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including tetrachloromethane, in water from well
adjacent to former Union Carbide India Ltd (UCIL)
pesticide plant, Bhopal (India)**

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Contents

Summary	3
Introduction.....	3
Materials and methods	4
Analysis of VOCs at the Greenpeace Research Laboratories.....	4
Analysis of VOCs at the accredited water laboratory.....	6
Results.....	6
Discussion.....	9
References.....	11

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Summary

In previous studies (Labunska *et al.* 1999, Stringer *et al.* 2002) we have demonstrated the scale and extent of environmental contamination on and around the site of the former Union Carbide India Ltd (UCIL) pesticide plant in Bhopal. This contamination includes the presence of toxic chlorinated solvents in groundwater reserves serving wells still in use by local communities, especially those located to the north of the plant boundary.

In this current study, we report the results of analysis of three additional samples of water collected from a well located immediately to the south of the UCIL plant, a well which is also still in regular use for drinking and other domestic uses. Analyses confirmed the presence in water from this well of the same chlorinated solvents reported in our earlier studies, including tetrachloromethane (otherwise known as carbon tetrachloride), chloroform and a range of chlorinated benzenes. Levels of tetrachloromethane, at 2-3 parts per million, were more than 500 times higher than the limits set for drinking water by the World Health Organisation (WHO). Limits for chloroform and 1,2,4-trichlorobenzene were also exceeded, though to a lesser extent. These values are broadly similar to those obtained from the most contaminated well identified to the north of the plant in our 1999 study.

It is known that tetrachloromethane and chloroform, and possibly also chlorinated benzenes, were used extensively on the UCIL site in the manufacture of pesticides, primarily Sevin, during its period of operation up until the 1984 disaster. Taken together, these results indicate that operations at the plant have resulted in substantial contamination of the aquifer underlying the site, and serving a number of community wells, with toxic chlorinated solvents. Their continued presence in groundwater two decades after operations ceased at the plant demonstrates the environmental persistence of these chemicals.

The long-term consequences for health of continuous use of this contaminated water by humans and livestock are not known and have never been subject to specific investigation in communities surrounding the UCIL site. Nevertheless, these latest results, combined with those from previous work, indicate the urgency with which alternative water supplies need to be installed to serve the total needs of the communities. In the longer term, efforts should be made to decontaminate the aquifer with a view ultimately to re-enabling use of the water.

Introduction

In October 2004 Greenpeace Research Laboratories (GRL) received three samples of well water for analysis. The samples had been collected by a third party from a water pump

located to the south of the boundary of the former Union Carbide India Ltd (UCIL) pesticide plant in Bhopal, India. We understand that observations made and photographs taken at the time of collection confirm that local people were using water from this well as a source of drinking water. Although we do not have precise details of the location of the well, we understand that it was not one of those from which our laboratory has previously sampled and analysed water (Labunska *et al.* 1999) and that, at the time of sampling, the well was not marked as being unsuitable for drinking.

Three samples of water, each of approximately 200ml, had been collected sequentially from the same water pump into clean, screw-cap, glass bottles. Care had been taken to ensure that the bottles were completely full, with no air bubbles which could otherwise have reduced concentrations of volatile compounds over time during storage. Samples were kept cold and dark before and during shipment to our laboratory.

On receipt by our laboratory, these samples were assigned the codes NGP04018, NGP04019, and NGP04020. As they were collected sequentially from a single well pump, they could be considered as replicates. Although the samples were colourless and non-turbid, a very small quantity of fine solid particles could be seen at the bottom of each sample. The precise nature of these solids is not known. However, there are no reasons to believe that this fine material did not originate from the water samples themselves.

Materials and methods

One of the samples, NGP04020, was subsequently used by our laboratories for an initial screening analysis using gas chromatography/mass spectrometry (GC/MS) operated in both SCAN (total ion chromatogram) and SIM (Selective Ion Monitoring) modes. SIM mode was used to verify the presence or absence of the full range of volatile organic compounds (VOCs) listed in Table 1, as well as to estimate concentration ranges to assist in subsequent quantification. Details of the analytical methods employed are given below.

