# ENVIRONMENTAL BEHAVIOUR OF THE CHLORINATED SOLVENTS: A BRIEF OVERVIEW OF A GLOBAL PROBLEM

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**INTRODUCTION- COMPOUNDS OF CONCERN**

Considerable attention has been paid in recent years to the environmental effects of the organochlorine pesticides and other similar compounds such as the PCBs. Comparatively little attention has been directed at solvents, despite their known toxicity and the huge volumes in which they are used and discharged to the environment. The purpose of this paper is to discuss the way in which the chlorinated solvents behave on being released to the environment and to identify the areas of maximum impact.

Many of the widely used and potentially problematic of the volatile chlorinated compounds are listed in priority pollutant lists. Those compounds listed in MINDEC (1990) are given as an example and are primarily in use as solvents: *=priority substance for water; #= for air.

dichloromethane $CH_2Cl_2$ (methylene chloride)
* trichloromethane $CHCl_3$ (chloroform)
*# carbon tetrachloride $CCl_4$
*# trichloroethene $C_2HCl_3$ (trichloroethylene, TCE)
*# tetrachloroethene $C_2Cl_4$ (tetrachloroethylene, perchloroethylene, per, PCE)
* 1,2-dichloroethane $C_2H_4Cl_2$ (ethylene dichloride) (pesticidal use)
*# 1,1,1-trichloroethane (trike, TCA)
hexachloroethane
chlorobenzene
1,2-dichlorobenzene (o-dichlorobenzene, o-DCB)
1,3-dichlorobenzene (m-dichlorobenzene, m-DCB)
1,4-dichlorobenzene (p-dichlorobenzene, p-DCB)
*# trichlorobenzene
*# hexachlorobenzene

Naturally this list is not comprehensive; many volatile and semi-volatile halogenated compounds have been left out, but this does not mean that they are not toxic or potentially environmentally harmful. Some do pose a known threat. The environmental database is simply too incomplete to allow such judgements about many of this diverse group. Some are produced in relatively low quantities. Many are not in use as solvents and therefore outside the brief of this document. A broader overview of the problem is given by Johnston (1987). For example, dibromoethene has been investigated as a contaminant of fruit because of its carcinogenicity (Nakamura *et al* 1988). However, its presence in fruit is due to use as an insecticidal fumigant and not use as a solvent.

The chlorobenzenes are widely used as solvents; they were frequently used as the solvent for the PCBs in transformers. Chlorobenzene is frequently detected in industrial effluents. It is, for example, used as a dye carrier in the textile industry (Tagatz *et al* 1985). The behaviour of the chlorobenzenes in the environment, though, has more in common with the organochlorine pesticides than with the other solvents. Indeed hexachlorobenzene is employed in insecticidal use. Moreover, because of their much less volatile

nature, the fugitive emissions of these compounds are much less, though still significant. Consequently it may be more suitable to class them with the heavier organochlorines than with the volatiles. Work is likely to concentrate on those compounds which are most wideky used by industry. These will largely be the chlorinated methanes and ethanes.

However, some recent work has shown that the dichlorobenzenes may be present in sewage sludge at levels up to 50ppm. Generally the pattern of occurrence of the group as a whole reflected known industrial usage patterns (Rogers *et al*. 1989). Direct industrial discharges have also been identified as significant medium range point sources in the Forth Estuary (Rogers *et al* 1989) with concentrations in excess of 10ug/l in estuary waters. Also, Tagatz *et al*. (1985) report appreciable alterations in the community structure of aquaria spiked with trichlorobenzenes at concentrations as low as 10ug/l. They are a group of solvents which should not be ignored.

The four most widely used chlorinated solvents however are tetrachloroethene, trichloroethene, dichloromethane and 1,1,1-trichloroethane (Lawrence and Foster 1987, Herbert *et al* 1986). Their use peaked in the 1970s, apart from 1,1,1-trichloroethane whose use as an alternative to trichloroethene has increased dramatically in the recent past. Because of the obvious threats posed by this group, they have been much discussed. Production figures and basic toxicological information are given by Costner (1989).

