

THE LEACHING OF SOME HALOGENATED ORGANIC COMPOUNDS FROM DOMESTIC WASTE

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Summary

Columns of domestic waste contaminated with halogenated organic compounds to the extent of 500 to 5,000 ppm w/w have been eluted with water to assess the extent to which these compounds may be leached from wastes in landfill sites. The concentrations of the solvents perchloroethylene, trichloroethylene and 1,1,2-trichlorotrifluoroethane in leachates were at least two orders of magnitude lower than in the solid waste being leached. It would seem, therefore, that evaporation and adsorption will serve to reduce the leachate concentrations of these compounds, which arise in domestic waste landfill as dry cleaning residues, to the part per million level or below. The low solubility of compounds such as dichlorobenzene, dichloronitrobenzene and chlorohydroxyquinoline appears to have prevented their appearance in the column leachates at concentrations above a few parts per million. In contrast, more soluble compounds such as dichlorophenol dichloroaniline, dibromoethane and dichloropropane were detected in leachates at concentrations of tens of parts per million. Adsorption processes appeared to have some capacity for reducing the concentrations of halogenated aromatic compounds in contaminated leachates.

Certain halogenated organic compounds present in or deposited with domestic waste could dissolve in leachates and subsequently pass into ground-water or surface water as pollutants. This paper describes an experimental study of the leaching of some compounds of this type from domestic waste. Apart from plastics the major halogenated organic contaminants of domestic wastes are solvents such as tetrachloroethylene and 1,1,2-trichlorofluoroethane arising as filtration or distillation residues from 'coin-op' dry-cleaning establishments [1]. Trichloroethylene and 1,1,1-trichloroethane from metal cleaning operations may also be present. Other compounds normally deposited only at industrial landfill sites [2] may prove acceptable for landfill with domestic wastes provided the quantities involved are shown to present no environmental hazard.

McConnell et al. have described [3] the environmental occurrence and degradation of several volatile [4] halogenated organic compounds containing one or two carbon atoms. Although widely distributed these compounds

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appear to degrade rapidly to a 'background' level of parts per billion. They also evaporate rapidly from aqueous solution, a process studied in more detail by Dilling et al. who found [5] 'half-lives' of less than one hour for many C₁ and C₂ halo-organics present in water at the 1 part per million level. This latter observation means that samples of leachate to be analysed for halogenated solvents must be protected from contact with the atmosphere, as well as from organic materials such as plastics which adsorb halosolvents. The environmental persistence of halogenated aromatic compounds other than polychlorinated biphenyls and pesticides has not been investigated in detail. However, halogenated aromatics are likely to pose greater problems than halogenated solvents owing to their lower volatility and relatively slow biodegradation rates.

A total of ten compounds was selected for investigation in the leaching tests described here, four were aliphatic and the remainder aromatic in nature. In addition to the commonly used solvents Perklone and Triklone, dibromoethane and dichloropropane were selected as examples of compounds containing two and three carbon atoms. The aromatic compounds used included two halogenated benzene derivatives and dichloronitrobenzene. Dichlorophenol and dichloroaniline were used to provide examples of aromatic compounds containing the acidic hydroxyl and basic amine substituents, while chlorohydroxyquinoline provided an example of a poly-aromatic molecule containing two fused rings. The aqueous solubilities of these compounds were determined and the results are presented in Table 1, along with the reported [3] solubilities of some halogenated solvents, their boiling points [4] and partition coefficients between water and air [3]. The observed solubilities ranged from 14 ppm for dichloronitrobenzene to 8,200 ppm for chloroform. Dichlorophenol was the most soluble aromatic compound studied.

The leaching of the two common solvents, Perklone and Triklone, was investigated using stainless steel columns 56 cm in diameter and ca. 90 cm in depth containing ca 50 kg of fresh, untreated domestic waste obtained from the Witney landfill site at Witney, Oxon. Perklone, in the form of filtration residues from dry-cleaning processes and Triklone, in the form of a 50/50 v/v mixture of trichloroethylene and used engine oil, were mixed with the refuse to give halogenated solvent loads of 200 and 500 ppm w/w. Three types of column condition were used for each concentration. One column was left open to the atmosphere to allow evaporation to occur, a second was closed off to restrict evaporation and a third was fully saturated with water. Samples of leachate were taken from the bottom of each column in a closed system to prevent evaporation of the solvent occurring after sampling. The columns were allowed to stand out of doors for approximately one month and were then eluted with successive 25 litre aliquots of water; in the case of the saturated columns leachate was removed to produce space for the added water. After 2 to 3 months all the leachate was drained from the unsaturated columns with the 500 ppm solvent loading and petroleum

