following conclusions (also repeated at the end of the paper):

1. The chloroform flux through the environment is constant at some  $660 \text{ Gg yr}^{-1}$ , released and destroyed. About 90% of emissions are natural in origin: the largest single source appears to be in offshore seawater (contributing  $360 \text{ Gg yr}^{-1}$ ), presumed to be natural, but from an undefined biological process. Littoral and coastal sources from macroalgae are much better defined, but do not appear to contribute much to the total release. Soil processes are the next most important source and the formation of chloroform has been shown to depend on oxidation of humic material in the presence of chloroperoxidase enzymes and chloride ions. The world's soils contribute about  $220 \text{ Gg yr}^{-1}$ . Other natural sources, mainly volcanic and geological, account for less than  $20 \text{ Gg yr}^{-1}$ .
2. The non-natural sources total  $66 \text{ Gg yr}^{-1}$  and are predominantly the result of using strong oxidising agents on organic material in the presence of chloride ions, a direct parallel with the natural processes occurring in soils. The wood pulp and paper industry, together with drinking and other water treatments, account for about  $55 \text{ Gg yr}^{-1}$  and the chemical and pharmaceutical industries account for the remainder.
3. Whatever the source, chloroform partitions preferentially into the atmosphere; the equilibrium distribution is greater than 99%. The average global atmospheric concentration is  $18.5 \text{ pmol mol}^{-1}$  (approximating to parts per trillion by volume: 18.5 in  $10^{12}$ ). It is sparingly soluble in water and can be washed out of the air by rainfall. However, the principal removal process from the environment is atmospheric oxidation and the calculated quantity that reacts annually in this way (to form hydrogen chloride and carbon dioxide) is approximately in balance with the identified sources. It is unlikely that there are significant other sources.
4. Chloroform is also removed by anaerobic and aerobic soil microorganisms and, although not important to the global balance, this can have a profound effect on local concentrations that cannot equilibrate into the atmosphere.
5. By virtue of the very small amounts that actually become transported to the stratosphere, chloroform does not deplete ozone materially. Nor is it a photochemically active Volatile Organic Compound (VOC); it exhibits a very low potential to enter into the reactions that produce ground level ozone and photochemical smog (local urban smog). It has a Global Warming Potential that is less than that of the photochemically active VOCs and is not classed as a greenhouse gas.

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6. The concentrations observed in raw water and groundwater are consistent with the sources identified, coupled with occasional wash-out from the atmosphere. Chloroform is widely dispersed in the aquatic environment (even naturally present in some mineral waters). Consequently, it is also widely dispersed in the tissue of living creatures and in foodstuffs, but there is little evidence of bioaccumulation.
7. The quantities in foodstuffs and drinking water are not problematical for human ingestion even at the highest concentrations.
8. Definitive studies have shown that current environmental concentrations of chloroform do not present an ecotoxicological risk, even to fish at the embryonic and larval stages when they are most susceptible.

## 1. Properties of chloroform (trichloromethane, CHCl<sub>3</sub>)

Table 1. Physical properties (generally from [Crookes et al., 1994], unless stated otherwise)

Property	
CAS number	67-66-3
Molecular weight	119.39
Boiling point	61.3°C
Melting point	-63.5°C
Density	1.4832 g mL <sup>-1</sup> @ 20°C
Vapour density	4.36 rel. to air @ 0°C
Vapour pressure	13 kPa @ 10.4°C 27 kPa @ 25.9°C
Log K <sub>ow</sub>	1.97 @ 25°C
Water solubility	8.22 g L <sup>-1</sup> @ 20°C
Henry's constant (dimensionless)	0.09 <sup>1</sup> 0.13 <sup>2</sup> 0.36 <sup>3</sup>
Henry's constant <sup>4</sup>	314.8 Pa.m <sup>3</sup> .mol <sup>-1</sup> @ 20°C
Water/air partition coefficient k <sub>w/a</sub>	8.6 <sup>5</sup> @ 15°C

Notes:

<sup>1</sup> from *Nightingale* [1991] for open sea conditions and 11.6°C.

<sup>2</sup> from *Dyrssen* et al. [1990] for estuarine conditions at 14°C.

<sup>3</sup> from *Khalil* et al. [1999] for open ocean conditions and 17°C.

<sup>4</sup> from *Zok* et al., [1998]

<sup>5</sup> from *Pearson* [1982]

The relatively high value for k<sub>w/a</sub> permits rainfall to scavenge chloroform from the atmosphere [*Class and Ballschmiter*, 1986]. However, because of its low solubility in water and relatively high vapour pressure, chloroform in water that is allowed to equilibrate with its surroundings can be expected to partition into the atmosphere [*Ballschmiter*, 1992]. Partitioning was calculated to be 99.1% to air using a Mackay "level 1" model [*Mackay and Patterson*, 1990; *Zok* et al., 1998] and was demonstrated practically in sewers by *Haas and Herrmann* [1996] and in river estuaries by *Dyrssen* et al. [1990] and *Dewulf* et al. [1998].

## 2. Fluxes into the environment

The major fluxes of chloroform into the environment were surveyed for the Reactive Chlorine Emissions Inventory by *Keene* et al. [1999]. Oceanic sources predominated at 360 Gg, followed by soil sources at 220 Gg. By contrast, all industrial sources together came to 70 Gg.

### 2.1 Seawater sources

#### *Off-shore*

Open ocean emissions of chloroform were calculated from atmospheric measurements and observations of supersaturation in the water column and by inverse modelling from the distribution of concentrations in the atmosphere [*Khalil* et al., 1999; *Khalil and Rasmussen*, 1999]. The fluxes so identified were 20 Gg yr<sup>-1</sup> in the northernmost semi-hemisphere (30°N to

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90°N), 150 Gg yr<sup>-1</sup> in each of the tropical semi-hemispheres and 40 Gg yr<sup>-1</sup> from 30°S to 90°S. Despite the importance of this flux, this is the only estimate from open ocean waters, with no information on the processes that are involved in chloroform formation. Presumably these are biological.

Other measurements show high concentrations of chloroform in remote waters of the North Sea [Nightingale, 1991], but the same work also showed significant contamination from river water (eg. The Scheldt). Using the mean concentration over the whole of the North Sea study area, a global oceanic flux of 330 Gg yr<sup>-1</sup> was calculated, although this fell to 90 Gg yr<sup>-1</sup> if coastal waters were excluded. It is difficult to ascertain the importance of the flux introduced by river water relative to that formed naturally in shallow water and in the coastal-terrestrial region.

### *Littoral*

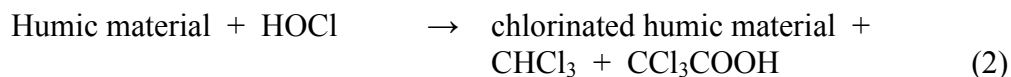
Compared to the oceanic sources, much more has been done to characterise littoral fluxes and it has been shown that the common brown seaweed *Laminaria saccharina*, red seaweeds *Gigartina stellata*, *Corallina officinalis* and *Polysiphonia lanosa* and green seaweed *Cladophora albida* all release significant quantities of chloroform into the surrounding water [Nightingale et al., 1995]. The formation of chloroform was considered to be intracellular with the mediation of chloroperoxidase. Based on this work, the global flux from sea shore kelp was calculated to be 0.23 Gg yr<sup>-1</sup>, although the authors point out that this is highly uncertain and that the real value could be much higher. Recent studies of seaweeds found off the north coast of East Anglia, UK, have confirmed the extent of generation of chloroform by coastal brown, red and green species at 0.25 Gg yr<sup>-1</sup> (global) [Baker et al., 2001]. Laturus and colleagues working with several species of Antarctic macroalgae described the production of methyl chloride and a number of bromine-containing methyl halides, but did not mention chloroform specifically [Laturus et al., 1998a; 1998b]. Other workers, however, have found very much larger amounts of chloroform to be emitted by red seaweeds in particular. The highest recorded flux was 2,400 ng g<sup>-1</sup> h<sup>-1</sup> from *Falkenbergia hillebrandii*, or 10,000 times the flux used to calculate the global generation rate reported by both Nightingale and Baker, above [Ekdahl, 1997].

Working in the coastal peatlands of western Ireland, Dimmer et al. [2000] showed significant halomethane emissions from these ecosystems and estimated a global annual flux of between 0.1 and 152 Gg yr<sup>-1</sup>. Enhanced atmospheric chloroform concentrations have also been observed to arise from natural coastal-terrestrial sources in Tasmania and the south eastern Australian mainland with maximum emissions in summer. The most likely sources in this region are from the Tasmanian west coast rainforest and button-grass moorlands [Cox et al., 2001].

The pattern of chloroform emissions shown by Lagrangian back trajectory modelling of the continuous atmospheric measurement at Mace Head, Ireland, gives further evidence that littoral sources are important [Ryall et al., 2001]. Significant chloroform emissions were identified in Ireland, Scotland and Scandinavia with little evidence of emissions from the populated regions of Europe; the total European emission strength was estimated to be 18 Gg yr<sup>-1</sup>.

## 2.2 Soil processes

The concentration of chloroform within soil pores is significantly higher than that in the surrounding air [Frank 1988; Frank et al., 1989; Frank and Frank, 1990]; the enhancement factor ranging from four times to 600 times. The source of this chloroform has been demonstrated to be chlorination of soil acids, mainly humic materials, by hypochlorous acid. This is generated from the chloride ion that is ubiquitous in soil, and hydrogen peroxide, in the presence of chloroperoxidase (CPO) enzyme.



Chloroperoxidase activity has been observed in many soil extracts [Asplund et al., 1993; Laturnus et al., 1995] and has been shown in the laboratory to catalyse the chlorination by hydrogen peroxide and chloride ion of simple organic compounds, such as acetone, propionic acid and citric acid, to chloroform [Walter and Ballschmiter, 1992]. Similar chlorination of humic acid in the laboratory was demonstrated by Hoekstra et al. [1995], who went on to show, by field studies and  $\text{Na}^{37}\text{Cl}$  enrichment of soil, that the production of chloroform is a natural process [Hoekstra et al., 1998a]. The source of chloroperoxidase enzyme is likely to be fungal; in laboratory studies chloroform production was detected from five varieties of basidiomycetes and one deuteromycete [Hoekstra et al., 1998b]. Seven of eight white rot fungi were shown to have chlorinating ability *in vitro* and also Wood Blewitt (fairy ring fungus, *Lepista nuda*) *in vivo* [Hjelm, 1996]. It has been suggested that the chloroform produced acts as a natural environmental stress agent, important to evolution [Urhahn and Ballschmiter, 1998]. The role of trichloroacetic acid in the production of chloroform in soil is not clear. On the basis of the equations above, it is a co-product, but the rate of production of chloroform in laboratory incubation studies was doubled by spiking the soils with trichloroacetic acid [Haselmann et al., 2000a], suggesting a possible influence of decarboxylation of the acid [Urhahn and Ballschmiter, 1998].

The conversion of inorganic chloride, present in fresh plant material, to aliphatic and aromatic chlorinated organic compounds as the material progresses naturally through senescence and humification in soil has been demonstrated by Myneni [2002]. Using X-ray absorption fine structure techniques, he showed the predominant forms to be: hydrated chloride ion in young and just yellowing leaves; hydrogen bonded chloride ion in reddish brown senescent leaves still connected to the plant; organically bound chlorine in reddish brown senescent and humified leaves in soil. While it is possible that this may be confounded by aqueous leaching of chloride ion, the results - when considered along with those described above - show that chlorination of organic compounds in humic materials is a fundamental process [Casey, 2002].