**ATMOSPHERE**

Most emissions are either directly to the atmosphere (evaporation from paints, fugitive emissions etc) or indirectly from evaporation of emissions to surface waters. Between 60% (for tetrachloroethene) and 85% (for dichloromethane) of volumes consumed are released directly by evaporation (Lawrence & Foster 1987). These emissions could be easily reduced a great deal by minor improvements in equipment design as evidenced by the research of Wadden *et al* (1989). They report emissions of trichloroethene of 2.6g/min from and open-top degreaser unit as opposed to 0.67g/min from an enclosed degreaser. Nonetheless, whatever containment system is employed, eventual 100% loss is inevitable. Emissions of carbon tetrachloride are estimated at $9x10^4$ t/yr (Simmonds *et al* 1988). Waste treatment and storage facilities will give rise to significant emissions of volatile organic compounds (VOCs), including the semivolatile compounds such as chlorobenzene; see for example Shen & Sewell (1988), Wadden & Berrafato-Triemer (1989). There are also emissions of chloroform, 1,1,1-trichloroethane, trichloroethene and tetrachloroethene from cars which could be significant in urban areas (Gilli *et al* 1989). In addition, dichloroethane, which is used as scavenger in petrol, is thought to be a precursor to dioxin.

Long term monitoring of these compounds carried out in urban areas of Japan unsurprisingly showed higher levels of chlorinated organic compounds in industrial and commercial regions than in residential areas (Urano *et al* 1988). 1,1,1-trichloroethane and tetrachlorethene were particularly elevated in industrial regions

because of their use in degreasing and cleaning activities. Tetrachloroethene, which is used as a drycleaning agent as well as a degreaser, was high in commercial as well as industrial regions. These researchers also found seasonal variations in the atmospheric levels of some chlorinated organics despite their emission rates being constant. Chloroform and carbon tetrachloride levels were fairly constant, but 1,1,1-trichloroethane, trichloroethene and tetrachloroethene tended to be lower in summer than in winter and lower in dry than rainy seasons. These changes seem mainly to be due to changes in wind speed rather than changes in temperature, sunlight or rainfall, suggesting no seasonal variation in breakdown, but more a variation in the distribution of the pollutants from their sources. Frank *et al* (1989) acknowledge that levels in rural areas can at times exceed those in cities.

Atmospheric pollution by these compounds is now global. Measurements in the remote Pacific Ocean atmosphere show the longer lived compounds at background concentrations of 137pptv for carbon tetrachloride and 108pptv for 1,1,1-trichloroethane (DeLorey *et al* 1988).

A breakdown of the emissions of volatile hydrocarbons in the vicinity of Chicago is given by Aronian *et al* (1989). The solvent found in the highest levels was 1,1,1-trichloroethane, with an average concentration in the urban area of 6.7 ug/m3. Other urban air concentrations were (in ug/m3): chloroform, 4.6; tetrachloroethene, 2.6; trichloroethene, 1.3; carbon tetrachloride, 0.6.

Assessment of the levels of various solvents is often confounded by the tendency of researchers to report only the group parameter of VOC (eg Shen & Sewell 1988). However, Shah & Singh (1988) have assembled a database of atmospheric concentrations of volatile organic compounds in the USA. This gives a great deal of data on indoor and outdoor levels; median values for some of the more common solvents are given below (in ppbv).

| Compound | Outdoor levels | Indoor levels |
|---|---|---|
| carbon tetrachloride | 0.122 | 0.0 |
| chloroform | 0.058 | 0.104 |
| 1,1,1-trichloroethane | 0.162 | ---- |
| dichloromethane | 0.774 | ---- |
| trichloroethene | 0.158 | 0.125 |
| chlorobenzene | 0.061 | ---- |
| tetrachloroethene | 0.350 | 0.737 |