TABLE 1

Properties of some halogenated organic compounds

Compound	Aqueous solubility (w/w, ppm)	Temperature (°C)	Boiling point at 1 atm (°C)	Partition coefficient water-air (w/v/w/v) 20°C
Chloroform [3]	8,200	20	61	8.6
Carbon tetrachloride [3]	785	20	77	1.1
Trichloroethylene [3] (Triklone)*	1,100	20	87	2.74
Tetrachloroethylene [3] (Perklone)*	150	20	121	1.22
1,1,1-Trichloroethylene [3] (Genklone)*	480	20	74	0.71
1,1,2-Trichlorotrifluoroethane (Arklone)*	170	20	48	—
1,2-Dibromoethane**	2,910	25	131 [4]	—
1,2-Dichloropropane**	2,096	25	94 [4]	—
1,4-Dichlorobenzene**	34	25	174 [4]	—
2,5-Dichloronitrobenzene**	14	25	267 [4]	—
2,4-Dichloroaniline**	166	25	245 [4]	—
2,4-Dichlorophenol**	2,650	25	260 [4]	—
5,8-Chlorohydroxyquinoline**	19	25	—	—
Fluorobenzene**	922	25	85 [4]	—

*ICI Trade name, other common Trade names are quoted in reference 1.

**Analyses were carried out by Detection and Analysis Group, Instrumentation and Applied Physics Division, Harwell Laboratory using gas chromatographic methods.

spirit passed through the column in an attempt to strip any adsorbed solvent from the refuse.

None of the aqueous leachates from any of the columns containing Perklone exhibited tetrachloroethylene concentrations above the detection limit of 2 ppm w/v. The petroleum extract of the closed column contained ca. 50 ppm of tetrachloroethylene and elution of this column with 68 litres of petroleum spirit removed approximately 15% of the tetrachloroethylene originally present. The petroleum extract from the open column did not contain detectable concentrations of tetrachloroethylene.

Only two of the columns containing Triklone produced leachates containing detectable (i.e. > 2 ppm w/v) concentrations of trichloroethylene. These were the closed and the saturated columns which had a 500 ppm loading of Triklone. Leachate from the closed column contained from 4 to 7 ppm of trichloroethylene over a four month elution period, while 9 to 13 ppm was detected in leachates from the saturated column over a similar time period. The petroleum extract of the open column containing a 500

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TABLE 2

Leaching test results for halogenated organics in domestic refuse

† Indication that the concentration was below the detection limit figure specified

Compound	Concentrations of halogenated compounds in leachate (w/v, ppm)*							
	Fresh refuse (1,000 ppm)**	Percent removal***	Mature refuse (1,000 ppm)**	Percent removal***	Fresh refuse (5,000 ppm)†	Fresh refuse 2nd stage †	Mature refuse 3rd stage †	
1,2-Dibromoethane	Maximum	<13	14	<6	54	22	<5	
	Minimum		<10		<5	<5	<5	
1,2-Dichloropropane	Maximum	<12	85		50	14	<3	
	Minimum		<3	<5	<3	<3	<3	
1,4-Dichlorobenzene	Maximum	<3	<5	<3	22	1	1	
	Minimum		<3		<1	<1	<1	
1,5-Dichloronitrobenzene	Maximum	<3	<3	<4	—	—	—	
	Minimum		<3		—	—	—	
1,4-Dichloroaniline	Maximum	<9	48	<10	25	<10	<10	
	Minimum		<15		<10	<10	<10	

4-Dichlorophenol	Maximum	50	8	75	<10	240	39	13
	Minimum	15		<15		37	13	<10
8-Chlorohydroxy-quinoline	Maximum	13	<11	5	<12	<5	<5	<5
	Minimum	<2		<2		<5	<5	<5
fluorobenzene	Maximum	19	<5	<5	<1	—	—	—
	Minimum	<1		<1		—	—	—

Analyses for halogenated organic compounds in leachate were carried out by Detection and Analysis Group using gas chromatography with a microwave plasma detector tuned for a halogen, usually chlorine. This method has the disadvantage of low sensitivity (1 to 25 ppm detection limit) but provides a specific method for determining halogenated compounds in the presence of the many other organic contaminants found in refuse leachates.

*Columns containing 1 kg of fresh or mature pulverised domestic refuse with 1 g of halogenated compound and 31 of water added, were allowed to equilibrate for 21 weeks. Samples of leachate were taken for analysis after 1 hour, 1, 3 and 21 weeks.