Field studies show a wide variation in the soil flux of chloroform, with the highest values being released by woodchip-covered forest soil ( $1,000 \text{ ng m}^{-2} \text{ hr}^{-1}$ ). Simultaneous air gradient analyses gave values similar to the mean flux calculated from soil measurements of about  $100 \text{ ng m}^{-2} \text{ hr}^{-1}$  from temperate Douglas fir forest [Hoekstra et al., 2001]. In Danish forest soils, rates of 4 to  $170 \text{ ng m}^{-2} \text{ hr}^{-1}$  were observed, while laboratory results from the same group indicated an expected flux of  $500 \text{ ng m}^{-2} \text{ hr}^{-1}$  [Haselmann et al., 2000b; 2000a]. Rice paddies in Asia have been shown to release  $300 \text{ ng m}^{-2} \text{ hr}^{-1}$  [Khalil et al., 1990].

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Based on these results, the global flux of chloroform from soil processes has been estimated at 220 Tg yr<sup>-1</sup> but with a relatively large uncertainty (range 100 to 400 Tg yr<sup>-1</sup>) [Khalil and Rasmussen, 1999; Khalil et al., 1999].

### *2.3 Other natural sources*

#### *Volcanic vents and mineral extraction*

While there is a significantly higher concentration of chloroform in the gases vented from solfataric volcanoes (at least 1000-fold over ambient) [Isidorov et al., 1990], the contribution of these sources to the global flux does not appear to be significant. On the other hand, the mining of silvinites and other potassium halides is calculated to release 10 to 15 Gg yr<sup>-1</sup> of chloroform, that was geologically associated with the halide ore, into the atmosphere [Isidorov et al., 1993].

#### *Landfills and ruminants*

Biogas generators, designed to anaerobically decompose wastes to generate methane, are also reported to generate chloroform [Khalil et al., 1990] with an emission ratio of  $2 \times 10^{-5}$  grams chloroform per gram of methane generated. Based on a global methane emission of 50 Tg from landfill and 100 Tg from ruminants [Bingemer and Crutzen, 1987; Hogan, et al., 1991], Aucott et al., [1999] concluded that 3 Gg yr<sup>-1</sup> of chloroform would be emitted from anaerobic processes producing methane.

### *2.4 Byproduct emissions from non-chemical industry and from water treatment*

Although these sources represent only a small fraction of the total emissions, their size and distribution is characterised better than any of the natural fluxes.

#### *Pulp and paper manufacture*

Chloroform is produced as a by-product during the delignification of wood and other cellulose pulps and the bleaching of paper by chlorine and other chlorine-containing oxidants, such as chlorine dioxide (ClO<sub>2</sub>) [Juuti et al., 1996]. Several reports [NCASI, 1988; Ayres, 1992; Blum, 1996; USEPA, 1984] provide information on the magnitudes of these emissions; calculations based on these reports, including an extrapolation of U.S. estimates to global estimates, suggest that yearly global emissions are in a range from 10 to 70 Gg yr<sup>-1</sup> CHCl<sub>3</sub>, with an approximate median of 30 Gg yr<sup>-1</sup> [Aucott, 1997]. Extrapolation of 1990 chloroform emissions from U.S. pulp and paper manufacturers, as reported on the U.S. Toxics Release Inventory [USEPA, 1994], provides a similar global estimate [Aucott, 1997].

Much variation exists in the ratio of chloroform released relative to the output of pulp and paper among US paper mills. The range is from  $2 \times 10^{-5}$  to  $1.78 \times 10^{-3}$  grams chloroform per gram of paper product, with a mean factor of  $1.38 \times 10^{-4}$  g g<sup>-1</sup> [Aucott et al., 1999]. This is lower than the draft EPA emission factor of  $1.95 \times 10^{-4}$  g g<sup>-1</sup> of air-dried pulp for chloroform emission from facilities that bleach with chlorine [USEPA, 1996].

Using the lower factor, the global total chloroform emission from the pulp and paper manufacturing sector was estimated to be 34 Gg yr<sup>-1</sup> [Aucott et al., 1999]. The uncertainty, based on U.S. emissions factors per mill as reported in the Toxics Release Inventory was estimated as  $\pm 8$  Gg yr<sup>-1</sup>.

*Drinking water and waste water treatment*

Chloroform and other tri-halogenated methanes are created when water is treated with chlorine through the haloform reaction involving humic compounds [Larson and Weber, 1994; Chaidou et al., 1999]. Dichlorine is not essential to the formation of chlorinated oxidation products from these organic acids; chloroperoxidase gives chloroform and trichloroacetic acid from humic material [Hoekstra et al., 1995; Niedan et al., 2000] and oxidation with  $\text{ClO}_2$  gives chloroform in almost the same way as  $\text{Cl}_2$  [Juuti et al., 1996]. In the first case, the sources of chlorine are chloride ions in solution or absorbed onto the humic substrate, so that the availability of chlorine as an oxidising agent does not determine the extent of chloroform formation. Furthermore, the abundance of chloroform relative to other trihalomethanes in treated water is highly variable. Golfinopoulos [2000] found chloroform at between 4 and 60% of the trihalomethanes in Greek drinking water; in Ireland similar determinations showed 10 to 44% [Stack et al., 2000] and in Florida chloroform amounted to 41 to 66% of trihalomethanes [Gibbons and Laha, 1999]. The other trihalomethanes contain both chlorine and bromine (up to tribromomethane) and, since none of the oxidising agents used contain significant bromine, the source must be bromide ions dissolved in the water or associated with the humic acid content.

Most potable water is treated with strong oxidising agent (such as chlorine or ozone) and trihalomethanes have been found to be ubiquitous (see Appendix 3); the observed average chloroform concentration of U.S. drinking water is approximately  $13,000 \text{ ngL}^{-1}$  [Krasner, et al., 1989]. However, water that has not been chlorinated, such as well-water or bottled mineral water, has also been shown to contain chloroform at approximately  $4000 \text{ ngL}^{-1}$  [Gibbons and Laha, 1999]. The authors ascribed this to errors in the sampling procedure, but the level detected amounted to between 1/4 and 1/2 of that found in samples of treated water, so this explanation does not seem sound.

Because of its generally lower humic content, waste water generates less chloroform when treated with chlorine than drinking water sources [USEPA, 1984]. With relatively low solubility in water and relatively high vapour pressure, the chloroform generated in water can be expected to partition into the atmosphere [Ballschmiter, 1992] and fluxes of chloroform have, indeed, been observed from potable water [Keating et al., 1997] and sewerage [Haas & Herrmann, 1996]. The processing involved in sewerage treatment generally reduces (by about 50%) the chloroform concentration in the water flowing through [Crookes et al., 1994]. In Europe, concentrations of chloroform in sewerage effluent were in the region of  $3,000 \text{ ngL}^{-1}$ , with a range of  $<1,000$  to  $20,000$ . In a survey of 50 treatment works in U.S.A., concentration in the influent ranged up to  $1,100,000 \text{ ngL}^{-1}$  with up to  $106,000 \text{ ngL}^{-1}$  in the effluent [Burns and Roe, 1982].

Based on quantities of water used and the U.S. EPA emission factor of 41 grams chloroform per  $10^6$  litres of chlorine-treated water [USEPA, 1984], overall fluxes of about 2 Gg chloroform per year for the U.S. and  $7 \text{ Gg yr}^{-1}$  for the rest of the world were calculated from potable water. Assuming that the quantity of publicly-supplied water and treated waste water is approximately the same, and using an emission factor of 14 grams per  $10^6$  litres of chlorine-treated waste water [USEPA, 1984], the chloroform produced globally from waste water treatment totals  $3 \text{ Gg yr}^{-1}$ . These sources together amount to  $12 \pm 7 \text{ Gg yr}^{-1}$  [Aucott et al., 1999].

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### *Cooling water and other water treatment*

Water used in cooling towers and other heat exchangers is typically treated with chlorine intermittently to remove slime organisms and other biota, such as mussels and, because cooling waters are mainly drawn from rivers which are likely to have similar humic matter content to drinking water sources, the emission factor used in the case of drinking water treatment also applies to cooling waters. This is likely to overestimate the chloroform emission; model studies have shown that, after dilution in receiving waters, the chloroform content is about 10% that of potable water [Mills et al., 1998].

Chloroform is also released from swimming pools treated with chlorine-containing compounds such as sodium hypochlorite (see Appendix 3). Concentrations in the water of Italian pools have been shown to range from 23,000 to 115,000 ngL<sup>-1</sup>, with up to 450,000 ngm<sup>-3</sup> in the air above the pool [Aggazzotti et al., 1995]. Similar concentrations (24,000 to 31,000 ngL<sup>-1</sup>) were recorded in Korean pools [Jo, 1994], with 28,000 to 34,000 ngm<sup>-3</sup> in the air above them; in the Korean cases, sodium hypochlorite and ozone were used together as disinfectants.

Apportionment of estimated quantities of chlorine used for cooling water and other water treatment, based on U.S. data [Jolley, et al., 1978; CMR, 1995a and b; CRA, 1993] suggests that approximately 70% as much chloroform is released from cooling and other water treatment as from drinking and waste water treatment [Aucott, 1997]. Assuming that a similar apportionment is valid globally, and that a similar uncertainty exists as for drinking and waste water treatment,  $9 \pm 6$  Gg yr<sup>-1</sup> of chloroform were released from cooling and other water treatment in the early 1990s [Aucott et al., 1999].

### *2.5 Chemical and pharmaceutical industries*

Chloroform is manufactured mainly in the U.S.A., European Union and Japan, the total global capacity in the late 1990s being about 520 Gg yr<sup>-1</sup> [Leder et al., 1998]. Its principal use is as a feedstock in chemical manufacturing, mainly to produce HCFC-22. Thus 96% of the quantity produced is converted to other materials and not released. The remainder is used as a solvent and extractant in various manufacturing and laboratory procedures and potentially may be released if it is not recovered for disposal.

Two manufacturing routes are employed: reaction of methanol with hydrogen chloride to form chloromethane and subsequent direct chlorination to yield *inter alia* chloroform, or direct chlorination of methane. In both cases, the processes operate under elevated pressure and are completely enclosed. Chloroform is also produced as a by-product of processes to make other chlorinated hydrocarbons in similarly enclosed processes. However, chloroform leaks into the environment in relatively small amounts both from these processes and when it is used as a chemical feedstock. A total of 419 Mg yr<sup>-1</sup> of chloroform emissions from HCFC manufacturers are listed in the US Toxic Release Inventory [USEPA, 1994].

The largest source of chemical industry emissions is from other manufacturing processes, particularly in the pharmaceuticals industry, in which chloroform is used as a solvent or process agent. These other industrial emissions have been estimated using a function derived from the data in the US Toxic Release Inventory and reported US consumptions of chloroform: 672 Mg total emissions from a consumption of 4.5 Gg [Leder et al., 1998]. Altogether the global release from industrial sources has been calculated to be 11 Gg yr<sup>-1</sup> with an uncertainty of 20% [Aucott et al., 1999].

The total calculated previously is markedly larger than that estimated, as a destruction rate based on atmospheric measurements, by Khalil and Rasmussen [1999] ( $470 \pm 120 \text{ Gg yr}^{-1}$ ). However it is consistent within the uncertainty and, as shown in Table 2, recalculation of the destruction rate produces a higher value that is closer to the emissions that were estimated directly. The hemispherical distribution for oceanic sources is taken from Khalil *et al.*, [1999] and, for other natural sources, two thirds of the emissions were considered to be from the northern hemisphere (consistent with the releases from terrestrial sources in Khalil *et al.*, [1999]). The non-natural sources were assumed to be predominantly in the northern hemisphere.