Samples NGP04018 and NGP04019 were thereafter divided each into two sub-samples following careful agitation to ensure homogeneity. Sub-samples were collected immediately after opening the sample bottles for the first time in order to minimise losses of volatile components. One set of sub-samples (diluted 1:500 with VOC-free deionised water) were forwarded to an accredited water laboratory for quantitative analysis of a range of halogenated (chlorinated and brominated) VOCs. These substantial dilutions were necessary in order to bring the concentrations of some analytes, present at high concentrations in the original samples down, within ranges which could be quantified. The second set of sub-samples were analysed in our laboratory also for VOCs (in part for interlaboratory calibration purposes) as well as for semi-volatile organic compounds (including chlorinated benzenes). Again, further details are given below.

Analysis of VOCs at the Greenpeace Research Laboratories

For VOC analysis, no further sample preparation was required. For each sub-sample, three portions or aliquots of 10ml each were transferred into pre-cleaned 20ml headspace vials and sealed with Teflon-lined caps. One of these 10ml aliquots for each sub-sample was used undiluted for the general GC/MS screen analysis to identify the full range of volatile

compounds in the sample. The second (NGP04018a and NGP04019a) and the third aliquots (NGP04018b and NGP04019b) were each diluted 1:100 with VOC-free deionised water to allow quantification in SIM mode of some of the more toxicologically significant compounds detected (see Table 2 below), using an external standards.

Name of compound	Target ion	Qualifying ions
Methane, dibromo-	93	95, 174, 176
Methane, bromodichloro-	83	85, 127, 129
Methane, dibromochloro-	129	127, 131, 79
Methane, trichloro-	83	47, 35, 118
Methane, tribromo-	173	175, 171, 254
Methane, tetrachloro-	117	35, 47, 82
Ethane, 1,1-dichloro-	63	27, 83, 98
Ethane, 1,2-dichloro-	62	27, 64, 49
Ethane, 1-bromo-2-chloro-	63	65, 27, 26
Ethane, 1,1,1-trichloro-	97	61, 26, 117
Ethane, 1,1,1,2-tetrachloro-	131	133, 117, 119
Ethane, 1,1,2,2-tetrachloro-	83	85, 95, 131
Ethane, pentachloro-	167	165, 169, 117
Ethane, hexachloro-	117	201, 203, 199
Ethene, chloro-	27	62, 37, 47
Ethene, 1,1-dichloro-	61	96, 26, 35
Ethene, 1,2-dichloro-, cis-	61	96, 26, 35
Ethene, 1,2-dichloro-, trans-	61	96, 26, 37
Ethene, trichloro-	95	130, 132, 97
Ethene, tetrachloro-	166	129, 94, 47
Butane, 1,3-dichloro-	55	54, 90, 63
Butane, 1,4-dichloro-	55	54, 90, 64
2-Butene, 1,4-dichloro-	53	75, 88, 27
1,3-Butadiene, 2-chloro-	53	50, 51, 52
1,3-Butadiene, hexachloro-	225	260, 190, 118
Propane, 2-bromo-1-chloro-	41	39, 77, 79
Chlorobenzene	112	77, 51, 38
Toluene	91	39, 65, 51
1,2-Dichlorobenzene	146	148, 150
1,3-Dichlorobenzene	146	148, 150
1,4-Dichlorobenzene	146	148, 150
1,2,3-Trichlorobenzene	182	180, 184
1,2,4-Trichlorobenzene	182	180, 184
1,3,5-Trichlorobenzene	182	180, 184
1,2,3,4-Tetrachlorobenzene	216	214, 218
1,2,3,5-Tetrachlorobenzene	216	214, 218
1,2,4,5-Tetrachlorobenzene	216	214, 218
Pentachlorobenzene	250	252, 248
Hexachlorobenzene	284	286, 282

Table 1. List of volatile organic compounds and appropriate ions that were monitored during GC/MS analysis using SIM method.