**After being allowed to equilibrate for 21 weeks the leachate was drained from the columns and replaced with an equal volume of fresh water. This process was repeated after 9 days and the percentage of halogenated organic removed was calculated on the basis of the volumes of leachate removed and the concentrations of halogenated organic compound therein. In most cases an upper limit on percent removal is set by the detection limits of the analytical technique used.

Data from experiments in which 5 g of halogenated organic and 1 kg of fresh pulverised domestic refuse were equilibrated with 31 of water for weeks. Leachate from these columns was then run into a second stage column containing 1 kg of fresh pulverised refuse and from there into a third stage column containing 1.5 kg of mature pulverised domestic refuse. The whole system was then eluted with 1.3 l of water (5 cm depth in the columns) per week for a further 15 weeks. Samples of leachate were taken from the base of each of the three columns for analyses at approximately 2 week intervals.

ppm load of Triklone did not contain detectable concentrations of trichloroethylene.

In domestic wastes the likely overall, combined concentration of Perklone and Arklone arising from coin-op dry-cleaners is of the order of 100 ppm [1]. Based on the experimental results, and noting that Arklone and Perklone are of similar aqueous solubility, it seems that evaporation and adsorption processes will combine to maintain the residual concentrations of these solvents in domestic landfill leachate below the part per mil. level. At such concentrations their presence in leachate constituents no significant threat to groundwater quality. In the case of Triklone the situation is a little less favourable owing to its higher aqueous solubility. The experimental results suggest that evaporation and adsorption may result in leachate concentrations of trichloroethylene which are about two orders of magnitude lower than its concentration in the domestic waste being leached. This figure is in accord with data from field studies [6]. Samples of liquid wastes and groundwater from one site showed concentrations of 1,000 to 5,000 ppm of 1,1,1-trichloroethane and tetrachloroethylene in oily wastes being deposited. The concentrations of these solvents in aqueous liquors contacting the oil were 50 to 100 ppm and they could not be detected above a limit of 1.5 ppm in groundwater from strata underlying the site. Thus there appears to be some scope for the controlled bulk disposal in domestic landfill of oily halogenated solvent containing wastes from metal cleaning.

Leaching tests on the other compounds studied were carried out in the laboratory on a smaller scale. Kilogram quantities of fresh, or 5 year old matured, pulverised refuse obtained from a site at Thriplow, Cambridgeshire were used. The refuse columns were prepared in 5 litre glass aspirators fitted with a fermentation lock (bubbler) which allowed gases to leave the column but prevented free ingress of air. Leachate could be drained from the columns through a stopcock at the base and 1,000 and 5,000 ppm loadings of halogenated organic on the refuse were used. Both saturated and elution conditions were established in the columns and the results of the leaching tests along with details of the column conditions are summarised in Table 2. The halogenated organic compound concentrations in the leachates proved to be highly variable so only the maximum and minimum concentrations found in the leachates are presented. Generally the maximum concentrations were observed after 3 weeks under saturated conditions. The variability of the results and the low solubilities of the compounds involved preclude any estimates of partition coefficients between refuse and leachate, as have been made for phenolic compounds [7].

The results of the tests in which a loading of 1,000 ppm of halogenated organic was allowed to equilibrate with saturated refuse broadly reflect the differing solubilities of the compounds studied. Mature refuse, which contains a higher proportion of un-degraded plastics, appeared to be a better adsorbant for dibromoethane and dichloropropane than fresh refuse. In contrast, fresh refuse, in which the cellulosic materials were not substantial-

ly degraded, proved a better adsorbant for dichlorophenol and dichloroaniline. At the 1,000 ppm loading dichlorobenzene and dichloronitrobenzene could not be detected in any of the column leachates. Elution of the columns with a total of 2 bed volumes of water failed to remove more than 15% of the halogenated organics studied and in the cases of dichlorobenzene and dichloronitrobenzene this figure was less than 5%.

In the tests using a 5,000 ppm halogenated organic load on refuse only dichlorobenzene and dichlorophenol reached substantially higher concentrations in leachate than in the 1,000 ppm loading tests. In terms of the peak concentrations in leachate, passage through a second column of uncontaminated fresh refuse produced an attenuation factor of at least 2 and, in one case of 22, in the halogenated organic concentration. Subsequent passage of the leachate through uncontaminated, mature refuse produced a further reduction in concentration by a factor ranging from 1 to at least 4.

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