**Table 2. Chloroform emission fluxes: Global totals and estimated hemispherical distribution**

Source category	Emission $\text{Gg yr}^{-1}$	Uncertainty $\text{Gg yr}^{-1}$	Estimated hemispherical emission rates $\text{Gg yr}^{-1}$	
			NH	SH
<i>Natural sources</i>				
Open ocean	360	90	170	190
Soil processes	220	100	150	70
Geological contribution	12	3	8	4
Anaerobic fermentation	3	1	2	1
<i>Total natural</i>	595			
<i>Non-natural sources</i>				
Pulp and paper manufacture	34	8	32	2
Drinking water treatment	12	7	11	1
Cooling water treatment	9	6	8	1
Other industrial sources	11	2	10	1
<b>Total: all sources</b>	<b>660</b>	<b>220</b>	<b>390</b>	<b>270</b>

### 3. Observed occurrence of chloroform in the environment

#### 3.1 Air

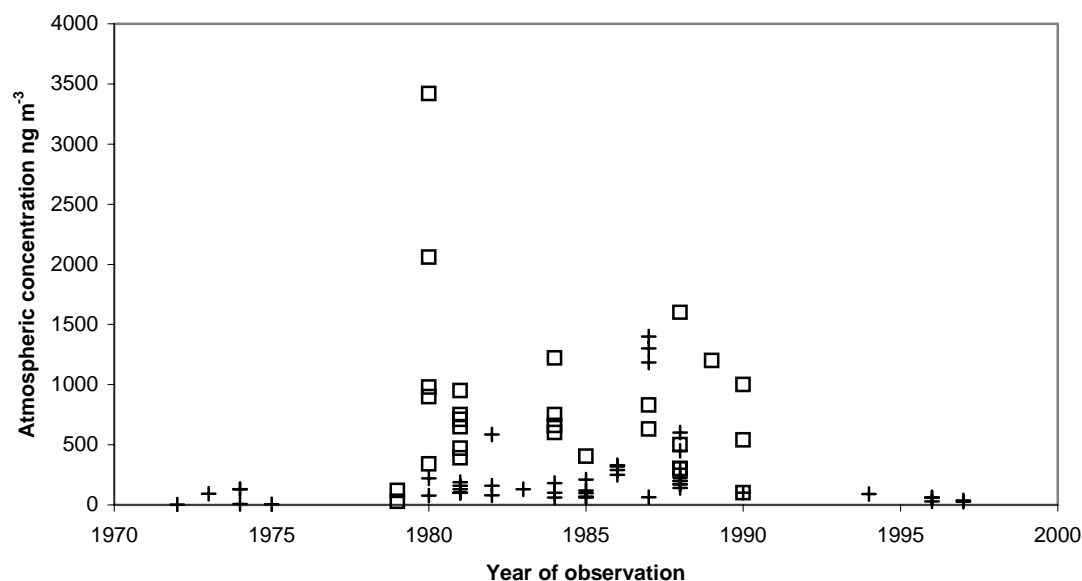
The comprehensive set of determinations is shown in Appendix 1 and, subdivided into background (or rural) and urban (or industrial), is also represented in Figure 1. Considerable overlap is evident between measurements in the two categories but the peak in concentration during the 1980s is more apparent than real: this was the period when most of the industrial and urban observations were made.

Global background concentrations are the principal source of information from which the major fluxes are calculated. Measurements at Cape Grim, Tasmania, indicated the average background concentration of chloroform to be apparently stable, in the long term, in the region of  $15 \text{ pmol mol}^{-1}$  ( $73 \text{ ngm}^{-3}$ ) [Fraser *et al.*, 1994]. However, other determinations show wide seasonal and geographical variations of the long-term average. Drawing on analyses from a number of laboratories, Khalil *et al.* [1999] showed that the mean concentration in the southern hemisphere was between  $12$  and  $15 \text{ pmol mol}^{-1}$  ( $59$ - $73 \text{ ngm}^{-3}$ ), but the concentration rose steadily from the equator northwards to become over  $30 \text{ pmol mol}^{-1}$  ( $146 \text{ ngm}^{-3}$ ) by

northern mid-latitudes. A mean global value of  $18.5 \text{ pmol mol}^{-1}$  ( $90 \text{ ngm}^{-3}$ ) in the lower troposphere was calculated. The average seasonal cycle in both hemispheres was  $3 \text{ pmol mol}^{-1}$ , calculated from long-term measurements, but local seasonal variations up to  $9 \text{ pmol mol}^{-1}$  are apparent from short-term measurements in the northern hemisphere [Khalil and Rasmussen, 1999].

The average chloroform concentration reduces with altitude: in northern high and mid latitudes, the concentration at the tropopause is 10 to  $15 \text{ pmol mol}^{-1}$  [Khalil et al., 1999]. However, at the tropical tropopause, the mean concentration was determined to be  $3.13 \pm 0.70 \text{ pmol mol}^{-1}$  [Schauffler et al., 1993]. The importance of this determination is in characterising the concentration that is carried with the main flow of air into the stratosphere, so enabling assessment of the extent of any effect of chloroform on stratospheric ozone.

Over land the variability is profound; Frank et al. [1991], analysing air from Madeira, the coast of Portugal and the Black Forest recorded median values from 41 to 123 and a range from 14 to  $1800 \text{ pmol mol}^{-1}$  ( $68\text{-}8700 \text{ ngm}^{-3}$ ). In earlier work they had showed similar values in Black Forest sites ( $51 \pm 29$  to  $68 \pm 31 \text{ pmol mol}^{-1}$ ) [Frank and Frank, 1988]. Measurements in rural areas of the United Kingdom ranged from 25 to  $120 \text{ pmol mol}^{-1}$  ( $120\text{-}590 \text{ ngm}^{-3}$ ) [Pearson and McConnell, 1975] and in the U.S.A., in New Jersey, the air concentration of chloroform ranged from 8 to  $900 \text{ pmol mol}^{-1}$  ( $39\text{-}4,400 \text{ ngm}^{-3}$ ) [Crookes et al., 1994].



**Figure 1.** Mean values of atmospheric concentrations of chloroform measured in remote (background) and rural locations (plus signs) and urban or industrial surroundings (open squares). Data from: Murray and Riley, 1973; Lovelock, 1974; Cox et al., 1976; Bätjer et al., 1980; Singh et al., 1982; Khalil et al., 1983; Harkov et al., 1984; Guicherit and Schulting, 1985; Singh et al., 1986; Frank and Frank, 1988; Levaggi et al., 1988; Frank et al., 1989; Kroneld, 1989; McAllister et al., 1989; Gilli et al., 1990; Bevan et al., 1991; Frank et al., 1991; McAllister et al., 1991a and 1991b; Singh et al., 1992; Sweet and Vermette, 1992; Crookes et al., 1994; Dewulf et al., 1998; O' Doherty et al., 2001.

The last reference contains a comprehensive list of measurements, all of similar magnitude and variability. Such measurements had led to the conclusion that industrial processes were a significant source of airborne chloroform, but the elevated levels in populated and forest covered regions of Europe and North America can be explained by the soil and water treatment sources identified above.

## *Chloroform in the environment*

Continuous monitoring of chloroform concentrations in the air of North Carolina and Massachusetts have enabled inversely modelled fluxes to be calculated from the eastern U.S.A.. In both experiments the flux was shown to be considerably larger than that estimated from identified anthropogenic sources, with the inference that soil processes account for the differences [Bakwin et al., 1997; Kleiman and Prinn, 2000].

### *3.2 Environmental water (see Appendix 2)*

#### *Rainfall, soil and groundwater concentrations*

The partitioning of chloroform between water and air, and the soil source of environmental chloroform, is demonstrated by results reported by McConnell [1976]: the waters of several upland reservoirs during relatively dry weather conditions, when a net outflow of chloroform from water to air is driven by volatility, contained less than 100 ngL<sup>-1</sup>. However, after prolonged heavy rain that, because of the small but significant solubility of chloroform, could be expected to scavenge it from the atmosphere, and would also wash out chloroform from its accumulation in soil water, the concentration in the same reservoirs averaged 20,000 ngL<sup>-1</sup>.

Rainwater chloroform concentrations were determined by Schleyer [1996] to be up to 97 ngL<sup>-1</sup> in the Black Forest of Germany, although previous studies in the same region had shown levels only between 11 and 17 ngL<sup>-1</sup> in open land [Schleyer et al., 1991]. There is a significant difference between the concentrations determined in forest clearings and that found under the tree canopy: median levels of 14 ngL<sup>-1</sup> against 56 ngL<sup>-1</sup>, respectively. The concentrations of chloroform in soil pore water follow an opposite tendency, with a range from 17-87 ngL<sup>-1</sup> in woodland and 30-540 ngL<sup>-1</sup> in the open. These observed concentrations are consistent with generation of chloroform *in situ* by soil micro-organisms (as described in 2.2) and the phenomenon of "guttation" whereby soluble materials are absorbed by plant roots, carried to the leafy extremities in the xylem system and then excreted onto the leaf surface, to be washed off by rainwater [Von Scheffer et al, 1965] (although guttation has not been demonstrated for chloroform, *per se*). The higher transpirational activity in forests could have the effect of diminishing the soil concentration of chloroform and, at the same time, increasing its concentration in rainfall beneath the forest canopy.

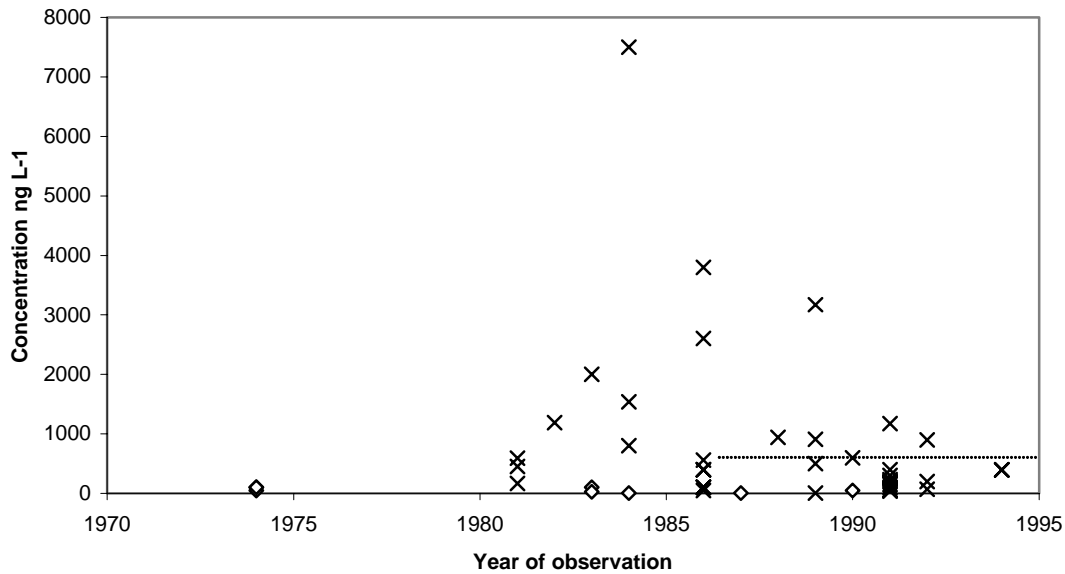
From below the water table, groundwater concentrations of chloroform in the region of 30 ngL<sup>-1</sup> were measured in the late 1980s and 200 to 500 ngL<sup>-1</sup> some five years later in similar locations [Schleyer et al., 1991; Schleyer, 1996]. The reason for the apparent difference is not clear but the higher more recent levels are consistent with the source of the groundwater being open land.

Potable well water contains up to 3,600 ngL<sup>-1</sup> in Germany [Schleyer, 1996], 4,000 ngL<sup>-1</sup> in Florida [Gibbons and Laha, 1999], 54,000 ngL<sup>-1</sup> in California and 61,000 ngL<sup>-1</sup> in Washington [Crookes et al., 1994]. The lower levels are consistent with the concentrations enhanced by rainfall reported by McConnell [1976] and the highest concentrations are likely to be geological [Isidorov et al., 1993], although they could also be the result of contamination.