Name of compound	Target ion	Qualifying ions
Methane, tetrachloro-	117	119, 121
1,2-Dichlorobenzene	146	148, 150
1,3-Dichlorobenzene	146	148, 150
1,4-Dichlorobenzene	146	148, 150
1,2,3-Trichlorobenzene	182	180, 184
1,2,4-Trichlorobenzene	182	180, 184

Table. 2. List of volatile organic compounds that were quantitatively identified during GC/MS analysis using SIM method.

All standards were obtained from Sigma-Aldrich Co. Ltd./Supelco UK. Working standard solutions were initially prepared in methanol with final dilution into 10ml of VOC-free deionised water for consistency with the samples. Calibration curves were prepared throughout the linear response range and were routinely checked during analysis of samples using additional standard solutions of known concentration prepared separately.

Following separation using a Hewlett Packard 5890 Series II gas chromatograph, linked to an Agilent 7694 Headspace Sampler, VOCs were identified and quantified using a Hewlett Packard 5972 Mass Selective Detector operated in SCAN or SIM mode. Compounds were identified through a combination of computer matching against the Hewlett Packard Wiley275 spectral library and expert interpretation.

Analysis of VOCs at the accredited water laboratory

The methodology used in this case was a standard accredited method for the determination of trihalomethanes and selected volatile solvents in raw and potable waters and leachates by gas chromatography/mass spectrometry. Aside from differences in the compounds specifically quantified, this method is almost identical to that used in our laboratory as described above. Further details, including limits of detection for all analytes included, can be provided on request.

Results

The total ion GC/MS screen analysis confirmed the presence in the samples of the following organic contaminants; tetrachloromethane, 3-chlorotoluene, 1,2-dichlorobenzene, 1,3-dichlorobenzene, 1,4-dichlorobenzene, phenol and 1,2,4-trichlorobenzene. A typical chromatogram for these samples is shown in Figure1 below. The largest peak, labelled "1", corresponds to tetrachloromethane, otherwise known as carbon tetrachloride

GC/MS analysis in more sensitive SIM mode revealed the presence of additional compounds in the samples, including trichloromethane (or chloroform) and 1,2,3-trichlorobenzene. Figure 2 shows a typical chromatogram obtained in SIM mode, showing peaks corresponding to various chlorinated benzenes.

