

**ORGANOHALOGENS: THE CASE FOR DEVELOPING
REGULATION BY CHEMICAL CLASS**

A Submission to the Workshop on Products Containing
or Generating Organohalogen Compounds

15-16 May 1995, Amsterdam

Paul Johnston, Ruth Stringer & David Santillo,
Greenpeace Research Laboratories,
University of Exeter,
United Kingdom

1. INTRODUCTION

Sewage discharges account for a significant proportion of discharges to the aquatic environment. It has been estimated for the UK alone (Lester 1990) that, exclusive of cooling water, 23 million cubic metres of water are used daily. 8 million of these are accounted for by domestic use and together with 6.8 million cubic metres from industry, this water is discharged to sewer. Overall, it has been estimated (McIntyre 1990) that 180 litres of sewage are produced *per capita* each day. On a global basis, Nriagu and Pacyna (1988) estimated that domestic wastewaters amounted to around 150×10^9 cubic metres per annum.

The potential environmental effects of discharges of sewage to aquatic systems are now well understood. These include health risks due to the presence of pathogenic organisms, oxygen depletion of receiving waters, eutrophication and modification of sediment quality and character (see: Clark 1992; NSTF 1993). Sewage also contains chemical contaminants which are then released into the environment. Regulation of sewage discharges and, in particular, their chemical content poses considerable problems as pointed out by Johnston *et al.* (1993).

Sewage discharges input substantial quantities of toxic trace metals into aquatic systems as estimated by Nriagu and Pacyna (1988). Coastal sewage discharges were estimated to contribute around 14,000 tonnes of oil to the North Sea, around 5% of the total (McIntyre 1988). The detergent content of domestic wastewater has been estimated at between 3 and 8% with an average content of 3-4g/kg in dried sewage sludges (Kuhnt 1993). Sewage treatment plants have also been found to be a significant source of organic micropollutants into the aquatic environment and the atmosphere (Clark *et al.* 1991; Mayer *et al.* 1994).

The organohalogen group of compounds is one which is receiving increasing international scrutiny. The International Joint Commission, a body which advises the Canadian and United States Government on water quality in the Great Lakes (IJC 1992; 1994), has concluded that in order to address the problems of organohalogens the use of chlorine and its compounds should be avoided in manufacturing processes. Similarly, the OSPAR Commission has prioritised organohalogen compounds which are toxic persistent and liable to bioaccumulate for reduction and elimination. A further step was taken recently, at the June 1995 fourth Ministerial North Sea Conference with the commitment to continuously work towards the cessation of all discharges, emissions and losses of hazardous substances within 25 years (Para 17, Ministerial Declaration).

The most important organohalogens are those containing chlorine which account for around 95% of organohalogen compounds (see: OSPAR 1994). Chlorine manufacture is currently around 40 million tonnes per annum of which around 30% is used to manufacture 1,2-dichloroethane. 15% is used in the production of other plastics and 15% in the production of diverse organic chemicals. The balance is used for industrial bleaching, manufacture of

chlorinated solvents, titanium dioxide and bromine. Around 1% is used for drinking water disinfection (Nisbet 1989; Verbanic 1990). On the basis of what is known about the quantities of natural organohalogens produced (see: Stringer & Johnston 1995) it can be estimated that industrial organohalogen production exceeds natural production by up to a factor of seven.

The organohalogens predominate in lists drawn up for regulatory purposes. While they account for around 10% of chemical production, they comprise around 70% of the chemicals identified as of potential environmental concern as exemplified by Annex 1D of the 1990 Third Ministerial Declaration on the Protection of the North Sea (MINDEC 1990). This weight of regulation has resulted from the identification of a variety of actual and possible human and environmental health effects (see: Johnston & Stringer 1994) due to organohalogens and in particular organochlorines. This paper outlines some of the difficulties likely to be encountered in regulating organohalogens in sewage. It argues that the most successful regulation is likely to be based upon a chemical class approach, rather than upon the current method of "substance by substance" evaluation.

1. SOURCES OF ORGANOHALOGENS IN SEWAGE EFFLUENTS

Measurements of the group parameters AOX and EOX have shown that in the Netherlands alone, emissions of organohalogens from municipal sewage treatment works (MSTWs) alone are 65 and 20 tonnes respectively. This compares with figures of 75 tonnes and 50 tonnes from industrial installations (OSPAR 1994). Of the MSTW emissions, only 15 tonnes has been accounted for, mainly due to use of hypochlorite bleaches. Organohalogens are likely to enter sewerage systems from two main sources: Uses of domestic products and effluents from industry tie-ins to the sewer system. Few attempts have been made to characterise the organohalogens present in domestic products. Hathaway (1980) listed common products containing a variety of toxic chemicals including organohalogens, but the current significance of this is unclear given possible product and commercial changes since this time.

Nonetheless, it is clear that organohalogens have substantial applications in domestic products, ranging from fire retardants, cleansers and toothpaste to non-ethical pharmaceutical preparations. (see: Document ORGHAL 95/IIIa/Add.2). Some of these such as the flame retardants, chlorinated paraffins and chlorobenzenes are clearly toxic, persistent and bioaccumulative. In other cases, such as the Triclosan used in antiseptic hand cleansers or bromocyclene fragrance, their properties have not been fully evaluated. Pharmaceutical products also enter sewerage systems both after passage through the body and through disposal of unused preparations (see: Richardson & Bowron 1985). Many of these are halogenated. Organohalogens are also present in sewage sludges, to which many organic micropollutants may become bound (Rogers 1987a; Rogers *et al.* 1989). Dichlorobenzenes may be found at concentrations of up to 50ppm, while PCBs and chlorinated pesticides can be found at concentrations of 1-10ppm dry weight in sewage sludge.