Most degradation takes place in the troposphere, the primary mechanism being oxidation by -OH radicals, leading to the formation of ozone as a byproduct. For this reason, some of the more reactive solvents require regulation because of their ozone production potential (Arnts *et al* 1989). Half-lives for the more common solvents are generally in the in the region of 0.2 years, apart from 1,1,1-trichloroethane which can last as long as 5 years (Lawrence and Foster, 1987). This extended lifetime for 1,1,1-trichloroethane allows it to reach the upper atmosphere where it

can become involved in ozone depletion. Carbon tetrachloride, with an atmospheric halflife of some 40 years, is also of concern because it is involved both in stratospheric ozone depletion and the greenhouse effect (Simmonds *et al* 1988).

The solvents, like other pollutants, are removed from the atmosphere by the mechanisms of dry and wet deposition. Wet scavenging, whereby solvents in gaseous form become dissolved in falling rain, is the primary mechanism of solvent deposition (Ligocki *et al* 1985, Atlas & Giam 1988). In this way, aquifers and surface waters can become contaminated but such problems are more often due to landfill leaching or industrial discharge. More significantly Frank *et al* 1989) consider that wet deposition may also be responsible for the elevated levels of volatile HHCs found in the atmosphere on high ground in forest environments. There is evidence to suggest that these compounds are involved in forest decline which is more extensive on high ground. These researchers found high levels of chlorinated solvents and CFCs in the air 30cm below the ground, where adverse effects upon the trees' root systems might be exerted.

In another study, Frank & Frank (1989) found tetrachloroethene in pine needles at bioconcentration factors of between 50 and 520 over atmospheric concentrations, indicating another possible toxicological pathway. In this context it must be remembered that as many as 209 organic compounds including 16 HHCs have been identified in forest atmospheres (Helmig *et al* 1989). The possible synergies and interactions of this number of compounds add considerably to the difficulties inherent in attributing effects to any particular compound or group of compounds.

**WATER**

The following compounds are recognised as priority substances for water among North Sea states (MINDEC 1990) : chloroform, carbon tetrachloride, trichloroethene, tetrachloroethene, 1,2-dichloroethane and 1,1,1-trichloroethane. Interestingly, chloroform was removed from the proposed UK "redlist" in the UK after consultation with industry (DoE 1988).

There is little information on the chlorinated solvents in waters as compared to the heavier organochlorines. Data on background levels of chlorinated methanes and ethanes in surface waters are not available, though is likely that they occur in measurable quantities. Similarly, little research is available on the toxicity of the solvents to aquatic life. That which exists suggests that at the high levels tested, anaesthetic effect of an additive nature was the primary mode of action.

Few chronic or sublethal data are available for the $C_1$ or $C_2$ compounds, probably partly as result of their volatility and the difficulty of keeping them in the test waters. The paucity of data is indicative of the low priority attached to the study of chlorinated solvents in environmental media other than air or groundwater. Effort directed at their study has been almost entirely focussed on these two fields, leaving a large gap in the data available. However, in relation to surface waters adjacent to

known point source discharges, they have been reported at high levels. For example tetrachloroethene (up to 190,000 ng/l) and chloroform (up to 5000 ng/l) were recorded in the St Lawrence River (Allan 1988). The possibility of wet scavenging or dry deposition implies a potential interaction with the marine microlayer, already identified as a potential sink for these types of compounds in laboratory studies (Sodergren *et al* 1990).