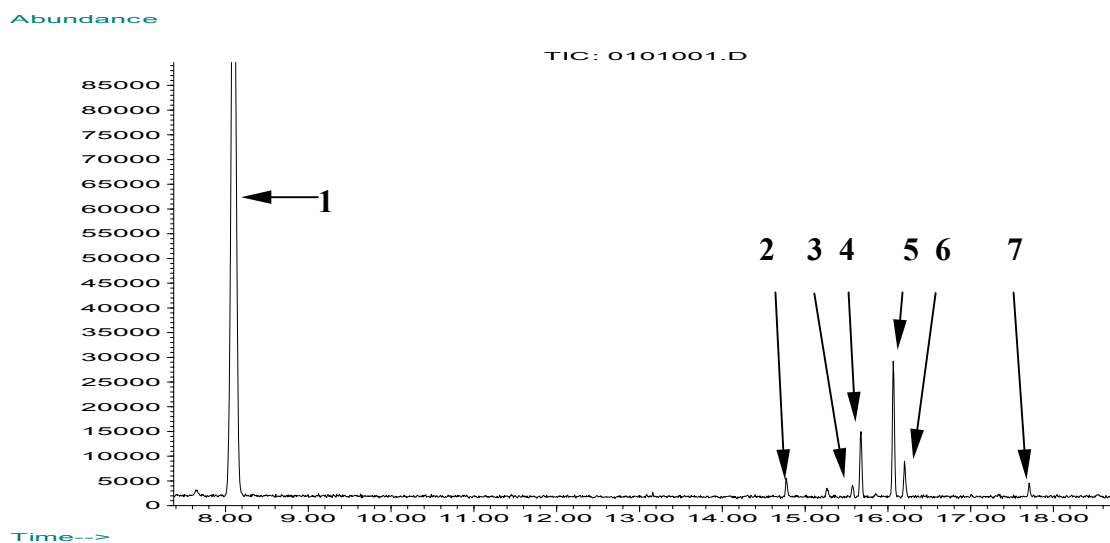


Fig. 1. Total Ion Chromatogram of drinking water sample NGP04020 collected near to former UCIL plant, Bhopal, India, October 2004. 1- Methane, tetrachloro-; 2 - Toluene, 3-chloro-; 3 - Benzene, 1,3-dichloro-; 4 - Benzene, 1,4-dichloro-; 5 - Benzene, 1,2-dichloro-; 6 - Phenol; 7 - Benzene, 1,2,4-trichloro-.

The results of the quantitative analyses conducted both by our laboratory and by the accredited water laboratory are presented in Table 3. Concentrations reported have been corrected for the dilution of the samples. Limits of detection are those for the method (*i.e.* are not adjusted for dilution of the samples).

Note that, of the various analytes present in the samples, our method yielded quantitative data for tetrachloromethane and four chlorinated benzenes only. Although we were able to detect the presence of chloroform, we were not able to determine absolute concentrations. In contrast, the method applied by the accredited laboratory quantified tetrachloromethane and chloroform only; as chlorinated benzenes are not commonly encountered in drinking water, they are not routinely quantified in standard drinking water analyses of this type.

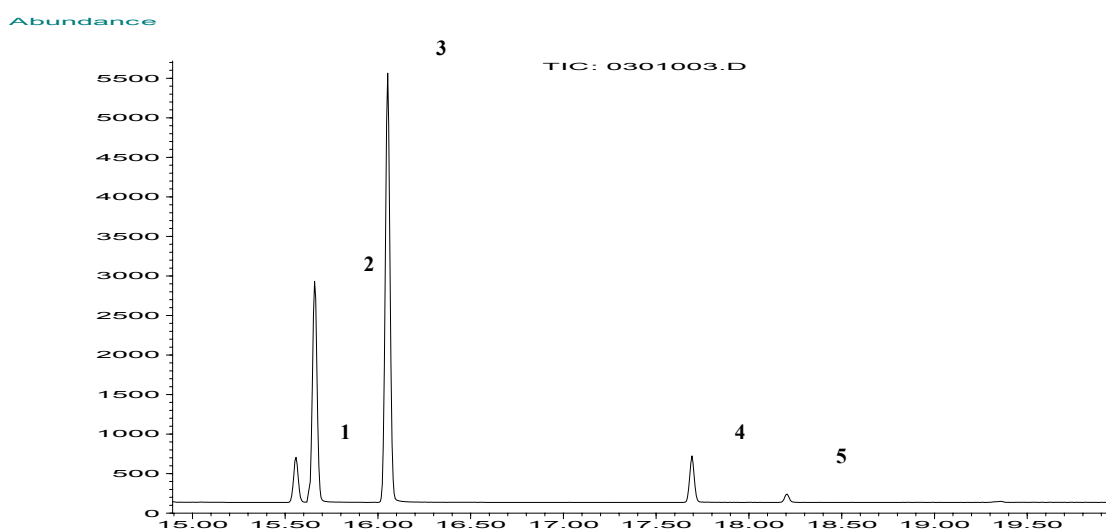


Fig. 2. SIM chromatogram of drinking water sample NGP04020 collected near to former UCIL plant, Bhopal, India, October 2004. 1- Benzene, 1,3-dichloro-; 2 - Benzene, 1,4-dichloro-; 3 - Benzene, 1,2-dichloro-; 4 - Benzene, 1,2,4-trichloro-; 5 - Benzene, 1,2,3-trichloro-.