In many cases, it appears that householders may not be aware of the hazardous properties of materials, prompting disposal in inappropriate ways including to sewer. Table 1, taken from Bass *et al.* 1990, reports the distribution of hazardous items present in a sample of 21 households in the US. The majority (65%) were liquids amounting to 482 US gallons overall. 53% of all products were between one and five years old. This is clearly an area requiring further scrutiny on two levels. Firstly, the composition of these products needs to be determined and the organohalogen component characterised. Secondly, there is a need for a similar survey in Europe to establish the age and nature of the chemicals stockpiled by householders since these may be disposed of to sewer particularly if in liquid form. One particular difficulty is that primary manufacturers have little control over the end use to which their products may be put. Hence, a full characterisation of organohalogens present in household products is likely to prove elusive.

ITEM	NUMBER	PERCENT
Cleaners	117	10.5
Polishes and Waxes	85	7.6
Disinfectants	58	5.2
Solvents	44	3.9
Paint Products	235	21.1
Ignitable Products	44	3.9
Car Products	111	10.0
Insecticides	155	13.9
Herbicides	52	4.7
Fungicides	13	1.2
Rodenticides	4	0.4
Outdated Medicines	10	0.9
Drain openers	17	1.5
Adhesives	9	0.8
Detergents	13	1.2
Other	53	4.8
Home repair products	54	4.8
Wood working products	41	3.7
TOTAL	1,115	100

TABLE 1: Distribution of household hazardous waste items in a sample of 21 homes.

Organohalogens entering the sewerage system from industrial sources could, in theory, cover the full spectrum of industrial chlorinated chemicals. In the UK, industry is actively encouraged to connect to the public sewer network as part of generalised pollution control initiatives (Belshaw & Fisher 1992). Industrial effluents may contain a wide variety of chemicals and they may vary considerably with time (Johnston *et al.* 1991; Matthiessen *et al.* 1993). Under UK domestic legislation, the details of the discharges remain confidential between the industrial concern and the utility company responsible for the sewer system. The complexity of industrial effluent and sewage mixtures adds considerably to the difficulties of analytical determinations.

TABLE 2

COLUMN 1			COLUMN 2			
3,8	1AW		methane, trichloro-	12		1,3-cyclopentadiene 1,2,3,4,5,5-hexachloro-
3	1AW	1AA	methane, tetrachloro-	4,6		benzene, chloro-
4			methane, bromodichloro-	4		benzene, 1-chloro-2-methyl-
10			methane, dibromochloro-	4		benzene, dichloromethyl-
4			methane, dibromo-	4		benzene, 1,2-dichloro-4-methyl-
4,7			methane, tribromo-	4		benzene, 1,3-dichloro-2-methyl-
3,4,6			ethane, 1,1-dichloro-	4		benzene, 1-chloro-4-(chloromethyl)-
8,9	1AW	1AA	ethane, 1,2-dichloro-	6,12	1AW 1AA	benzene, 1,2,4-trichloro-
4,10,13		1D	ethane, 1,1,2-trichloro-	12	1AW 1AA	benzene, 1,3,5-trichloro-
12		1D	ethane, 1,1,2,2-tetrachloro-	12		benzene, 1,2,3,4-tetrachloro-
12		1D	ethane, hexachloro-	6		benzene, 1,2,3,5-tetrachloro-
2			ethane, bromodichloro-	12		benzene, 1,2,4,5-tetrachloro-
12			ethane, 1,1'-oxybis[2-chloro]-	12		benzene, pentachloro-
12			ethane, 1,1'-oxybis[1-chloro]-	12	1AW 1AA	benzene, hexachloro-
5,13		1D	ethene, chloro-	14		benzenamine, 2-chloro-
3,6	1AW	1AA	ethene, trichloro-	8		benzenamine 3,5-dichloro-
3,4,6	1AW	1AA	ethene, tetrachloro-	14		phenol, 2,4-dibromo-
14			acetonitrile, dichloro-	14		2,4,6-tribromo-phenol
11		1D	propane, 1,2-dichloro-	12		ethanone, 1-(4-chlorophenyl)-
6		1D	1-propene, 2,3-dichloro-	12		benzamide, 4-chloro-N-(1,1-dimethyl)-
12			1-propene, 1,2,3,3-tetrachloro-	12		oxazole, 5-bromo-4-phenyl-
12			1-propene, 3,3,3-trichloro-2-methyl-	12		benzene, pentachloro-(trichloroethenyl)-
12			butane, 1,2,2,4-tetrachloro-	12		benzene, pentachloro-(trichloroethyl)-
12			butane, 1,2,3,4-tetrachloro-	12		4-fluorobenzoylcarboxylic acid
12			2-butene, 1,4-dichloro-	12		benzamide, 2-chloro-N-(4-hydroxyphenyl)-
6			cyclobutene, 3,4-dichloro-	13		benzene, 1-chloro-2-[2,2-dichloro-1-
6			1,3-butadiene, 1,1,3,4-tetrachloro-	13		-(4-chlorophenyl)ethenyl]-
12			1,3-butadiene, pentachloro-	13		benzene, 1,1'-(2,2,2-trichloroethylidene),
12			1,4-butadiene, pentachloro-	13		bis[4-chloro]-
3,6,12	1AW		1,3-butadiene, 1,1,2,3,4,4-hexachloro-			

TABLE 2: Halogenated compounds identified in effluents and wastes at PVC manufacturing sites in Europe Sites 1-4: Belgium; Sites 5,6: France; Sites 7-9: Germany; Sites 10,11: Netherlands; Site 12: Sweden; Site 13-14: United Kingdom

Key to results tables: Compounds listed have been identified to >90% certainty.