Some acute toxicity data is summarised below.

| Compound | Species | $LC_{50}$(mg/l) | Source |
|---|---|---|---|
| 2-chloroethane | P. promelas | 39.5 | Phipps & Holcombe (1985) |
| hexachloroethane | P. promelas | 1.23 | " " " |
| 1,2-dichloroethane | P. promelas | 118.0 | Benoit et al (1982) |
| tetrachloroethylene | P. promelas | 13.4 | Walbridge et al (1983) |
| 1,1,2,2-tetrachloroethane | P. promelas | 20.4 | " " " |
| pentachloroethane | P. promelas | 7.34 | " " " |
| 1,1,2-trichloroethane | P. promelas | 81.6 | " " " |
| 1,1,2-trichloroethene | P. promelas | 45.0 | " " " |
| 1,2-dichloroethane | P. promelas | 116.0 | " " " |
| hexachloroethane | P. promelas | 1.51 | " " " |
| chlorobenzene | D. magna | 12.9 | Gersich et al (1986) |
| chloroform | D. magna | 65.7 | " " " |
| p-dichlorobenzene | D. magna | 11.6 | " " " |

**SEDIMENTS**

Because of the volatility and comparatively high solubility of the chlorinated solvents, they will not tend to partition to aquatic sediments in the same way as the chlorinated pesticides. They will however be present in some regions at least in concentrations which could exert an effect on benthic life and organisms such as flatfish which contact the sediments over prolonged periods. Toxic effects at this level would have ramifications for the local ecosystem of unknown magnitude. The situation regarding research information on the chlorinated solvents in sediments is, if anything, worse than that for surface waters. Occurrence will only be at elevated levels in the vicinity of pollution sources but can still give a slightly longer-term indication of contamination problems and discharges variety than analysis of surface waters.

Sediment analysis therefore could be used, albeit on a more local basis, as a tool for assessing anthropogenic contamination of an area, as is currently done for other contaminants. Some data currently available are those reported by Johnston & Stringer (1989) which relates to the highly contaminated Weston Canal in northwest England. The following compounds were detected in the samples; the maximum concentrations are given in terms of ppm wet weight: dichloroethane (3), chloroform (60), carbon tetrachloride (300), trichloroethene (1), tetrachloroethene (30), hexachloroethane (trace), hexachlorobutadiene (6), dichlorobenzene (1) and hexachlorobenzene (250). There is a developing literature on the biodegradation of the solvents in sediments. This research seems

to have been largely prompted by groundwater and soil remediation concerns.

**SOIL/GROUNDWATER**

In the longer term, groundwater contamination is one of the most potentially harmful of the contamination problems associated with the solvents. Many populations take their drinking water from aquifers and contamination will need to be cleaned up or alternative sources found, with huge economic repercussions. It is estimated that 50% of the US population depends on groundwater for its drinking water (Waterstone, 1987). Contaminated groundwater can also act as sources of pollution to surface waters through seepage into streams, lakes and spring discharges.

The topsoil can act as a barrier to solvent contamination of the underlying strata. Firstly, it contains a large bacterial population which can break pollutants down. Also, the large content of organic matter and the large surface area can retain the compounds significantly by sorption and surface tension. Additionally, this retention enhances the time available for biodegradation and evaporative processes to occur, decreasing the eventual overall impact on the aquifer (Lawrence & Foster 1987). These processes have nonetheless failed to protect many aquifers and such contamination is of growing concern.

Major sources of pollution to groundwater are landfills, leaking tanks/pipes, spillages, soakaway drainage, lagoons, well injection, sewage landspreading/infiltration and leakage from other aquifers. Surface water, irrigation and rainfall are comparatively minor sources.

Where point sources discharge significant quantities, the solvents may form an immiscible layer separate from the main body of water. This is because of the low solubility of the solvents, and their different physical characteristics (ie, denser and less viscous than water). This layer may penetrate deeper and faster than the aqueous phase through most types of subsurface media. In addition, no biological degradation will occur in this layer; it is restricted to the aqueous sections where concentrations are low enough for microorganisms to survive.

Once an aquifer has become contaminated it may remain contaminated for hundreds, perhaps thousands of years. It should also be remembered that because of the low guideline values, it may only take a few litres of solvent to contaminate millions of gallons of groundwater. Water flow can be anything from a fraction of a metre per year to several metres per day. In fissured aquifers, transport can be even faster. Natural degradation of the solvents can occur both biologically or abiotically, but both take place at such slow rates it is only of very low significance in remediation terms.