#### *River water*

Apart from water courses that are clearly and grossly contaminated, such as a canal in UK that contained 177,000 ngL<sup>-1</sup> of chloroform [Crookes et al., 1994], the levels in most other freshwater samples are of the same order as those found in well water and, on average, greater than the quantities found in soil pore water reported above. Using the statistical methods

devised by Govaerts *et al.* [2001] on the data in COMMPS [1998], the mean concentration of chloroform in European rivers is calculated to be 500 ngL<sup>-1</sup>.



**Figure 2.** Mean concentrations of chloroform determined in lake water (open diamonds) and rivers (crosses). The line at 500 ng L<sup>-1</sup> represents the statistical mean for European rivers. Data from: *McConnell*, 1976; *W-Elbe*, 1982; *Ballschmitter et al.*, 1988; *Fahrni*, 1985; *FHH*, 1988; *Gewässergutebericht*, 1986; *Ott*, 1990; *Bohlen et al.*, 1989; *Malle*, 1990; *BLW*, 1991; *ARGE Elbe*, 1991; *RIWA*, 1993; *Fleig and Brauch*, 1991; *ARW*, 1994; *Crookes et al.*, 1994; *Zok et al.*, 1998.

**Table 3. Chloroform concentrations in river and lake water**

Country	Year	Minimum ngL <sup>-1</sup>	Maximum ngL <sup>-1</sup>	Reference
Belgium	1992		200	<i>RIWA</i> , 1993
Canada (Gt. Lakes)	1980s	18	830	<i>Crookes et al.</i> , 1994
France	1995	<1000		<i>Zok et al.</i> , 1998
Germany	1986-90	<100	3800	<i>Zok et al.</i> , 1998
The Netherlands	1990-92	<10	900	<i>RIWA</i> , 1998
Switzerland	1982-84	<10	1190	<i>Ballschmitter et al.</i> , 1998
UK	1993	<1000	>2000	<i>Zok et al.</i> , 1998
USA (Ohio)	1970s	4	2100	<i>Dreisch et al.</i> , 1991

## Seawater

**Table 4. Chloroform concentrations  
in seawater**

Location	Year	Minimum ngL <sup>-1</sup>	Maximum ngL <sup>-1</sup>	Reference
<i>Estuarine and Coastal</i>				
Elbe, Germany	1993	<10	90	<i>G-Elbe</i> , 1994
Ostsee, Germany	1983	60	170	<i>Hellmann</i> , 1984
Nordsee, Germany	1983	560	3800	<i>Hellmann</i> , 1984
Weser, Germany	1993	<20	200	<i>AR-Weser</i> , 1994
Rhine, NL	1993-95	5	91	<i>Zok et al.</i> , 1998
Scheldt, NL	1980s	10	5190	<i>van Zoest and van Eck</i> , 1991
Scheldt, NL	1993		150	<i>MVW</i> , 1994
Humber, UK	1992	<10	36.4	<i>Dawes and Waldock</i> , 1994
Liverpool Bay, UK	1970s		<1000	<i>Pearson and McConnell</i> , 1975
Mersey, UK	1987-90	2700	70000	<i>Rogers et al.</i> , 1992
Poole Harbour, UK	1992	<10	36.4	<i>Rogers et al.</i> , 1992
Solent, UK	pre 1991	10	7500	<i>Bianchi et al.</i> , 1991
Tees, UK	1992	<10	11500	<i>Rogers et al.</i> , 1992
Tyne/Wear, UK	1992	<100	242	<i>Rogers et al.</i> , 1992
<i>Open Ocean</i>				
North East Atlantic	1972		8	<i>Murray and Riley</i> , 1973
Eastern Pacific	1970s		15	<i>Su and Goldberg</i> , 1976
Gulf of Mexico	1990	40	200	<i>Sauer</i> , 1991
North Atlantic and Pacific	1980s	330	1090	<i>Khalil et al.</i> , 1983
North Atlantic	1985	1.6	7	<i>Class and Ballschmiter</i> , 1986

While there is clear evidence for industrial contamination in some of these determinations, it is also clear that the chloroform concentration in much of the estuarine water is similar to that in groundwater and the rivers feeding into the estuaries. Furthermore, it is a ubiquitous component of seawater.

### 3.3 Biota

As might be expected from the concentrations observed in most of the aqueous environment, chloroform is widely dispersed in botanical and zoological species. Concentrations in living tissue are reported in Appendix 4, summarised here, and those in prepared foodstuffs are reported in Appendix 5. All data are from the compendium in Crookes *et al.* [1994] and almost all are from the UK.

#### *Marine species*

There are no obvious correlations between chloroform concentrations in animal tissue and the position of the animal in the food chain or the organ sampled. Concentrations in plankton ranged from 0.02 ng.g<sup>-1</sup> in Liverpool Bay to 5 ng.g<sup>-1</sup> in Torbay (UK). Among molluscs, the lowest concentration was 3 ng.g<sup>-1</sup> (wet weight basis) in mussels (*Mytilus edulis*) and oysters (*Ostrea edulis*) in the Thames Estuary; the highest was 1040 ng.g<sup>-1</sup> (dry weight basis) in the

gill of a scallop (*Pecten maximus*) from the Irish Sea. The lowest crustacean concentration was 15 ng.g<sup>-1</sup> in a shore crab (*Carcinus maenas*) from the Firth of Forth and the highest, at 180 ng.g<sup>-1</sup>, both expressed as wet weight, in a specimen of *Cancer pagurus* from the same waters.

Individual tissues and organs were sampled in the determinations carried out on fish but there is no clear pattern. The lowest concentrations in the broad definition of flesh were 5 ng.g<sup>-1</sup> in sprat (*Clupea sprattus*) and mackerel (*Scomber scombrus*) from Torbay and the highest 110 ng.g<sup>-1</sup> in a spurdog (*Squalus acanthias*) from the Thames estuary (all wet weight). Concentrations in the flesh of some six other species of coastal fish were all in the range 20 to 50 ng.g<sup>-1</sup>. It is not clear how "flesh" differs from "muscle" but, within the latter narrower definition, dry weight concentrations ranged from 168 ng.g<sup>-1</sup> in a cod (*Gadus morhua*) to 649 ng.g<sup>-1</sup> in a dogfish (*Scylliorhinus canicula*), both from the Irish Sea. Only 6 ng.g<sup>-1</sup> (wet weight) was present in the liver of a flounder (*Platycthus flesus*) caught in Liverpool Bay, but 851 ng.g<sup>-1</sup> (dry weight) was present in the same organ of a coalfish (*Pollachius birens*) caught in the Irish Sea. A similarly broad range of concentrations is apparent in the organs in more direct contact with seawater: 50 ng.g<sup>-1</sup> in the gill of a conger eel (*Conger conger*) and 755 ng.g<sup>-1</sup> in the dogfish gill, 7 ng.g<sup>-1</sup> in the gut of a cod and 544 ng.g<sup>-1</sup> in the dogfish gut; all from the Irish Sea and all expressed on a dry weight basis.

Finally, the blubber of grey seals (*Halichoerus grypus*) from the Farne Islands contained between 7.6 and 22 ng.g<sup>-1</sup> of chloroform on a wet weight basis and common shrews (*Sorex araneus*) from a Merseyside location contained 41 to 66 ng.g<sup>-1</sup>.

#### *Aquatic birds*

Determinations on birds' eggs and the livers of mature examples are reported. The eggs have a very wide range of chloroform concentrations: from 0.7 ng.g<sup>-1</sup> (shag, *Phalacrocerax aristotelis*, Irish sea) to 65 ng.g<sup>-1</sup> (guillemot, *Uria aalge*, Irish sea), with almost as great a range within the same species. The liver samples have a smaller range: from 1.3 ng.g<sup>-1</sup> in a moorhen (*Gallinula chloropus*) to 17.3 ng.g<sup>-1</sup> in a kittiwake (*Rissa tridactyla*) from the same area of Merseyside.

#### *3.4 Foodstuffs*

Traces of chloroform are present in a wide variety of foodstuffs at highly variable concentrations; those in over 60 different foodstuffs are documented in Crookes *et al.* [1994] (Appendix 5). This list drew on several studies in the UK and U.S.A. In one study of 231 food samples, chloroform was detected in 94 at an average level of 52 ng.g<sup>-1</sup> [Daft, 1988]; in another it was detected in 302 out of 549 samples and the average level was 71 ng.g<sup>-1</sup> [Daft, 1989, 1991].

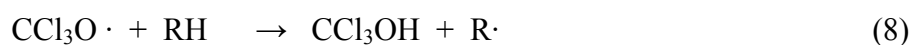
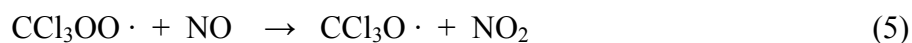
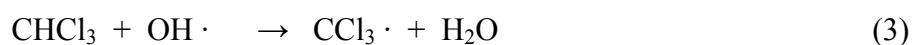
Given its lipophilicity, it should not be surprising that chloroform is present in butter, but there is no obvious explanation for the difference between English (22 ng.g<sup>-1</sup>) and American butter (80 - 1100 ng.g<sup>-1</sup>). Cheese concentrations are more consistent at 33 ng.g<sup>-1</sup> for cheese made from cows' milk in UK and 18 - 80 ng.g<sup>-1</sup> for a similar product in the U.S.A. [McConnell *et al.*, 1975; Heikes, 1987]. Chloroform is present in a variety of legumes, for example, lentils contained the lowest (6.6 ng.g<sup>-1</sup>) and almost the highest (57.2 ng.g<sup>-1</sup>) observed concentrations; among the 14 varieties of legume examined, only split peas were higher at 74.1 ng.g<sup>-1</sup> [Lovegren *et al.*, 1979].

Wheat is reported to contain between 105 and 3,400 ng.g<sup>-1</sup> (the latter is the highest for any foodstuff in the survey), bleached flour contains much less, 9.3 - 63 ng.g<sup>-1</sup> [Heikes and Hopper, 1986]. Both of these were determined in the U.S.A. and in the UK fresh bread contains only 2 ng.g<sup>-1</sup> [McConnell et al., 1975]. This sequence could be rationalised by the extent and temperature of processing as wheat is milled to flour and then baked to bread.

#### 4. Removal processes

##### 4.1 Air

Given that the equilibrium partitioning of chloroform is over 99% into air, atmospheric removal processes are the most important. Chloroform is oxidised by atmospheric hydroxyl radicals, present naturally at an average  $9.5 \times 10^5$  mol cm<sup>-3</sup> [Prinn et al., 2001]. The mechanism is typically:



The first step (equation 3) is rate determining, with a rate constant of  $8.1 \times 10^{-14}$  cm<sup>3</sup> molecule<sup>-1</sup> sec<sup>-1</sup> at 277 °K, so that the atmospheric lifetime of chloroform is 0.5 years [Prather, 1995]. Based on the concentrations reported in Khalil and Rasmussen [1999], the removal rates of chloroform by atmospheric oxidation are in the region of 410 Gg yr<sup>-1</sup> in the northern hemisphere and 190 Gg yr<sup>-1</sup> in the southern. Particularly in view of the vertical concentration profile, but also because of uncertainties in the reaction rate constant and the measurements themselves, these mass destruction rates have wide ranges: from 570 to 250 Gg yr<sup>-1</sup> (NH) and 260 to 120 Gg yr<sup>-1</sup> (SH). These calculations were performed using the simple two-box hemispherical model described in McCulloch and Midgley [1996] and so are illustrative, rather than substantive. Nevertheless, the global total is in good agreement with the emissions that were calculated directly in section 2 above, even if the distributions reflect a bias in the latter towards the northern hemisphere.