1AW: compound is listed in annex 1A of the Final Declaration of the Third International Conference on the Protection of the North Sea (MINDEC 1990) as a priority hazardous substance in water; scheduled for >50% reduction

1AA: compound is listed in annex 1a of the Final Declaration of the Third International Conference on the Protection of the North Sea (MINDEC 1990) as a priority hazardous substance in air; scheduled for >50% reduction

1D: compound is listed in Annex 1D of the Final Declaration of the Third International Conference on the Protection of the North Sea (MINDEC 1990) for consideration for prioritisation.

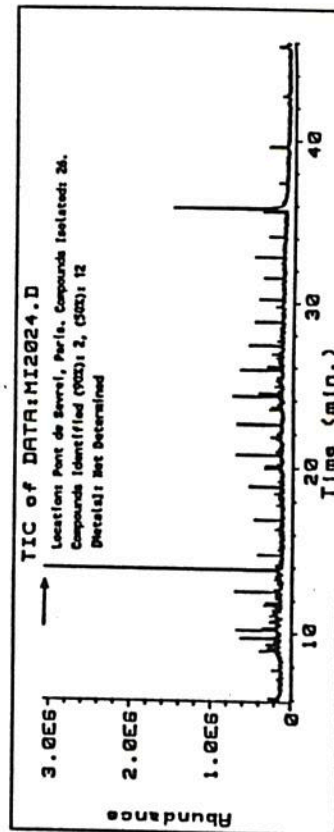
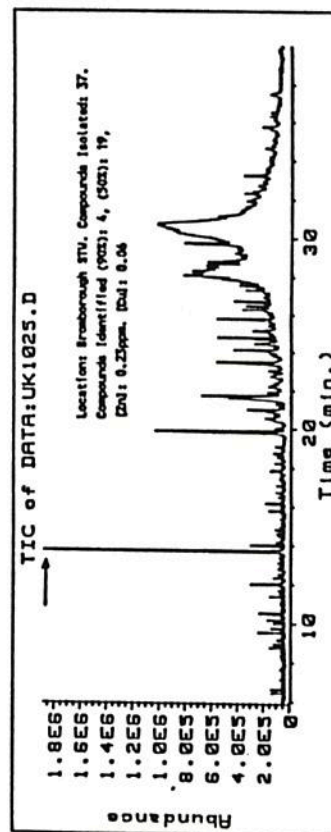
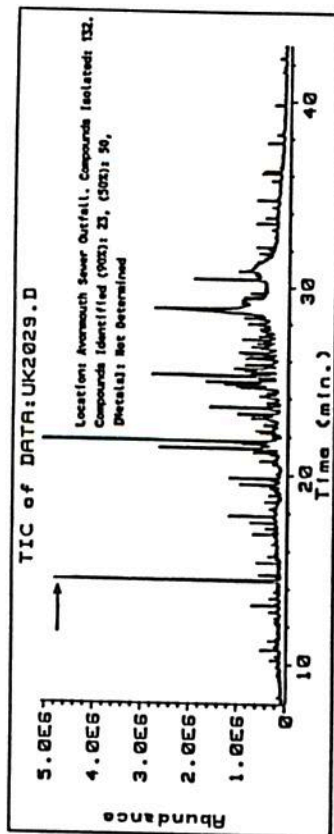
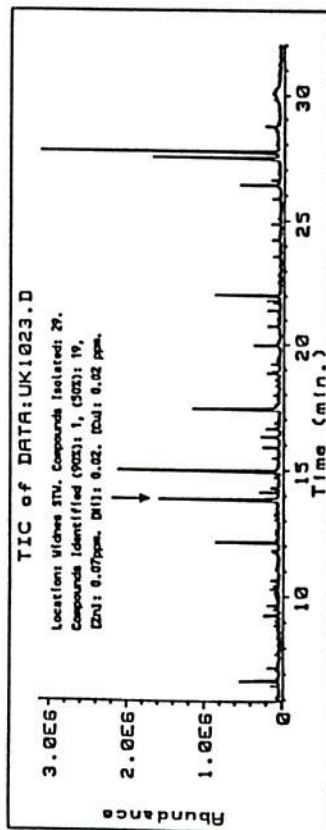
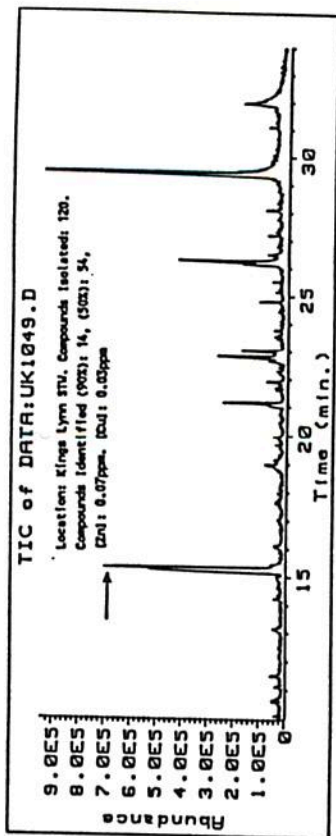
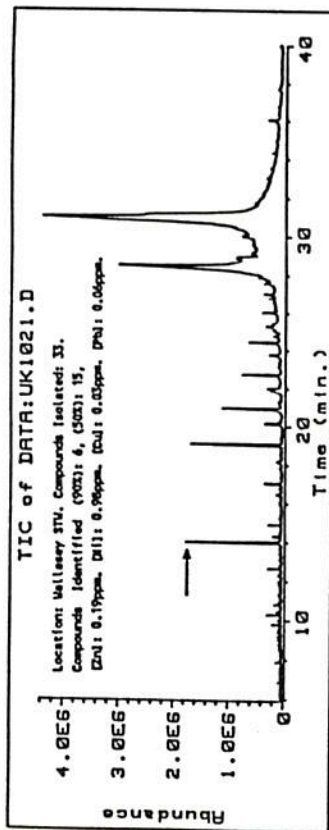