Remediation of groundwater problems can be tried principally by one of three methods, physical containment, removal and treatment of water, and *in situ* treatment whereby the contaminants are encouraged to degrade underground. Different sites require exten-

sive investigation of the hydrology and chemistry of the site to assess the best cleanup strategy for a contaminated aquifer (see eg, Bouwer *et al*, 1988, Edworthy, K.J. 1987). Physical containment, either by hydraulic means or by the imposition of a plastic or similar barrier, is hardly to be recommended. It is an inadequate and temporary measure at the best of times, and is only appropriate in any case for the shallowest of aquifers.

*In situ* methods generally comprise the introduction of suitable substrates to the aquifer, in order to facilitate biological degradation by the native microorganisms. This method is most suitable for comparatively mildly contaminated sites, since biodegradation is at best slow and partial (Mayer *et al* 1988).

The third option is to pump the water from the aquifer as normal and treat it at the surface, either by air stripping or biodegradation. Trichloroethene and tetrachloroethene at low levels can also be broken down by treatment with ozone and gamma radiation (Gehringer *et al* 1988). There is considerable research into the biodegradation of the solvents (eg Oldenhuis *et al* 1989), but it is still in its infancy as an industrial technique. Transformation can still take place only at low concentrations and then it is often slow or incomplete. Trichloroethene degradation, for example, is inhibited at above 25ppm for some bacterial strains and the accumulation of toxic products of partial degradation, such as dichloroethenes or vinyl chloride, sometimes occurs (Oldenhuis *et al* 1989). Also, co-contaminants can interfere with biodegradation (Sholz-Muramatsu *et al* 1988), making it very difficult to predict how laboratory studies will relate to actual operational conditions.

**HUMAN EXPOSURE & EFFECTS**

Several of the organochlorine solvents are either known or suspected carcinogens. The following table gives New York State Department of Health (NYDH) and US Environmental Protection Agency (EPA) carcinogenicity potencies after Bro *et al* 1987.

| SUBSTANCE | NYDH | EPA |
|---|---|---|
| tetrachloroethene | 0.016 | 0.04 |
| trichloroethene | 0.008 | 0.013 |
| carbon tetrachloride | 0.008 | 0.083 |
| 1,2-dichloroethane | ----- | 0.037 |

(Potencies are expressed as the inverse of mg of contaminant per kg of body weight per day dosed: $(mg/kg/day)^{-1}$. It assumes that a 70kg individual is exposed over a 70-year lifetime; values are upper 95% confidence limits of estimated potencies.

Other major biochemical effects are liver and kidney damage and CNS depression. Narcotic effects are particularly notable at high concentrations.

The majority of the solvent burden is rapidly excreted unchanged in the breath (Droz *et al* 1988) and metabolites are excreted in

the urine or via the breath. What proportion remains in the body is generally distributed in the fatty tissues (Fawell & Hunt, 1988).

The World Health Organisation (1984) has set guideline values for certain of the chlorinated solvents in drinking water. They are listed below, and are based on a 70kg individual consuming two litres of water per day. For those compounds of concern for which sufficient data was not available, tentative guidelines have been set. Guideline values have not been set for dichloromethane, 1,1,1-trichloroethane, 1,2-dichloroethene, vinyl chloride, chlorobenzenes other than hexachlorobenzene, or the trihalomethanes other than chloroform.

WHO guidelines for chlorinated solvents in drinking water

| Contaminant | Guideline value (ug/l) |
|---|---|
| 1,2-dichloroethane | 10 |
| 1,1-dichloroethene | 0.3 |
| chloroform | 30 |

Tentative guideline values:

| | |
|---|---|
| Carbon tetrachloride | 3.0 |
| tetrachlorethene | 10.0 |
| trichlorethene | 30.0 |

Major exposures take place by inhalation, both at home and at work. Wallace (1987) estimates that the atmospheric route provides more than 99% of the exposure of people to the chlorinated solvents and other VOCs. Andelman (1985) identified evaporation from contaminated tapwater as a major source of indoor atmospheric contamination and therefore human exposure. McKone (1987) estimates that the amount of solvents from contaminated tapwater inhaled exceeds the amount ingested by a factor of between 1.5 and 6. Inhaled material will primarily reach the peripheral circulatory system where it will have its most intense effects on the kidneys. In the case of trichloroethene, for example, one of the major problems with chronic exposure is nephrotoxicity, with effects being visible in men occupationally exposed to around 15ppm (Nagaya *et al* (1989).