Laboratory	Code	CCl ₄	CHCl ₃	1,2-DCB	1,3-DCB	1,4-DCB	1,2,4-TCB
		Concentration, ug/l					
Greenpeace Research Laboratories	NGP04018a	3190	n/a	595	67	216	64
	NGP04018b	2980	n/a	522	49	159	56
	NGP04019a	3080	n/a	289	43	97	54
	NGP04019b	3260	n/a	331	59	101	72
	Detection limit	1	n/a	0.4	0.4	0.4	1
Accredited water laboratory	NGP04018	2300	750	n/a	n/a	n/a	n/a
	NGP04019	2200	800	n/a	n/a	n/a	n/a
	Detection limit	0.04	0.04	n/a	n/a	n/a	n/a

Table 3. Concentrations of tetrachloromethane (CCl₄); chloroform (CHCl₃); 1,2-dichlorobenzene (1,2-DCB); 1,3-dichlorobenzene (1,3-DCB); 1,4-dichlorobenzene (1,4-DCB) and 1,2,4-trichlorobenzene (1,2,4-TCB) in drinking water samples collected near former UCIL, Bhopal, India, October 2004. n/a – not analysed

Concentrations of tetrachloromethane determined in our laboratory, and of both tetrachloromethane and chloroform determined by the accredited laboratory, showed close *within*-laboratory agreement between duplicate measurements and/or samples. Comparison *between* laboratories is only possible for one analyte, namely tetrachloromethane. In this case, the levels reported by the accredited laboratory were approximately 30% lower than values determined in our laboratory. Although this is not a negligible difference, it could possibly have arisen in part from the process of preparing high factor dilutions in order to facilitate quantitative analysis (*i.e.* 100 x dilution in the case of our laboratory and 500 x dilution in the case of the accredited laboratory). Any slight heterogeneity of contaminant distributions in the original samples could well have been compounded in these dilution procedures and contributed to this difference.

In any case, despite this 30% difference, the concentrations recorded for tetrachloromethane by both laboratories (approximately 2000-3000 ug/l, or 2-3 parts per million [ppm]) are remarkably high for groundwater or, indeed, for drinking water from any source. At 750-800 ug/l (0.75-0.8 ppm), levels of chloroform in the samples (accredited laboratory only) are also indicative of substantial contamination of the aquifer. These findings are discussed further below.

Concentrations of chlorinated benzenes (our laboratory only) are also significant. Concentrations varied from around 50 ug/l for 1,3-dichlorobenzene and 1,2,4-trichlorobenzene to between 100 and 200 ug/l for 1,4-dichlorobenzene and up to 300-500 ug/l (0.3-0.5 ppm) for 1,2-dichlorobenzene. For organic compounds with a relatively low solubility in water, these concentrations are high. This maybe because these contaminants are associated in part with the fine particulate matter in the samples (see below), although the presence of high concentrations of chlorinated solvents such as chloroform and tetrachloromethane may also have augmented the solubility of the chlorinated benzenes.

Concentrations of individual chlorinated benzenes showed greater disagreement between duplicates than those for the more volatile compounds discussed above. Again this may result in part from the presence in the water samples of a small quantity of fine solid particulate matter with which semi-volatile chemicals such as chlorinated benzenes may be

expected to associate. For example, dichlorobenzenes are expected to sorb moderately to soils and sediments, based on measured soil organic carbon partition coefficient (Koc) values. Sorption is primarily to the soil organic phase and, therefore, depends on the organic content of the soil (Rao and Asolekar 2001). However, sorption is likely to be reversible; therefore, dichlorobenzenes may leach from hazardous waste sites and be transported to the groundwater (ATSDR 2000). In order to investigate this possibility, the samples NGP04018 and NGP04019 were filtered through the pre-cleaned paper filter, diluted 1:50 with VOC-free deionised water and analysed for chlorinated benzenes in duplicate using the same GC/MS method in SIM mode. The results of these additional analyses are presented in Table 4.