##### 4.2 Soil and sediment

Several methanogens have been shown in the laboratory to dechlorinate chloroform, particularly when promoted by metallic iron (Fe<sup>0</sup>) [Novak et al., 1998a; 1998b]. While this has little importance in the global mass balance of chloroform, it may be significant for groundwater; based on the kinetic data given for carbon tetrachloride degradation by the same organisms, the half-life for chloroform is about 12 hours. Although the product(s) were not

identified, the mechanism of the process, driven by an unidentified enzyme, suggests methane.

Chloroform is also degraded aerobically by *Methylosinus trichosporium*. In this case, the responsible enzyme is methane monooxygenase and the loss of chlorine follows insertion of oxygen into the C-H bond. The product is carbon dioxide [Bartniki and Castro, 1994].

#### 4.3 Water

Although chloroform hydrolyses readily in alkaline conditions [Hine and Dowell, 1954], the dependence of the rate on hydroxyl ion concentration means that, under the conditions prevailing in environmental water, its half-life is greater than 1,000 years and hydrolysis is not an effective sink. The principal removal process from water is volatilisation, as described in section 1 above.

### 5. Environmental effects

#### 5.1 Effects on atmospheric ozone

##### *Photochemical ozone formation at low altitude*

Due to its relatively slow reaction with hydroxyl radical, chloroform does not participate in the localised runaway atmospheric oxidations that give rise to photochemical ozone and peroxyacetyl nitrate (PAN) incidents near ground level. On a scale where ethene is 100, the photochemical ozone creation potential (POCP) of chloroform is 2.3 and, on a scale where propene is 100, its photochemical PAN creation potential (PPCP) is 1.7 [Derwent et al., 1998]. To place these values into context, ethane, which is regarded in US legislation as the boundary of such photochemical activity, scores 12.3 (POCP) and 17.3 (PPCP) and toluene, a recognised photochemically active VOC, scores 63.7 and 47.8. Hence, in the context of legislation on transboundary fluxes of air pollutants, chloroform is not a photochemically active VOC (volatile organic compound).

##### *Stratospheric ozone depletion*

Given the concentration of  $3.13 \text{ pmol mol}^{-1}$  determined at the tropical tropopause [Schauffler et al., 1993], chloroform contributes less than  $10 \text{ pmol mol}^{-1}$  to the total  $3,400 \text{ pmol mol}^{-1}$  of stratospherically active halogen (expressed as chlorine) that is advected through there into the stratosphere [Madronich et al., 1999]. The effect of the total quantity of chloroform released from land and seas is, therefore, about one third of a percent of the total effect on ozone. In view of the fact that non-natural releases are 10% of all chloroform emitted and an even smaller fraction is amenable to direct control, it is not classed as an ozone depleting substance under the Montreal Protocol.

#### 5.2 Climate change

With a Global Warming Potential (GWP) of only 4 (relative to  $\text{CO}_2$  at 100 years), chloroform is not classed as a greenhouse gas under the Kyoto Protocol [Albritton et al., 1996]. For comparison, the GWP of methane is 21 (other hydrocarbons: 11) on the same scale [Shine et al., 1990].

### 5.3 Ecotoxicity

Studies of toxicological tests on chloroform and comparisons with calculated and observed aquatic environmental concentrations have enabled the conclusion that "Levels of chloroform in surface water are generally low and would not be expected to present a hazard for aquatic organisms" [IPCS, 1994]. More specifically, after a study of 33 tests on aquatic organisms, Crookes *et al.* [1994] concluded that, while chloroform is moderately toxic to aquatic organisms, particularly at the embryo/larval stage of development, the general background concentrations of chloroform in UK waters is unlikely to affect these organisms adversely. Furthermore, with an environmental quality standard (EQS) of 12,000 ng L<sup>-1</sup>, even waters that are potentially contaminated should not pose a threat.

In a recent comprehensive evaluation of toxicological data, Zok *et al.* [1998] considered the results of 10 sets of data on algae, 17 on aquatic invertebrates and 23 on fish. Acute and chronic toxicity studies were taken into account and a Predicted No-Effect Concentration (PNEC) value of 72,000 ng L<sup>-1</sup> was defined (after allowing for a fifty-fold safety factor). The actual environmental levels in surface water shown in Tables 3 and 4 and Appendix 2 are currently very much lower than this.

### 5.4 Human health

The US Environmental Protection Agency concluded that chloroform is not likely to be carcinogenic to humans by any routes of exposure at a dose level that does not cause cytotoxicity and cell regeneration. This conclusion is supported by the finding that chloroform is not a strong mutagen and is not likely to cause cancer through a genotoxic mode of action. Hence daily exposure to 10,000 ng kg<sup>-1</sup> body weight (600,000 ng day<sup>-1</sup> for an adult person) is safe from the point of view of carcinogenicity [USEPA, 2001]. Based on a daily consumption of 1.5 kg food and 2 litres of water based beverages per person, the chloroform levels in drinking water (13 µg L<sup>-1</sup> [Krasner, *et al.*, 1989]) and foodstuff (52-71 µg.kg<sup>-1</sup> [Crookes *et al.* [1994]]) are of no concern.

## 6. Conclusions

The chloroform flux through the environment is constant at some 660 Gg yr<sup>-1</sup>, released and destroyed. About 90% of these emissions are natural in origin. The largest single source appears to be in offshore seawater (contributing 360 Gg yr<sup>-1</sup>), presumed to be natural, but from an undefined biological process. Littoral and coastal sources from macroalgae are much better defined, but do not appear to contribute much to the total release. Soil processes are the next most important source and the formation of chloroform has been shown to depend on oxidation of humic material in the presence of chloroperoxidase enzyme and chloride ion. The world's soils contribute about 220 Gg yr<sup>-1</sup>. Other natural sources, mainly volcanic and geological, account for less than 20 Gg yr<sup>-1</sup>.

The non-natural sources total 66 Gg yr<sup>-1</sup> and are predominantly the result of using strong oxidising agents on organic material in the presence of chloride ions, a direct parallel with the natural processes occurring in soils. The wood pulp and paper industry, together with drinking and other water treatments, account for about 55 Gg yr<sup>-1</sup> and the chemical and pharmaceutical industries account for the remainder.

Whatever the source, chloroform partitions preferentially into the atmosphere; the equilibrium distribution is greater than 99%. However, it is sparingly soluble in water and can be washed out of the air by rainfall. The average global concentration is  $18.5 \text{ pmol mol}^{-1}$  (approximating to parts per trillion by volume: 18.5 in  $10^{12}$ ). The principal removal process from the environment is atmospheric oxidation and the calculated quantity that reacts annually in this way (to form hydrogen chloride and carbon dioxide) is approximately in balance with the identified sources. It is unlikely that there are significant other sources.

Chloroform is also removed by anaerobic and aerobic soil microorganisms and, although not important to the global balance, this can have a profound effect on local concentrations that cannot equilibrate into the atmosphere.

By virtue of the very small amounts that actually become transported to the stratosphere, chloroform does not deplete ozone materially. Nor is it a photochemically active Volatile Organic Compound (VOC); it exhibits a very low potential to enter into the reactions that produce ground level ozone and photochemical smog. It has a Global Warming Potential that is less than that of the photochemically active VOCs and is not classed as a greenhouse gas. The concentrations observed in raw water and groundwater are consistent with the sources identified, coupled with occasional wash-out from the atmosphere. Chloroform is widely dispersed in the aquatic environment and is even naturally present in some mineral waters. Consequently, it is also widely dispersed in the tissue of living creatures and in foodstuffs but there is little evidence of bioaccumulation. The quantities in foodstuffs and drinking water are not problematical for human ingestion even at the highest concentrations.

Definitive studies have shown that current environmental concentrations of chloroform do not present an ecotoxicological risk, even to fish at the embryonic and larval stages when they are most susceptible.

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## Appendices

### APPENDIX 1. Atmospheric concentrations

Date	Location	Country	ng m <sup>-3</sup>			ng m <sup>-3</sup>		Comment (area)	Source (of data)
			mean	SD	n	min	max		
<i>Outdoor atmosphere</i>									
1970s	Merseyside	UK				11900	47400	Industrial	(2) in (1)
1970s	Merseyside	UK				3600	9500	Industrial	(2) in (1)
1970s		UK	4					Rural (unspecified)	(3) in (1)
1970s		UK				<120	590	Rural (unsp.)	(2) in (1)
1972	NE Atlantic		2					Oceanic	(3) in (1)
1973	North Atlantic		92					Oceanic	(4) in (1)
1974	NH background		130					Background	(5) in (1)
1974	SH background		<15					Background	(5) in (1)
1974	West Coast	Ireland	130					Coastal	(4) in (1)
1974-75	Brussels	Belgium				2390	14600		(6) in (1)
1975	Michigan	USA				100	150	Rural	(7) in (1)
1977-82	Cape Meares, OR	USA	220					Coastal	(8) in (1)
1979	Bremen	Germany	120						(9) in (1)
1979	Bremerhaven	Germany	30						(9) in (1)
	Pacific coastline	USA	83					Coastal	(10) in (1)
	Washington	USA	44					Rural	(11) in (1)
	Washington	USA				39	430	Urban	(11) in (1)
	Talladega	USA	490					Forest	(12) in (1)
1979-81	S. Pole	Antarctica	78					Remote	(8) in (1)
1980		The Netherlands	980			9.8	36600		(13) in (1)
1980	Houston, TX	USA	2060						(14) in (1)
1980	Denver, CO	USA	900						(14) in (1)
1980	Riverside, CA	USA	3420						(14) in (1)
1980	St. Louis	USA	340						(14) in (1)
1981	Staten Island, NY	USA	710						(14) in (1)
1981	Pittsburgh, PA	USA	470						(14) in (1)
1981	Chicago, IL	USA	390						(14) in (1)
1981	Barrow, AK	USA	190					Coastal/Oceanic	(8) in (1)
1981	Cape Kumukahi, HI	USA	160					Coastal/Oceanic	(8) in (1)
1981		Marshall Islands	130					Coastal/Oceanic	(8) in (1)
1981		Samoa	100						(8) in (1)
1981	Melbourne	Australia	100					semi-Rural	(8) in (1)
1981-82	Newark, NJ	USA				300 mean	1000 mean		(15) in (1)
1981-82	Elizabeth, NJ	USA				500 mean	1400 mean		(15) in (1)
1981-82	Camden, NJ	USA				200 mean	1300 mean		(15) in (1)
1982		Azores	160					Oceanic/Coastal	(16)
	New Jersey	USA	4400					Industrial	(17) in (1)
	New Jersey	USA				500 mean	1200 mean	Residential	(17) in (1)
1982-83	South Coast, CA	USA				390 mean	780 mean	Coastal	(18) in (1)
1982	Arctic	Norway	81						(19) in (1)
1983	Arctic	Norway	130						(19) in (1)
1983-85	6 cities	USA				280 mean	1220 mean		(20) in (1)
		NE Atlantic				59	110	Oceanic	(21) in (1)
		USA	200	med.				Rural	(22) in (1)
		USA	350	med.				Urban	(22) in (1)