Figure 1: Total ion chromatograms obtained from the analysis of sewage effluents from various locations. The arrow identifies the peak due to the internal standard added at 200ppb. Numbers of isolated and identified compounds are given in each case, together with metal concentrations determined by ICPAES

A characteristic of industrial chlorine chemistry is that many of the reactions are non-specific. A wide variety of organochlorine compounds are present in the effluents derived from the oxychlorination process used in the manufacture of ethylene dichloride (EDC) for example. Other chlorinated compounds are used in PVC formulation. Table 2 lists the chlorinated compounds present in effluent samples obtained from PVC manufacturing facilities in Europe as published by Johnston *et al.* (1994). The acceptance of such effluents for treatment in municipal sewage treatment works is questionable since many of the organohalogenes will resist degradation. This practice has the potential to add considerably to both the regulatory burden and to the chemical analysis required for its policing and enforcement. One aspect of this is that the partial degradation of organohalogenes may result in a suite of further uncharacterised compounds (Dolfing 1994). At present, knowledge of the significance of partial degradation products and metabolites is limited.

2. CHEMICAL COMPLEXITY OF SEWAGE EFFLUENTS

Sewage effluents are complex mixtures of chemicals. Rogers (1987b) provides an analysis of the major organic constituents in solution and in suspension in domestic sewage (TABLE 3).

Component	Concentration (mg/l)	%Total Organic Carbon
IN SOLUTION		
Sugars	70	31.3
Non-volatile acids	34	15.2
Volatile acids	25	11.3
Free amino acids	5	3.1
Bound amino acids	13	7.6
Detergents	17	11.2
Uric acid	1	0.5
Phenols	0.2	0.2
Organic Carbon	90	-
IN SUSPENSION		
Fats	140	50
Proteins	42	10
Carbohydrates	34	6.4
Detergents	5.9	1.8
Amides	2.7	0.6
Amino acids	1.7	0.3
Soluble acids	12.5	2.3

TABLE 3: Composition of domestic sewage effluent with respect to the major organic components present in suspension and in solution

This "average" composition can be modified considerably by the

admixture of industrial effluents and urban runoff. Figures 1 shows gas chromatograms obtained from the simple hexane extraction of various sewage effluents as published by Johnston *et al.* (1993). Of these, the one most closely representing a "normal" domestic effluent is designated as UK1025.D. The presence of the large baseline excursion around 30 minutes is due to the presence of fatty acids. The smaller this excursion, the lower the proportion of domestic sewage actually present in the influent stream. Hence sample UK1023.D appears to be almost entirely composed of industrial wastewater. The figures also show the number of peaks isolated under identical conditions of data analysis and those positively identified by GC-MS (90% probability) and those tentatively identified (50% probability).

The figures and the data illustrate some important points, equally applicable to industrial effluents. The numbers of compounds positively identified as a proportion of those isolated is consistently low. The chemical "fingerprint" established by the analyses differs markedly between samples. Jobling *et al.* (1995) note that they were unable to find a report of a full chemical characterisation of sewage effluent. They also make the point that microbial transformation of chemicals will result in a wide range of products, many of which are unidentified. They suggest that the numbers of chemicals actually identified in analyses may represent only 20% of those actually present. The data of Johnston & Stringer (1991) suggest that the figure may be considerably lower than this. In addition, analysis using solvent extraction will only address compounds having a limited range of polarities. Highly polar compounds will not be extracted. Moreover, analytical interferences are likely to further restrict the numbers of compounds isolated, particularly those present at low concentration. The selectivity of the detector used by the analytical instrument must also be considered, since this too will restrict the number of compounds actually registered in the analysis.

It seems unlikely, therefore, that full characterisation of a sewage effluent will be possible in practice. This extends equally to the organohalogenes present and to their degradation products. This premise is borne out by the results of the AOX and EOX analyses reported in OSPAR (1994). The presence of organohalogen compounds present in sewage effluents has largely been determined by the use of the AOX (Adsorbable organohalogenes) or EOX (Extractable organohalogenes) (see: Document ORGHAL II/1.Add 1; OSPAR 1994). These group parameters, while useful for the purposes of broad evaluation, have limitations as discussed by Stringer & Johnston (1995).

There are serious concerns about the AOX parameter and indeed other sum parameters used for regulatory purposes. It has been stated that there is no direct link between AOX levels in effluent and its toxicity (Folke 1991). Neilson *et al.* (1991) also consider EOCl and AOX sum parameters to be of value only for control programmes. Such measurements provide only a circumstantial link with putative discharges and do not allow a causal relation between the compounds produced and the observed ecosystem effects to be determined. With specific reference to

cyclohexane extracts for determination of EOC1 the major organochlorine fraction remains unidentified.