Urano *et al* 1988) have calculated the yearly atmospheric exposure of individuals exposed to ambient levels of chlorinated organic solvents in urban areas of Japan. They calculate that daily intake from atmospheric sources exceeds that implied by the WHO or US EPA recommended levels for drinking water at many sites. Trichloroethene intakes were exceeded at four of the fifteen sites monitored, carbon tetrachloride at ten and tetrachloroethene at all fifteen.

Wallace *et al* 1989 investigated activities and personal exposures

to a variety of volatile organic compounds. Chloroform exposure was found to be mainly a result of washing dishes, because of the content of the hot water. Visitors to drycleaners were exposed to elevated levels of 1,1,1-trichloroethane and tetrachloroethene and persons working in laboratories were exposed to a number of compounds. Indoor air concentrations of p-dichlorobenzene, trichloroethene, 1,1,1-trichloroethane and carbon tetrachloride accounted for high exposure in some individuals, though no sources were identified.

Whilst the inhalation route is naturally of primary concern, the ingestion route and its possible implications should not be discounted. Ingestion is predominantly through drinking water though various foodstuffs will be contaminated either through processing or atmospheric transport. Fawell and Hunt (1988) note that levels of 1,1,1-trichloroethane of 10ppb in butter, 16ppb in steak and 60ppb in tea have been recorded. They suggest that this could be due to the use of 1,1,1-trichloroethane as a solvent during food processing. This compound has also been used in coffee preparation, though it is often replaced by methylene chloride. Coughlin (1987) investigates the use of methylene chloride in the production of decaffeinated coffee where residues of up to 10ppm are allowed, despite its being a proven carcinogen. The recording of tetrachloroethene at elevated levels in butter in shops near to drycleaners (Miller & Uhler 1988) are an indication of the potential for aerial contamination of all kinds of fatty foods.

**CONCLUSION**

There is a great deal of literature on the occurrence and behaviour of the chlorinated solvents in the environment. However, it is badly skewed, with a preponderance of research in the areas of atmospheric and groundwater contamination and biodegradation. This research indicates very strongly the considerable effects already being experienced as a result of overuse of these compounds, particularly with reference to human health and exposure. It is certain, however that the solvents will have unpredictable adverse effects on sectors of the ecosphere because current emission controls are highly permissive. For example, chlorinated solvents seem to be involved in European forest die-off, though the link is far from clear.

They are clearly a group of chemicals to which the general population may be substantially exposed. The exposure pathway is predominantly through the atmosphere. There may also be significant exposure through drinking water and through food.

The lipophilic nature of these chemicals suggests that they may, in aquatic systems, be involved with the surface microlayer, through transport by and wet scavenging from the major environmental compartment: the atmosphere. Unsurprisingly, perhaps, given the global emission figures, chlorinated solvents interact to contribute to the global phenomena of ozone depletion and greenhouse warming. It is clear that emissions need to be prevented given the atmospheric lifetime of some solvents.

Destruction using current technology is of growing general con-

cern. When incinerated, chlorinated solvents can give rise to an unpredictable array of products of incomplete combustion and secondary combustion products. This subject will be dealt with in a later document. There is no doubt that the chlorinated solvents should be withdrawn from use as a result of their involvement in large scale deleterious environmental changes. Proscription would also treat in a precautionary way the very real concerns posed by uncertain health effects of solvents, their uncertain fate in aquatic systems and interactions with the microlayer.

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