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Date	Location	Country	ng m <sup>-3</sup>			ng m <sup>-3</sup>		Comment (area)	Source (of data)
			mean	SD	n	min	max		
		USA	4000				Industrial "source dominated"	(22) in (1)	
1984	Downey, CA	USA	660					(23) in (1)	
1984	Houston, TX	USA	1220					(23) in (1)	
1984	Denver, CO	USA	600					(23) in (1)	
1984		Madeira	180				Oceanic/Coastal	(16)	
1984		Madeira	100				Oceanic/Coastal	(16)	
1984		Madeira	60				High altitude/Oceanic	(16)	
1985	40° N	USA	98				Background	(23) in (1)	
1985	San Jose, CA	USA				310 mean	500 mean	(23) in (1)	
	Bayonne, NJ	USA				40 mean	130 mean	Industrial (24) in (1)	
	Los Angeles, CA	USA				30 mean	620 mean	(24) in (1)	
	Pittsburgh, CA	USA				30 mean	620 mean	(24) in (1)	
1985		Bermuda	60					Oceanic (16)	
1985	S. Atlantic		70					Oceanic (16)	
1985	N. Atlantic		120					Oceanic (16)	
1985	Bay of Biscay	N. Atlantic	210					Oceanic (16)	
1986-90	Chicago, IL	USA	300					(25) in (1)	
1986-90	St. Louis	USA	500					(25) in (1)	
1986	Mauzenberg	Germany	320	150				Forest (26)	
1986	Bernstein	Germany	250	140				Forest (26)	
1986	Sulzbach	Germany	290	100				Forest (26)	
1986	Schonbuch	Germany	330	150				Forest (26)	
1987	San Francisco Bay, CA	USA				10 mean	630 mean	Coastal (27) in (1)	
1987		Finland	95000					Industrial (28) in (1)	
1987		Finland	63					Suburban (28) in (1)	
1987	Mauzenberg	Germany	1300	329	6			Forest (29)	
1987	Bernstein	Germany	1400	337	4			Forest (29)	
1987	Schonbuch	Germany	1183	279	6			Forest (29)	
	Hamburg	Germany	360					(30) in (1)	
1987-88	Turin	Italy	830					Winter (31) in (1)	
1987-1990	Champaign IL	USA	300					Rural (25) in (1)	
1988	Mauzenberg	Germany	173	50	13			Forest (26)	
1988	Bernstein	Germany	200	122	13			Forest (26)	
1988	Sulzbach	Germany	225	116	12			Forest (26)	
1988	Schonbuch	Germany	171	90	14			Forest (26)	
1988	Turin	Italy	140					Summer (31) in (1)	
1988	19 sites	USA	1600					Urban (32) in (1)	
	Spitzbergen	Norway	98					Coastal (33) in (1)	
	Birkenes	Norway	73					Rural (33) in (1)	
1988-89	Tubingen	Germany	300	med.				City centre (34)	
1988-89	Rastatt	Germany	200	med.				Forest (34)	
1988-89	Lisbon	Portugal	600	med.				Coastal (34)	
1988-89		Madeira	450	med.				Coastal/Oceanic (34)	
1989	14 sites	Eastern USA	1200					Urban (35) in (1)	
1990	12 sites	USA	540					Urban (36) in (1)	
1990	Southampton	UK	1000					Roadside (37) in (1)	
1990	Southampton	UK	nd (<200)					Town centre (37) in (1)	
1990	Southampton	UK	nd (<200)					Semi-rural (37) in (1)	

## Appendices

Date	Location	Country	ng m <sup>-3</sup>			ng m <sup>-3</sup>		Comment (area)	Source (of data)
			mean	SD	n	min	max		
1980s & 90s	SH background NH background					50	100	Background	(38)
1986-1992	Cape Grim, Tas	Australia	73		continuous			Background	(39)
1994-1995	Scheldt Estuary	The Netherlands	90					Coastal	(40)
1994-98	Mace Head	Ireland	61	0.5	continuous			Background	(41)
1995-98	Trinidad Head, CA	USA	61	0.5	continuous			Background	(41)
1996-98	Ragged Point	Barbados	38	0.4	continuous			Background	(41)
1996-98	Cape Matatula	Samoa	27	0.2	continuous			Background	(41)
1994-98	Cape Grim, Tas	Australia	29	0.3	continuous			Background	(41)

### Soil pores

1992	Spuelderbos	The Netherlands	1148	396	4			Atmosphere = 140	(42)
1992	Spuelderbos	The Netherlands	548	103	4			Atmosphere = 190	(42)
1992	De Wieden	The Netherlands	2827	2269	3			Atmosphere = 150	(42)
1992	Fochteloo	The Netherlands	220					Atmosphere = 130	(42)
1994-95	Spuelderbos atmos.	The Netherlands	138	13	4			Atmosphere above soil	(43)
1994-95	Spuelderbos 10cm	The Netherlands	7383	4331	6				(43)
1994-95	Spuelderbos 20cm	The Netherlands	17500	2811	6				(43)
1994-95	Spuelderbos 40cm	The Netherlands	24000	4690	6				(43)
1994-95	Spuelderbos 80cm	The Netherlands	22833	3371	6				(43)
1994-95	Spuelderbos 120cm	The Netherlands	32000	6928	3				(43)
1994-95	Spuelderbos 160cm	The Netherlands	22000	5888	4				(43)

### Sources of data

- (1) Crookes et al., 1994; (2) Pearson and McConnell, 1975; (3) Murray and Riley, 1973; (4) Lovelock, 1974; (5) Cox et al., 1976; (6) Su and Goldberg, 1976; (7) Russell and Shadoff, 1975; (8) Khalil et al., 1983; (9) Bätjer et al., 1980; (10) Singh et al., 1977; (11) Harsch et al., 1977; (12) Holzer et al., 1977; (13) Guicherit and Schulting, 1985; (14) Singh et al., 1982; (15) Harkov et al., 1984; (16) Class and Ballschmiter, 1986; (17) Bozzelli and Kebbekus, 1982; (18) Shikiya et al., 1984; (19) Hov et al., 1984; (20) Singh et al., 1986; (21) Tille and Bachmann, 1987; (22) Brodzinsky and Singh, 1983; (23) Singh et al., 1992; (24) Hartwell et al., 1987; (25) Sweet and Vermette, 1992; (26) Frank and Frank, 1988; (27) Levaggi et al., 1988; (28) Kroneld, 1989a; (29) Frank et al., 1989; (30) Bruckmann et al., 1988; (31) Gilli et al., 1990; (32) McAllister et al., 1989; (33) Müller and Oehme, 1990; (34) Frank et al., 1991; (35) McAllister et al., 1991a; (36) McAllister et al., 1991b; (37) Bevan et al., 1991; (38) Khalil et al., 1999; (39) Fraser et al., 1994; (40) Dewulf et al., 1998; (41) O' Doherty et al., 2001; (42) Hoekstra and de Leer, 1993; (43) Hoekstra et al., 1998.

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### APPENDIX 2. Concentrations in environmental water

Date	Location	Country	ng L <sup>-1</sup>		Comment / location	Source
			mean	SD		
<i>Freshwater</i>						
1974	Rivington	UK	100		Upland reservoir without rain	(1)
1974	Blackstone Edge	UK	<100		Upland reservoir without rain	(1)
1974	Delph	UK	100		Upland reservoir without rain	(1)
1974	Rivington	UK	15000		Upland reservoir, prolonged rain	(1)
1974	Blackstone Edge	UK	21000		Upland reservoir, prolonged rain	(1)
1974	Belmont	UK	20000		Upland reservoir, prolonged rain	(1)
1974	Hollingworth	UK	11000		Upland reservoir, prolonged rain	(1)
1981	Elbe	Germany	594		Geesthacht	(3) in (2)
	Elbe	Germany	450		Wedel	(3) in (2)
	Elbe	Germany	168		Scharhoern	(3) in (2)
	Schuylkill R.	USA			<4 13500	Philadelphia (5) in (4)
	Ohio R., Huntingdon	USA			<4 2100	(5) in (4)
1982	Rhine	Switzerland	1190			(6) in (2)
1981-1983	rivers	Switzerland	62		1000	"Typical" (7) in (2)
	Welland R.	Canada			600	(8) in (4)
	St. Clair R.	Canada			5 4480	(9) in (4)
	Lower Niagara R.	Canada	22		110	(10) in (4)
	Lake Ontario	Canada	18		830	(10) in (4)
	Canal water	UK			12800 177000	(4)
1984	lakes	Switzerland			<10	"Typical" (7) in (2)
1983-85	Elbe	Germany	1540			Hamburg (11) in (2)
1983	Rhine	Germany	2000			Constanz (6) in (2)
1983	Bodensee	Germany	100			Lindau (6) in (2)
1983	Bodensee	Germany			<50	Uberlingen (6) in (2)
1984	Ruhr	Germany			150 15000	Duisburg (6) in (2)
1984	Mosel	Germany			500 1100	(12) in (2)
1985-89	Rhine	Germany	2600		9000	Hessen (13) in (2)
1985-89	Main	Germany	3800		12000	Hessen (14) in (2)
1986	Rhine	Germany			400	Bad-Honnef (12) in (2)
1986	Seig	Germany			<100	(12) in (2)
1986	Wupper	Germany			400	(12) in (2)
1986	Ruhr	Germany			100	(12) in (2)
1986	Emscher	Germany			100	(12) in (2)
1985-87	Unterweser	Germany	560		5000	(14) in (2)
1984-90	Bodensee	Germany			10 29	(2)
1988	Elbe	Germany	940		2700	(15) in (2)
1989	Inn	Germany			<16	Kirschdorf am Inn (16) in (2)
1989	Donau	Germany			<1017	Bofinger Halde (16) in (2)
1989	Donau	Germany	908			Jochenstein (16) in (2)
1989	Main	Germany	3170			Kahl am Main (16) in (2)
1990	Elbe	Germany	595			Schnackenburg (17) in (2)
1990-91	Ijsselmeer	The Netherlands			<100	Andijk (18) in (2)
1991	Rhine	The Netherlands	200			Lobith (18) in (2)
1991	Rhine	The Netherlands	300			Hagestein (18) in (2)

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Date	1991 Rhine Location	Germany Country	100 ng L <sup>-1</sup>		ng L <sup>-1</sup>		Karlsruhe (19) in (2) Comment / location	Source
			mean	SD	min	max		
	1991 Rhine	Germany	1170				Worms (19) in (2)	
	1991 Rhine	Germany	400				Koblenz (19) in (2)	
	1991 Rhine	Germany	230				Dusseldorf (19) in (2)	
	1991 Rhine	Germany	150				Bimmen (19) in (2)	
	1991 Weser	Germany	40				(2)	
	1991 Ems	Germany	60				(2)	
	1992 Meuse	Belgium	200				Tailfer (18) in (2)	
	1992 Meuse	The Netherlands	900				Eijsden (18) in (2)	
	1992 Meuse	The Netherlands	70				Keizersveer (18) in (2)	
	1993 26 monitoring stations	UK	3500			5000	(2)	
	1993 12 sites	UK				>2000	(2)	
	1993 17sites	UK			1000	2000	(2)	
	1993 181 sites	UK			<1000		(2)	
	1994 Rhine	Germany				390	Koln (20) in (2)	
	1994 Rhine	Germany				400	Wiesbaden (20) in (2)	
	European Rivers		500				Statistical analysis (21, 22)	

### Estuarine

1981-82	Untereibe	Germany	40					(6) in (2)
1983	Ostsee	Germany			60	170		(23) in (2)
1983	Nordsee coast	Germany			560	3800		(24) in (2)
	Liverpool Bay	UK			<1000			(24) in (4)
1986	Elbe	Germany			<20	1500		(25)
	Estuarine water	UK			<20	2400		(4)
1987-90	Mersey Estuary	UK			2700	70000		(26) in (2)
1988	Stenungsund	Sweden			5.4	14.8		(27) in (2)
	Scheldt Estuary	NL/B			10	5190		(28) in (4)
1990	Firth of Clyde	UK			9	13		(29)
	Solent	UK			10	7500		(30) in (2)
1992	Humber	UK			<10	36.4		(31) in (2)
1992	Poole Harbour	UK			<10	36.4		(31) in (2)
1992	Tees Estuary	UK			<10	11500		(31) in (2)
1992	Tyne & Wear	UK			<100	242		(31) in (2)
1992	Mersey Estuary	UK			28.3	88.9		(31) in (2)
1992	Other UK estuaries				<10			(31) in (2)
1993	Scheldt Estuary	NL/B	150					(32) in (2)
1993	Elbe	Germany			<10	90		(33) in (2)
1993	Weser	Germany			<20	200		(34) in (2)
1993-95	UK Estuaries				<25	1500		(35, 36) in (2)
1994-95	Scheldt Estuary	The Netherlands	73					(37)
1995	Seine	France			<1000			(2)