The experience in the Netherlands where it has not proven possible to reconcile AOX in sewage with either specific compounds, or to a source, is common to other attempts to effect such a reconciliation in other environmental media. For example, Wesen *et al.* (1990) found that only 10-15% of the extractable organically bound chlorine present in fish in the Baltic could be attributed to specific organochlorines. This figure fell to 5% when sediments were considered. The authors concluded that much of the chlorine was bound in the form of esters and would require chemical derivatisation for successful analysis. Similarly, Newsome *et al.* (1993) found that levels of organically bound chlorine in Great Lakes fish were between 5 and 72 times higher than those attributable to PCBs and chlorinated pesticides. Moreover, the list of persistent chlorinated organic compounds found to be accumulated by aquatic species continues to grow. Two recent examples are the finding of tris(4-chlorophenyl) methanol and tris (chlorophenyl) methane as apparently ubiquitous contaminants (Jarman *et al.* 1992 and the isolation of bis(4-chlorophenyl) sulfone in Baltic wildlife species (Olsson & Bergman 1995). No unequivocal source attribution for these chemicals has been made to date.

Hence, attempts to control organochlorines present in sewage simply by restricting AOX may not adequately control those of the greatest environmental significance. AOX cannot be used as a sole regulatory parameter, only as an indicator. Many highly significant halogen compounds are likely to be present at low concentration. For example, the chlorinated dioxins and dibenzofurans may only be present at the part per trillion level, and would be impossible to control using AOX as a sole parameter. Further, full characterisation of the organohalogens present in complex effluents is beyond the capacity of current, routine analytical methods.

4. THE EXAMPLE OF CHLORINATED PARAFFINS

The points raised above may be illustrated by reference to the chlorinated paraffins. These have been manufactured on a commercial basis for some 50 years (Houghton 1994). They are produced in a variety of chain lengths from ten to thirty carbons by the direct chlorination of paraffinic feedstock and the chlorine content varies between 30-70% by weight. Hence, these products are mixtures rather than single substances. These chemicals are relatively inert, exhibit considerable resistance to chemical attack and are hydrolytically stable. The chlorinated paraffins are versatile materials with numerous applications. They are used as plasticisers in sealants, PVC, surface coatings, paints and varnishes. In these applications their fire retardant properties are also exploited. Metal working lubricants and heavy duty gear oils subject to extreme working pressures contain chlorinated paraffins. They are used in fat liquors in the leather industry and as solvents in carbonless copy papers. The largest single national market is in the US at approximately

40,000 tonnes per annum with Europe as a whole utilising some two and a half times this amount. Excluding China and the former Soviet Union, global production is around 300kt per year. Production increased steadily throughout the 1980's

Although the paraffinic feedstocks are relatively pure, they may still contain 1-2% isoparaffins and around 100mg/kg of aromatics. This raises the possibility that aromatic chlorinated compounds may be produced as by-products in the production process.

The precise uses of the chlorinated paraffins differ regionally as indicated by the relevant Figure in Document ORGHAL 1/2-Add.1, produced from the data of Svanberg (1983). Other reviews have recently been conducted on various aspects of chlorinated paraffins in the environment (BUA 1992; Back et al. 1994). The conclusions of all three reports are broadly similar. These compounds are often used in open ended applications and there are considerable gaps in the data concerning environmental fate and effects. They are known to be widely distributed in the environment with high concentrations in sewage sludge, sediments and in biota.

In terms of monitoring, chlorinated paraffins are difficult to extract and clean up for analysis. They are difficult to identify unequivocally using standard GC-MS techniques and to date there appears to have been no international intercalibration exercise designed for the purposes of quality assurance/control of the data. In addition there appear to be no data on individual toxicities. There appear to be no simple and generalised analytical techniques for the analysis of these compounds. As a group they have a low acute toxicity, but there is great interspecific variation in toxicological response. They cause skin and eye irritation and exhibit hepatotoxic, nephrotoxic, thyrotoxic and neurotoxic properties. Reproductive toxicity has also been observed. The short chained highly chlorinated paraffins are regarded as carcinogenic and teratogenic. The toxicological responses are generally greater for short chain chloroparaffins and for more highly chlorinated paraffins. It is possible that the long and medium chain paraffins may degrade to form short chain compounds but this needs confirmation.

Hence, these compounds are known to be toxic, persistent and bioaccumulative. Although there is little evidence that they are biomagnified, the highly chlorinated short chain paraffins are retained by fish and shellfish for long periods after exposure is terminated. Indeed, their potential environmental hazards have been known since the 1970's. Regulation, however, has centred upon only the short chain chloroparaffins culminating in the recent OSPAR agreement to phase out the use of the short chain compounds with greater than 48% chlorine by weight. This phase out, with certain national reservations by the UK, Portugal and Spain is set to become effective by the end of 1999.

The chlorinated paraffins highlight a number of issues in relation to the control of organohalogens entering sewerage systems. They can enter this area of the hydrocycle as a result of release from numerous open ended applications. They are not easily analysed in

environmental media and their environmental fate is poorly understood. Given the concentrations likely to be present in sewage effluent (in the microgramme per litre range) the control of AOX will not effectively control emissions of chloroparaffins. Moreover, considerable uncertainties exist in relation to the environmental fate and behaviour of the medium and long chain chloroparaffins, suggesting that precautionary regulation is appropriate.

5. TOWARDS REALISTIC REGULATION AND CONTROL

On the basis of the considerations above several points emerge which have a direct impact upon the regulation of organohalogen substances emitted into and ultimately from sewerage systems.

- 1) It is unlikely that a full characterisation of the organohalogens present in domestic products will be feasible.
- 2) It is unlikely that a full characterisation of organohalogens present in sewage effluents will be feasible.
- 3) The admixture of industrial effluents with domestic sewage is likely to considerably complicate the analysis of organohalogens in sewage, and their subsequent regulation.
- 4) Full source reconciliation of organohalogens present in sewage is not likely to be feasible.
- 5) AOX is likely to be of limited utility as a regulatory parameter or as a gauge of the effectiveness of regulatory action to reduce organohalogens emitted with sewage.