Clearly, filtration both reduced the overall concentrations of chlorinated benzenes in the samples, suggesting a significant association with particulate matter, and resulted in closer agreement between duplicates. Filtration led to greatest reductions in concentration for 1,4-dichlorobenzene, possibly as this compound has a relatively higher melting point than the other dichlorobenzenes and is a solid at room temperature.

Discussion

In a previous study we have demonstrated the presence of high concentrations of chlorinated solvents, in particular tetrachloromethane (or carbon tetrachloride), in water abstracted from wells serving communities located around the site of the former Union Carbide India Ltd (UCIL) pesticide plant in Bhopal (Labunska *et al.* 1999). In that study, tetrachloromethane and chlorinated benzenes were detected in groundwater collected from three wells located close to the northern boundary of the plant, with concentrations of tetrachloromethane ranging from 200 to 3410 ug/l (0.2 to 3.4 ppm). High concentrations of chloroform were also present in those samples (160 to 2590 ug/l, or 0.16 to 2.6 ppm). Although both these compounds were also detected in one of the sampled wells located to the south of the plant in 1999, levels there, especially for tetrachloromethane (50 ug/l, or 0.05 ppm) were considerably lower. Other wells sampled at that time which were located to the south of the site did not yield detectable levels of any volatile organic compounds.

Although we do not have a precise location for the well sampled in this current study, we understand that it is located close to the southern boundary of the UCIL site and that it is not one of the wells that we have previously sampled.

Sample code	1,2-DCB		1,3-DCB		1,4-DCB		1,2,4-TCB	
	Concentration, ug/l							
	Before filtration	After filtration	Before filtration	After filtration	Before filtration	After filtration	Before filtration	After filtration
NGP04018a	595	96	67	46	216	26	64	19
NGP04018b	522	97	49	45	159	28	56	17
NGP04019a	289	22	43	29	97	n/d	54	11
NGP04019b	331	23	59	25	101	n/d	72	12
Detection limit	0.4		0.4		0.4		1	

Table 4. Concentrations of 1,2-dichlorobenzene (1,2-DCB); 1,3-dichlorobenzene (1,3-DCB); 1,4-dichlorobenzene (1,4-DCB) and 1,2,4-trichlorobenzene (1,2,4-TCB) in drinking water samples before and after filtration. Samples collected near former UCIL, Bhopal, India, October 2004. n/d – not detected

Unfortunately, therefore, we cannot make a direct comparison with results previously recorded in order to determine whether the presence of high levels of chlorinated VOCs in these samples indicates further southward spread of contamination in the aquifer compared to the situation in 1999.

Nevertheless, the high levels recorded are of concern in themselves, particularly as, according to information provided to us, this well was not labelled in any way to indicate contamination and was still in use by the local community as a source of water for drinking and other uses. The concentrations of tetrachloromethane, chloroform and chlorinated benzenes in water from this southern well are only slightly lower than those recorded previously in the most contaminated well supply (sample code IT9030) identified to the north of the plant (Labunska *et al.* 1999).

It is known that carbon tetrachloride and chloroform were used extensively as solvents in the pesticide manufacturing processes used at the UCIL plant during its operation up to the disaster in 1984 (Behl *et al.* 1978, NEERI 1990). Carbon tetrachloride was the primary solvent used in production of the pesticide Sevin (also known as carbaryl), as well as in the manufacture of raw materials including methyl isocyanate (MIC). Chloroform was also used in the conversion of phosgene to MIC (Behl *et al.* 1978, UCC 1985). Some documentation suggests that dichlorobenzenes were also used on site as solvents, in this case in the production of naphthol which was then reacted with MIC to produce the final product Sevin, though their presence may also arise from historic operations at the site before pesticide manufacture began. Given their extensive use on site, the propensity for these volatile and semi-volatile chemicals to migrate through soils into groundwater (*e.g.* Howard 1989) and the fact that the aquifer serving the wells sampled lies in part directly beneath the site, it seems highly likely that the occurrence of these contaminants in the groundwater has resulted directly from historic use, release and disposal of these solvents on and/or around the UCIL site.