### Oceanic

1972	NE Atlantic		8					(38) in (4)
	E Pacific		15					(39) in (4)
	NH				330	1090	Open ocean	(40) in (4)

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Date	Location	Country	ng L <sup>-1</sup>		ng L <sup>-1</sup>		Comment / location	Source
			mean	SD	min	max		
1985	Madeira	N Atlantic	1.6					(41) in (2)
1985	W Africa	N Atlantic	1.6					(41) in (2)
1988	North Sea		4.8	1	0.3	84	Includes estuarine	(42)
	Gulf of Mexico				40	200		(43) in (4)
<i>Soil pore water, groundwater and well water</i>								
	Well water	USA			1700	4400		(44) in (4)
	36 sites	UK			<100	4600	Groundwater	(45) in (4)
	Rhine-Sieg area	Germany			<3000	0	Groundwater	(46) in (4)
	Washington	USA			nd	61000	Well water	(47) in (4)
1986-88	Birmingham	UK			nd	5000	Supply boreholes	(48, 49) in (4)
	Groundwater	UK	160					(4)
	Groundwater	UK	12800 and 20800				Contaminated site	(50) in (4)
	California	USA			200	54000		(51) in (4)
	Galicia	Spain			9000	48000		(52) in (4)
	Gibbstown	USA	28000 - 32000					(53) in (4)
	Coventry	UK	2100			21000	Supply boreholes	(54, 55) in (4)
1988-89	Morfelden	Germany	38				Soil pore, open land	(56)
1988-89	Konigstein	Germany	540				Soil pore, open land	(56)
1988-89	Grebenau	Germany	30				Soil pore, open land	(56)
1988-89	Witzenhausen	Germany	41				Soil pore, open land	(56)
1988-89	Morfelden	Germany	87				Soil pore, forest	(56)
1988-89	Konigstein	Germany	17				Soil pore, forest	(56)
1988-89	Grebenau	Germany	33				Soil pore, forest	(56)
1988-89	Witzenhausen	Germany	31				Soil pore, forest	(56)
1988-89	Morfelden	Germany	30				Groundwater	(56)
1988-89	Konigstein	Germany			27	190	Well water	(56)
1988-89	Grebenau	Germany			430	2100	Well water	(56)
1988-89	Witzenhausen	Germany			87	220	Well water	(56)
1993-94	Morfelden	Germany			0	240	Groundwater	(57)
1993-94	Konigstein	Germany			0	480	Well water	(57)
1993-94	Grebenau	Germany			240	3600	Well water	(57)
1993-94	Witzenhausen	Germany			60	240	Well water	(57)
1998	Florida	USA	3600				Well water	(58)

### *Rain and snow*

		UK			<200		Rain	(24) in (4)
		Alaska	94	20			Snow	(39) in (4)
1988-89	Morfelden	Germany	17				Rain, open land	(56)
1988-89	Konigstein	Germany	13				Rain, open land	(56)
1988-89	Grebenau	Germany	11				Rain, open land	(56)
1988-89	Witzenhausen	Germany	16				Rain, open land	(56)
1988-89	Morfelden	Germany	39				Spruce throughfall	(56)
1988-89	Konigstein	Germany	39				Spruce throughfall	(56)
1988-89	Grebenau	Germany	47				Spruce throughfall	(56)
1988-89	Witzenhausen	Germany	97				Spruce throughfall	(56)
	Black Forest	Germany	97				Rain	(57)

## Appendices

### Sources of data

(1) McConnell, 1976; (2) Zok et al., 1996; (3) W-Elbe, 1980-82; (4) Crookes et al., 1994; (5) Dreisch et al., 1981;  
(6) Ballschmiter et al., 1988; (7) Fahrni, 1985; (8) Kaiser and Comba, 1983; (9) Kaiser and Comba, 1986; (10) Kaiser et al., 1983;  
(11) FHH, 1988; (12) Gewässergutebericht, 1986; (13) Ott, 1990; (14) Bohlen et al., 1989; (15) Malle, 1990; (16) BLW, 1991;  
(17) ARGE Elbe, 1991; (18) RIWA, 1993; (19) Fleig and Brauch, 1991; (20) ARW, 1994;  
(21) COMMPS, 1998; (22) Govaerts et al., 2001; (23) Hellmann, 1984; (24) Pearson and McConnell, 1975;  
(25) Dyrssen et al., 1990; (26) Rogers et al., 1992; (27) Abrahamsson et al., 1989; (28) van Zoest and van Eck, 1991;  
(29) Nightingale et al., 1995; (30) Bianchi et al., 1991; (31) Dawes and Waldock, 1994; (32) MVW, 1994; (33) G-Elbe, 1994;  
(34) AR-Weser, 1994; (35) MAFF, 1995; (36) NRA, 1996; (37) Dewulf et al., 1998; (38) Murray and Riley, 1973;  
(39) Su and Goldberg, 1976; (40) Khalil et al., 1983; (41) Class and Ballschmiter, 1986; (42) Nightingale, 1991;  
(43) Sauer, 1991; (44) Bellar et al., 1974; (45) Folkard et al., 1984; (46) Schöler et al., 1984; (47) Suffet et al., 1985;  
(48) Rivett et al., 1990a; (49) Rivett et al., 1990b; (50) Eastwood et al., 1991; (51) Hadley and Armstrong, 1991;  
(52) Freiria-Gándara et al., 1992; (53) Rosen et al., 1992; (54) Burston et al., 1993; (55) Nazari et al., 1993;  
(56) Schleyer et al., 1991; (57) Schleyer, 1996; (58) Gibbons and Laha, 1999.

## Appendices

### APPENDIX 3. Concentrations in potable and treated water

Date	Location	Country	ng L <sup>-1</sup>		ng L <sup>-1</sup>		Comment	Source
			mean	SD n	min	max		
<i>Potable water</i>								
1970s		USA			37000	152000	Treated surface water	(1) in (2)
1970s	80 cities	USA	21000	med.	<100	311000	Drinking water	(3) in (2)
1970s		UK			700	38000	Tapwater	(4) in (2)
1970s		Japan			2600	17200		(5) in (2)
1970s	Salisbury	UK	400					(6) in (2)
1970s	Wareham	UK	300					(6) in (2)
1970s	Ringwood	UK	1300					(6) in (2)
1970s	Launceston	UK	34500					(6) in (2)
1980s	Dade Co. FL	USA	67000				2 yr. mean	(7) in (2)
1980s	Osaka	Japan			12500	37000		(8) in (2)
1980s	30 facilities	Canada			700	3400	Raw	(9) in (2)
1980s	30 facilities	Canada			21000	37000	Treated	(9) in (2)
1980s	Welland R.	Canada			7400	252000	Tapwater	(10) in (2)
1980s	97 public supplies	Nebraska USA	1800			6300		(11) in (2)
1980s	Bayonne NJ	USA	16000	med.			Winter	(12) in (2)
1980s	Bayonne NJ	USA	54800	med.			Summer	(12) in (2)
1980s	Los Angeles CA	USA	14500	med.			Winter	(12) in (2)
1980s	Los Angeles CA	USA	33000	med.			Spring	(12) in (2)
1980s	Pittsburg CA	USA	48900	med.			Spring	(12) in (2)
		UK	25000		2000	114000	River source	(2)
1990s	8 cities	Saudi Arabia			0	290 med.		(13) in (2)
1990s	Cairo	Egypt			500	96000		(14) in (2)
1990s	Cairo	Egypt			4200	13000		(15) in (2)
1990s		India	5210	med.				(16) in (2)
1990s		Ireland			<2800	21300		(17)
1993-95	Athens	Greece			5000	106000		(18)
1993-95	Mytilene	Greece			4000	27000		(18)
1993-95	Chalkida	Greece			5000	96000		(18)
1990s	Dade Co. FL	USA	7100				Tapwater	(19)
1990s	Broward Co. FL	USA	14800				Tapwater	(19)
1990s	Bottled water	USA	4000					(19)
1990s	Bottled water	USA	3800					(19)
1990s	Home filtration	USA	4900					(19)
1990s	Commercial filtration	USA	7300					(19)
<i>Waste water treatment</i>								
	Sewerage treatment	UK			1600	7000	Influent	(4)
	Sewerage treatment	UK			800	3000	Effluent	(4)
	50 treatment works	USA			1000	1100000	Influent	(20) in (2)
	50 treatment works	USA			1000	106000	Effluent	(20) in (2)
	50 treatment works	USA			1000	366000	Raw sludge	(20) in (2)
	Waste water treatment	The Netherlands			<100	90000 med.	Influent	(21) in (2)
	Waste water treatment	The Netherlands			<100	20000 med.	Effluent	(21) in (2)
	Sewerage treatment	USA			4000	4400	Influent	(22) in (2)

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Date	Sewerage treatment Location	USA Country	ng L <sup>-1</sup>		ng L <sup>-1</sup>		Effluent Comment	(22) in (2) Source
			mean	SD n	min	max		
	Digested sludge	UK	1300					(2)
	Sewerage treatment	USA			1000	7000	Influent	(23) in (2)
	Sewerage treatment	USA			2000	15000	Effluent	(23) in (2)

### *Swimming pools*

	Northern Italy				9000	145000		(24)
		Korea	23900	6600				(25)
			19500	7500				(25)
			31100	8500				(25)
		Ireland			87700	115600		(17)

### Sources of data

(1) Bellar et al., 1974; (2) Crookes et al., 1994; (3) Symons et al., 1975; (4) McConnell, 1977; (5) Fujii, 1977;  
 (6) Simmonds and Kerns, 1979; (7) Wood et al., 1980; (8) Kajino and Yagi, 1980; (9) Otson et al., 1982;  
 (10) Kaiser and Comba, 1983; (11) Goodenkauf and Atkinson, 1986; (12) Hartwell et al., 1987; (13) Fayad and Tawabini, 1991;  
 (14) Badaway, 1992; (15) El-Dib and Ali, 1992; (16) Thakkar and Vaidya, 1993; (17) Stack et al., 2000; (18) Golfopoulos, 2000;  
 (19) Gibbons and Laha, 1999; (20) Burns and Roe, 1982; (21) Van Luin and Van Starckenburg, 1984;  
 (22) Namkung and Rittman, 1987; (23) Michael et al., 1991; (24) Aggazzotti et al., 1995; (25) Jo, 1994.