Under these circumstances a regulatory approach based upon analytical characterisation of single organohalogens present in the products and the subsequent use/disposal chain is not likely to address the problem. The regulation of organohalogens using solely AOX as a parameter may well lead to reductions, but leave significant compounds unregulated. The production of specific organohalogens is generally accompanied by the co-production of unwanted halogenated compounds since reactions are generally non-specific.

Accordingly, a different approach is required based upon the organohalogens being treated as a class, or at least as a series of subgroups. This approach is not without precedent. The PCBs, chlorinated naphthalenes, certain of the chlorinated pesticides (eg chlordane and toxaphene), the chlorinated dioxins, the chlorinated solvents and the CFCs are all regulated to a greater or lesser extent on a group basis. Indeed it would make little sense in regulatory terms to do otherwise. In addition the production of organohalogens inevitably results in the by-production of a spectrum of other chemicals. As indicated by the data in Table 2, a suite of chlorinated benzenes and butadienes can be produced by oxychlorination. Extending this regulatory approach to include, for example, the chlorinated benzenes, the

chlorinated butadienes, the chlorinated toluenes the chlorinated nitrobenzenes, would be a logical progression and a further step away from the substance by substance approach currently prevalent.

Ultimately, however, the most simple way to address the problem of regulating organohalogenes from both diffuse and point sources would be to regulate the whole class of chemicals. Indeed, this is likely to prove to be the only effective way to meet the commitments of both the 1992 OSPAR Ministerial Declaration and the goals set in Para 17 of the 4th North Sea Ministerial Declaration. On the basis that the simplest method of controlling pollution is to prevent it, then such a strategy would target the production of these chemicals and their subsequent use. In this way progressive control over the problems of disposal and release through usage would automatically follow.

REFERENCES

Back, J., Olesen, S.I. & Andersen, S.H. (1994) Klorparaffiner i Danmark. Miljøprojekt nr. 248. Publ Miljøministeriet Miljøstyrelsen, Copenhagen 139pp.

Bass, E.S., Calderon, R.L. & Khan, M.E. (1990) Household hazardous waste: A review of public attitudes and disposal problems. J. Environ. Health 52 (6): 358-361.

Belshaw, C. & Fisher, T.J. (1992). Control of industrial waste discharges in the North West of England. Wat. Sci. Technol. 25 (1): 53-59.

BUA (1992) Chlorparaffine. Beratergremium für umweltrelevante Altstoffe (BUA) der Gesellschaft Deutscher Chemiker. Publ. VCH, Weinheim 227pp.

Clark, L.B., Rosen, R.T., Hartman, T.G., Alaimo, L.H., Louis, J.B., Hertz, C., Ho, C.-T. & Rosen, J.D. (1991). Determination of non-regulated pollutants in three New Jersey publicly owned treatment works (POTWs). JWPCF 63 (2): 104-113.

Clark, R.B. (1992) Marine Pollution. Third Edition. Publ. Clarendon Press, Oxford. pp9-27

Dolfing, J. (1994). Aerobic and Anaerobic biodegradation of organochlorines. Proc. Symposium: Chlorinated Organic Chemicals, Their Effect on Human Health and the Environment, Berlin, 19-21 Sept 1994. Publ. Toxicology Forum, Washington DC. pp270-298

Folke, J. (1991) Regulatory requirements for pulp and paper mill effluent control: Scientific basis and consequences. Water Science & Technology 24 (3/4): 19-31.

Hathaway, S.W. (1980) Sources of toxic compounds in household waste water. USEPA Report No: EPA-600/2-80-128. Published United States Environmental Protection Agency, Washington DC. 83pp.

Houghton, K.L. (1994) Chlorinated Paraffins. In: Kirk-Othmer

Encyclopedia of Chemical Technology. 4th Edition, Volume 4. Publ. Wiley Interscience pp. 79-87.

IJC (1992) Sixth Biennial Report on Great Lakes Water Quality. Publ. International Joint Commission, Ontario Canada. 59pp.

IJC (1994) Seventh Biennial Report on Great Lakes Water Quality. Publ. International Joint Commission on the Great Lakes.

Jarman, W.M., Simon, M., Norstrom, R.J., Burns, S.A., Bacon, C.A., Simoneit, B.R.T. & Riseborough, R.W. (1992) Global distribution of tris(4-chlorophenyl) methanol in high trophic level birds and mammals, Environ. Sci. Technol. 26 (9): 1770-1774.

Jobling, S., Reynolds, T., White, R., Parker, M.G. & Sumpter (1995) A variety of environmentally persistent chemicals including some phthalate plasticizers are weakly estrogenic. Environ. Health Perspect. 103 (6): 582-587

Johnston, P.A. & Stringer, R.L. (1991) Analytical difficulties in the full characterisation of effluents. Analytical Proceedings 28: 249-250.

Johnston, P.A., French, M.C. & Stringer, R.L. (1991) Pollution of UK estuaries: historical and current problems. Sci. Tot. Environ. 106: 55-70

Johnston, P.A., MacGarvin, M., Stringer, R.L., Troendle, S. & Swindlehurst, R.J. (1993) Sewage: Towards realistic environmental protection. Wat. Sci. Technol. 27 (5/6): 481-491.

Johnston, P.A. & Stringer, R.L. (1994) Environmental Significance and Regulation of Organochlorines. Proceedings of the Symposium on Chlorinated Organic Chemicals, Sept 19-21, 1994, publ. Toxicology Forum, Berlin.