The high concentrations of chlorinated solvents found in drinking waters collected from around the site are of particular concern given the known toxicological properties of many of these chemicals.

For example, tetrachloromethane (carbon tetrachloride) is a substance which can cause cancer in animals and humans (USEPA 1997) and has been classified as Group 2B carcinogen (possibly carcinogenic to humans) by the International Agency for Research on Cancer (IARC 1999). Carbon tetrachloride induces proliferation of liver cells and DNA synthesis. It also has a mutagenic effect (IARC 1999). High exposure to carbon tetrachloride can cause liver, kidney, and central nervous system damage.

Chloroform is also listed as a Group 2B carcinogen (IARC 1998), with the main target organs again being the liver and kidney but with concerns also for the intestine (Dunnick & Melnik 1993; Snyder & Andrews 1996; Chiu *et al.* 1996). The liver and kidney are also the primary target organs for toxic effects of the di- and trichlorinated benzenes (Meek *et al.* 1994b).

Furthermore, all these chlorinated solvents have a high persistency (*i.e.* low tendency to degrade) especially when released to groundwater. Moreover, because chlorinated solvents have a density greater than water (CRC 1969), groundwater plumes of these contaminants may form pools of residual solvent below the water table (Rivett *et al.* 1994). Chlorinated

solvents may undergo slow reductive dechlorination under anaerobic conditions, though it has been reported that final transformation into methane and ethane is significantly retarded if several of these compounds are present together (Hughes & Parkin 1996a, Hughes & Parkin 1996b). For these reasons, once contamination has occurred, without intervention it can be expected to persist for years or even decades, with concentrations attenuating only through gradual spread of the contamination through the aquifer.

Because of the toxicological concerns, maximum limits have been set by the World Health Organisation (WHO) for concentrations of certain VOCs, including tetrachloromethane (carbon tetrachloride), chloroform and certain chlorinated benzenes, in drinking water. For carbon tetrachloride (tetrachloromethane) the original limit of 2 ug/l (WHO 1993) has recently been increased to 4 ug/l (or 0.004 ppm). Nevertheless, the levels recorded for the well waters analysed in this current study (approximately 2-3 ppm) are clearly substantially (more than 500 times) higher than this WHO limit. The levels also exceed the level set nationally for the USA (5 ug/l, USEPA 1999) by a similar degree.

The WHO and US EPA have also established limits for chloroform in drinking water, at 200 ug/l and 100 ug/l respectively. Concentrations found in the waters sampled in this study again exceed these limits, albeit to a lesser degree than for tetrachloromethane. WHO limits for 1,2,4-trichlorobenzene (20 ug/l) were also exceeded, while levels of 1,2-dichlorobenzene and 1,4 dichlorobenzene fell only slightly below WHO limits of 1000 ug/l and 300 ug/l respectively.

In summary, the presence of chlorinated solvents such as tetrachloromethane in the well waters sampled close to the former UCIL pesticide plant, both in this and in previous studies, is undoubtedly due to the long-term industrial contamination of the surrounding environment from this plant. Consumption of water, contaminated by the chemicals identified above, for long periods could cause significant health damage. Measures are therefore urgently required in the short term 1) to seal off the wells in the vicinity of the plant to prevent further use and 2) to provide simultaneously a sufficient, continuous and reliable source of clean drinking water to serve the total needs of the local communities. In the longer term, it may be possible to emplace measures capable of separating the contaminants from the waters in the aquifer such that further spread of contamination may be prevented and that the aquifer may again serve as a safe supply of water for use by humans and livestock.

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