## Appendices

### APPENDIX 4. Concentrations in biota - all from UK, reported in Crookes *et al.*, 1994.

Species		Location	Organ	ng/g	Comment	Source
<i>Marine species</i>						
Plankton		Liverpool Bay		0.02-0.09	wet weight	(1)
Plankton		Torbay		5	wet weight	(1)
Ragworm	<i>Nereis diversicolor</i>	Mersey Estuary		nd		(1)
Mussel	<i>Mytilus edulis</i>	Liverpool Bay		9-10	wet weight	(1)
Mussel	<i>Mytilus edulis</i>	Firth of Forth		8	wet weight	(1)
Mussel	<i>Mytilus edulis</i>	Thames Estuary		3	wet weight	(1)
Whelk	<i>Baccinum undatum</i>	Irish Sea	digestive gland	117	dry weight	(2)
Whelk	<i>Baccinum undatum</i>	Irish Sea	muscle	129	dry weight	(2)
Mussel	<i>Modiolus modiolus</i>	Irish Sea	digestive tissue	56	dry weight	(2)
Mussel	<i>Modiolus modiolus</i>	Irish Sea	mantle	438	dry weight	(2)
Mussel	<i>Modiolus modiolus</i>	Irish Sea	muscle	200	dry weight	(2)
Scallop	<i>Pecten maximus</i>	Irish Sea	gill	1040	dry weight	(2)
Scallop	<i>Pecten maximus</i>	Irish Sea	mantle	224	dry weight	(2)
Scallop	<i>Pecten maximus</i>	Irish Sea	muscle	440	dry weight	(2)
Scallop	<i>Pecten maximus</i>	Irish Sea	ovary	720	dry weight	(2)
Scallop	<i>Pecten maximus</i>	Irish Sea	testis	448	dry weight	(2)
Cockle	<i>Cerastoderma edule</i>	Liverpool Bay		4-150	wet weight	(1)
Oyster	<i>Ostrea edulis</i>	Thames Estuary		3	wet weight	(1)
Whelk	<i>Baccinum undatum</i>	Thames Estuary		10	wet weight	(1)
Slipper limpet	<i>Crepidula fornicata</i>	Thames Estuary		6	dry weight	(1)
Crab	<i>Cancer pagurus</i>	Tees Bay		nd		(1)
Crab	<i>Cancer pagurus</i>	Liverpool Bay		3-115	wet weight	(1)
Crab	<i>Cancer pagurus</i>	Firth of Forth		180	wet weight	(1)
Shore Crab	<i>Carcinus maenas</i>	Firth of Forth		15	wet weight	(1)
Hermit crab	<i>Eupagurus bernhardus</i>	Firth of Forth		73	wet weight	(1)
Hermit crab	<i>Eupagurus bernhardus</i>	Thames Estuary		20	wet weight	(1)
Shrimp	<i>Crangon crangon</i>	Firth of Forth		45	wet weight	(1)
Starfish	<i>Asterias rubens</i>	Thames Estuary		13	wet weight	(1)
Sunstar	<i>Solaster sp.</i>	Thames Estuary		3	wet weight	(1)
Sea urchin	<i>Echinus esculentus</i>	Thames Estuary		2	wet weight	(1)
Flounder	<i>Platycthyes flesus</i>	Liverpool Bay	flesh	21	wet weight	(1)
Flounder	<i>Platycthyes flesus</i>	Liverpool Bay	liver	6	wet weight	(1)
Eel	<i>Conger conger</i>	Irish Sea	gill	50	dry weight	(2)
Eel	<i>Conger conger</i>	Irish Sea	gut	43	dry weight	(2)
Eel	<i>Conger conger</i>	Irish Sea	liver	474	dry weight	(2)
Eel	<i>Conger conger</i>	Irish Sea	muscle	219	dry weight	(2)
Cod	<i>Gadus morhua</i>	Irish Sea	brain	167	dry weight	(2)
Cod	<i>Gadus morhua</i>	Irish Sea	gill	156	dry weight	(2)
Cod	<i>Gadus morhua</i>	Irish Sea	heart	67	dry weight	(2)
Cod	<i>Gadus morhua</i>	Irish Sea	liver	19	dry weight	(2)
Cod	<i>Gadus morhua</i>	Irish Sea	muscle	168	dry weight	(2)
Cod	<i>Gadus morhua</i>	Irish Sea	skeletal tissue	29	dry weight	(2)
Cod	<i>Gadus morhua</i>	Irish Sea	stomach	7	dry weight	(2)

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Species		Location	Organ	ng/g	Comment	Source
Coalfish	<i>Pollachius birens</i>	Irish Sea	alimentary canal	51	dry weight	(2)
Coalfish	<i>Pollachius birens</i>	Irish Sea	gill	294	dry weight	(2)
Coalfish	<i>Pollachius birens</i>	Irish Sea	heart	112	dry weight	(2)
Coalfish	<i>Pollachius birens</i>	Irish Sea	liver	851	dry weight	(2)
Coalfish	<i>Pollachius birens</i>	Irish Sea	muscle	168	dry weight	(2)
Dogfish	<i>Scylliorhinus canicula</i>	Irish Sea	brain	404	dry weight	(2)
Dogfish	<i>Scylliorhinus canicula</i>	Irish Sea	gill	755	dry weight	(2)
Dogfish	<i>Scylliorhinus canicula</i>	Irish Sea	gut	544	dry weight	(2)
Dogfish	<i>Scylliorhinus canicula</i>	Irish Sea	heart	210	dry weight	(2)
Dogfish	<i>Scylliorhinus canicula</i>	Irish Sea	liver	76	dry weight	(2)
Dogfish	<i>Scylliorhinus canicula</i>	Irish Sea	muscle	649	dry weight	(2)
Dogfish	<i>Scylliorhinus canicula</i>	Irish Sea	spleen	80	dry weight	(2)
Mackerel	<i>Scomber scombrus</i>	Liverpool Bay	flesh	50	wet weight	(1)
Mackerel	<i>Scomber scombrus</i>	Liverpool Bay	liver	18	wet weight	(1)
Mackerel	<i>Scomber scombrus</i>	Torbay	flesh	5	wet weight	(1)
Dab	<i>Limanda limanda</i>	Thames Estuary	flesh	23	wet weight	(1)
Plaice	<i>Pleuronectes platessa</i>	Thames Estuary	flesh	17	wet weight	(1)
Sole	<i>Solea solea</i>	Thames Estuary	flesh	26	wet weight	(1)
Sole	<i>Solea solea</i>	Thames Estuary	guts	9	wet weight	(1)
Red gurnard	<i>Aspitrigla cuculus</i>	Thames Estuary	flesh	21	wet weight	(1)
Red gurnard	<i>Aspitrigla cuculus</i>	Thames Estuary	guts	2	wet weight	(1)
Scad	<i>Trachurus trachurus</i>	Thames Estuary	flesh	48	wet weight	(1)
Pout	<i>Trisopterus luscus</i>	Thames Estuary	flesh	15	wet weight	(1)
Spurdog	<i>Squalus acanthias</i>	Thames Estuary	flesh	110	wet weight	(1)
Sprat	<i>Clupea sprattus</i>	Torbay	flesh	5	wet weight	(1)
Grey seal	<i>Halichoerus grypus</i>	Farne Islands	blubber	7.6-22	wet weight	(1)
Gannet	<i>Sula bassana</i>	Irish Sea	liver	7.4	wet weight	(1)
Gannet	<i>Sula bassana</i>	Irish Sea	eggs	1.9-2.0	wet weight	(1)
Shag	<i>Phalacrocerax aristotelis</i>	Irish Sea	eggs	0.7	wet weight	(1)
Razorbill	<i>Alca torda</i>	Irish Sea	eggs	6.6-19.7	wet weight	(1)
Guillemot	<i>Uria aalge</i>	Irish Sea	eggs	8-65	wet weight	(1)
Kittiwake	<i>Rissa tradactyla</i>	North Sea	eggs	58	wet weight	(1)
Kittiwake	<i>Rissa tradactyla</i>	Mersey Estuary	liver	17.3	wet weight	(1)
Kittiwake	<i>Rissa tradactyla</i>	Merseyside	kidney	8.4	wet weight	(1)
<i>Freshwater and land species</i>						
Moorhen	<i>Gallinula chloropus</i>	Merseyside	liver	1.3	wet weight	(1)
Moorhen	<i>Gallinula chloropus</i>	Merseyside	muscle	8.2	wet weight	(1)
Moorhen	<i>Gallinula chloropus</i>	Merseyside	eggs	19.5-29	wet weight	(1)
Mallard	<i>Anas platyrhynchos</i>	Merseyside	eggs	10-22	wet weight	(1)
Common shrew	<i>Sorex araneus</i>	Merseyside		41-66	wet weight	(1)

Sources of data:

(1) Pearson and McConnell, 1975; (2) Dickson and Riley, 1976.

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### APPENDIX 5. Concentrations in foodstuffs - all data from Crookes *et al.*, 1994.

Foodstuff	Country	ng g <sup>-1</sup>			Source
		mean	min	max	
<i>Dairy produce</i>					
Unspecified	USA			17	(1)
Fresh Milk	UK	5			(2)
Cheshire cheese	UK	33			(2)
Butter	UK	22			(2)
Hen's eggs	UK	1.4			(2)
Cheddar cheese	USA		46	80	(3)
Mozzarella cheese	USA		27	260	(3)
Cheese	USA		18	21	(3)
Parmesan cheese	USA		190	810	(3)
Butter	USA		80	1100	(3)
Milk	Finland	0.8			(4)
Butter and fats	USA	92	30	255	(5)
<i>Meat and fish</i>					
Unspecified	USA			<18	(1)
Unspecified fish	Finland	2.6			(4)
Unspecified meat	Finland	3.3			(4)
Beef steak	UK	4			(2)
Beef fat	UK	3			(2)
Pork liver	UK	1			(2)
Breaded shrimp	USA	24			(3)
Frozen chicken dinner	USA	29			(3)
"High meat" dinner	USA	17			(3)
Fish sticks	USA	47			(3)
Pot pie	USA	32			(3)
<i>Oils and fats</i>					
Unspecified	USA			12	(1)
Margarine	UK	3			(2)
Olive oil	Spain	10			(2)
Cod liver oil	UK	6			(2)
Vegetable cooking oil	UK	2			(2)
Peanut butter	USA	29			(3)
Margarine	USA		nd	50	(6)
<i>Cereals and cereal products</i>					
Bread	UK	2			(2)
Wheat	USA		105	3400	(7)
Corn	USA		5.2	7.1	(7)
Oats	USA	8.3			(7)
Corn meal	USA		5	7.3	(7)
Corn grits	USA	1.4			(7)
Corn muffin mix	USA	12			(7)
Yellow corn meal	USA	9			(7)
Fudge brownie mix	USA		16	17	(7)

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Foodstuff	Country	ng g <sup>-1</sup>			Source
		mean	min	max	
Lasagna noodles	USA	0.5			(7)
Bleached flour	USA		9.3	63	(7)
Uncooked rice	USA	3.6			(7)
Yellow cake mix	USA	300			(7)
Chocolate chip cookies	USA		10	22	(3)
Plain granola	USA	57			(3)
Raisin granola	USA	46			(3)
Corn chips	USA		15	70	(3)
Shredded wheat cereal	USA	42			(3)
Oat ring cereal	USA	120			(3)
Mixed cereal	USA		54	230	(3)
<i>Fruit and vegetables</i>					
Potatoes	UK	18			(2)
Potatoes	UK	4			(2)
Apples	UK	5			(2)
Pears	UK	2			(2)
Tomatoes	UK	2			(2)
Black grapes	unknown	nd			(2)
Hopi lime beans	USA	11.5			(8)
Jackson Wonder beans	USA	8.2			(8)
Speckled lima beans	USA	12			(8)
Fordhook lima beans	USA	15.6			(8)
Pinto beans	USA		6.1	19.7	(8)
Red kidney beans	USA	23.2			(8)
Black beans	USA	18.6			(8)
Navy beans	USA	19.9			(8)
Mung beans	USA	6.5			(8)
Soybeans	USA	12.8			(8)
Split peas	USA	74.1			(8)
Lentils	USA		6.6	57.2	(8)
Dried lima beans	USA	4.2			(7)
Scalloped potatoes	USA	7.1			(3)
Creamed corn	USA	6.1			(3)
<i>Beverages</i>					
Unspecified	USA			32	(1)
Unspecified juice	Finland	7.3			(4)
Coffee	Finland	5.4			(4)
Canned fruit drink	UK	2			(2)
Light ale	UK	0.4			(2)
Canned orange juice	UK	9			(2)
Instant coffee	UK	2			(2)
Packet (loose) tea	UK	18			(2)

Sources of data:

(1) Entz et al., 1982; (2) McConnell et al., 1975; (3) Heikes, 1987; (4) Kroneld, 1989b; (5) Crookes et al., 1994; (6) Entz and Diachenko, 1988; (7) Heikes and Hopper, 1986; (8) Lovegren et al., 1979.

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