Johnston, P.A., Stringer, R.L., Clayton, R. & Swindlehurst, R.J. (1994) Regulation of Toxic Chemicals in the North Sea: The need for an Adequate Control Strategy. North Sea Monitor 12 (2): 9-16

Kuhnt, G. (1993) Behaviour and fate of surfactants in soil. Environ. Toxicol. Chem. 12 (10): 1813-1820.

Lester, J.N. (1990) Sewage and sewage sludge treatment. In: Harrison, R.M. (ed). Pollution: Causes, effects and control. Publ. Royal Society of Chemistry, Cambridge, UK. pp33-62.

Matthiessen, P., Thain, J.E., Law, R.J. & Fileman, T.W. (1993) Attempts to assess the environmental hazard posed by the complex mixtures of organic chemicals in UK estuaries. Mar. Poll. Bull. 26 (2): 90-95.

Mayer, G.J., Cheng, I.S., Pau, P. & Mohamed, F. (1994) Emissions of air toxics from wastewater treatment plants. Wat. Environ. Res. 66 (2): 140-144

McIntyre, A.D. (1990) Sewage in the Sea. Technical annexes to the

Report on the State of the Marine Environment Annex XII. UNEP Regional Seas Reports and Studies No. 114/2.

McIntyre, A.D. (1988) Pollution in the North Sea from oil related industry. In. Newman P.J. & Agg, A.R. (Eds) Environmental Protection of the North Sea. Publ. Heinemann pp.425-445

MINDEC (1990) Final Declaration of the 3rd Ministerial Conference on the Protection of The North Sea. Ministry of Transport Conference on the Protection of The North Sea. Ministry of Transport and Public Works, The Hague 36pp.

Newsome, W.H., Andrews, P., Conacher, H.B.S., Rao, R.R. & Chatt, A. (1993) Total organochlorine content of fish from the Great Lakes. J. Ass. Off. Anal. Chem. 76 (4): 703-706.

Nielson, A.H., Allard, A.-S., Hynning, P.-A. & Remberger, M. (1991). Distribution, fate and persistence of organochlorine compounds formed during production of bleached pulp. Toxicology & Environmental Chemistry 30: 3-41

Nisbet, A. (1989) Chlorine and Caustic Soda. Chemical Business, December 1989 pp24-26

Nriagu, J.O. & Pacyna, J.M. (1988) Quantitative assessment of worldwide contamination of air, water and soils by trace metals. Nature 333: 134-139

NSTF (1993) North Sea Quality Status Report. North Sea Task Force. Publ. Oslo and Paris Commissions, London 132pp.

Olsson, A. & Bergman, A. (1995). A new persistent contaminant detected in Baltic wildlife: Bis(4chlorophenyl) sulfone. Ambio 24 (2): 119-123.

OSPAR (1994) Products containing or generating Organohalogenes. Presentation by the Netherlands to the Oslo and Paris Convention Working Group on Diffuse Sources. Document No: DIFF 94/5/2. Oslo and Paris Commissions, London 13pp.

Richardson, M.L. & Bowron, J.M. (1985) The fate of pharmaceutical chemicals in the aquatic environment. J. Pharm. Pharmacol. 37: 1-12

Rogers, H.R. (1987a) Occurrence and Fate of Synthetic Organic Compounds in Sewage and Sewage Sludge- A Review: Appendices. Publ. WRC Environment, Medmenham UK. Report No:PRD 1540-M.

Rogers, H.R. (1987b) Occurrence and Fate of Synthetic Organic Compounds in Sewage and Sewage Sludge: A Review. Publ. WRC Environment, Medmenham UK. Report No:PRD 1539-M.

Rogers, H.R., Campbell, J.A., Crathorne, B. & Dobbs, A.J. (1989) The occurrence of chlorobenzenes and permethrins in twelve U.K. sewage sludges. Wat. Res. 23 (7): 913-921.

Stringer, R.L. & Johnston, P.A. (1995) Occurrence and toxicology of

natural and anthropogenic organohalogenes and relevance to environmental protection. In; Grimvall, A. & de Leer, E.W.B. (Eds) Naturally Produced Organohalogenes Publ. Kluwer Academic Publishers pp. 415-427.

Svanberg, O., Chlorinated Paraffins: A review of environmental behaviour and effects. Report No: PM 1614. Publ. National Swedish Environment Protection Board 52pp.

Verbanic, C.J. (1990) The Challenge for Chlorine and Caustic Soda in the 90's. Chemical Business, September 1990 pp21-35.

Wesen, C., Carlberg, G.E. & Martinsen, K. (1990) On the identity of chlorinated organic substances in aquatic organisms and sediments. Ambio 19 (1): 36-38

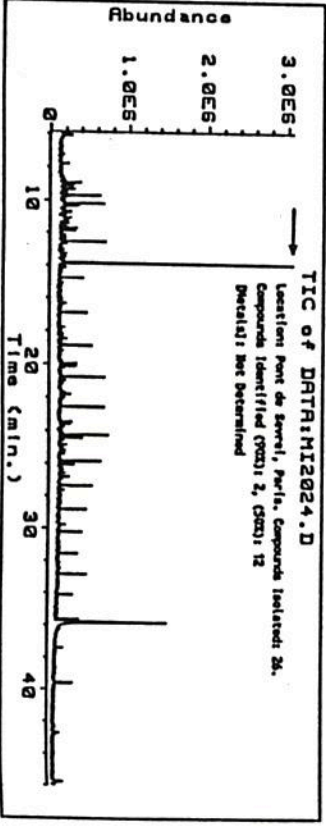
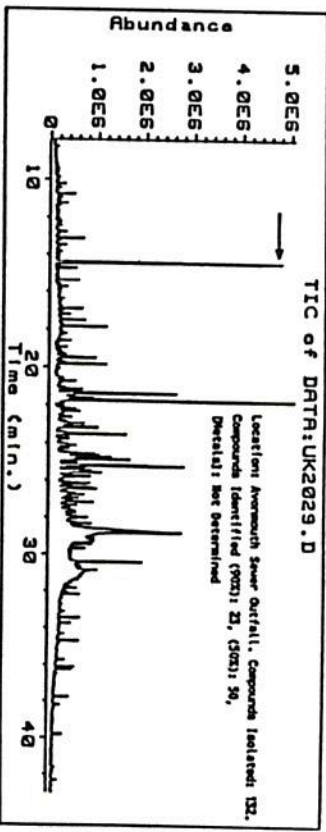
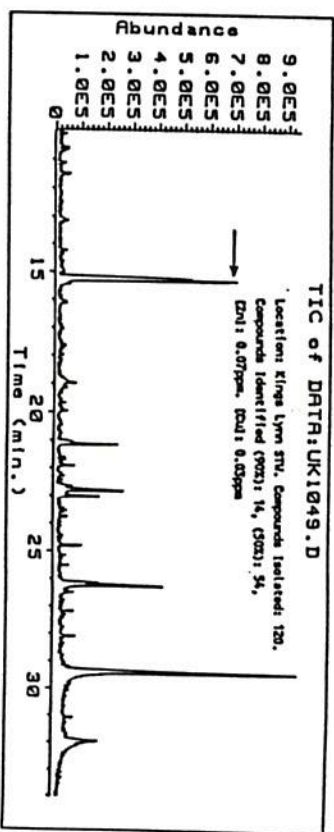
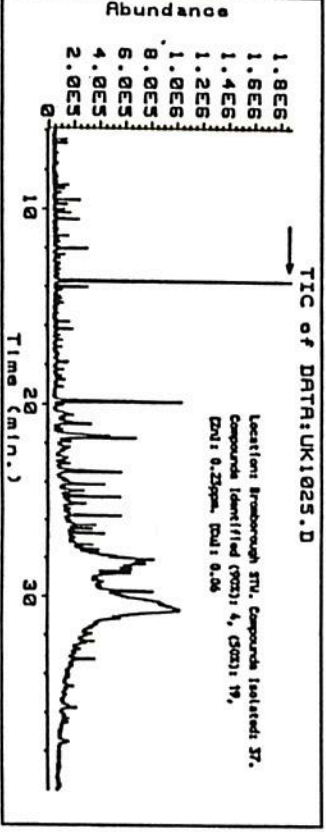
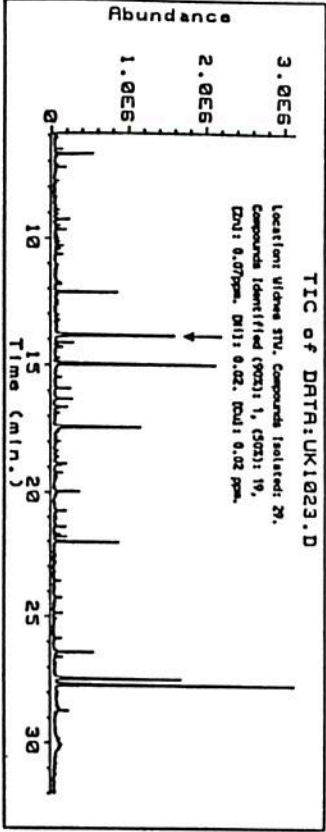
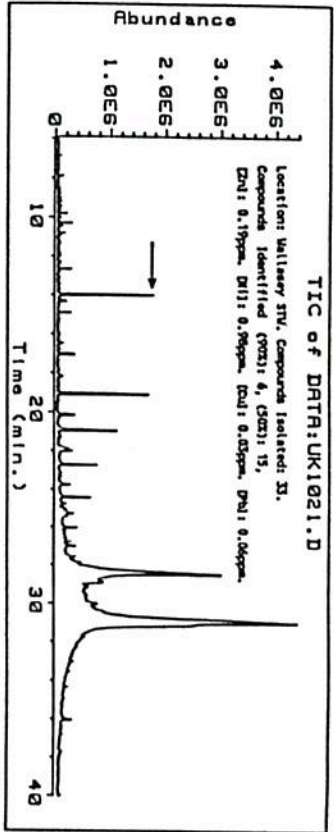


Figure 1: Total ion chromatograms obtained from the analysis of sewage effluents from various locations. The arrow identifies the peak due to the internal standard added at 200ppb. Numbers of isolated and identified compounds are given in each case, together with metal concentrations determined by ICPAES

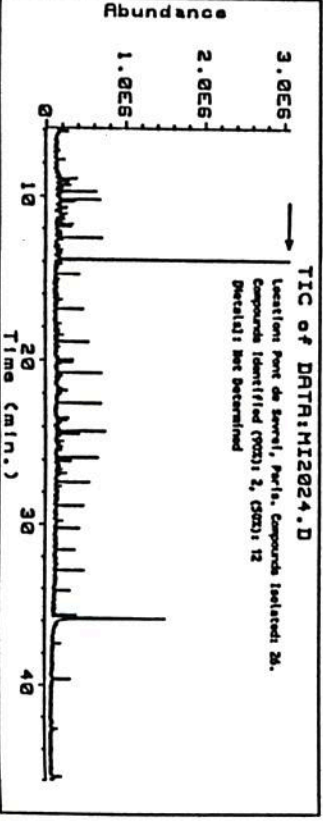
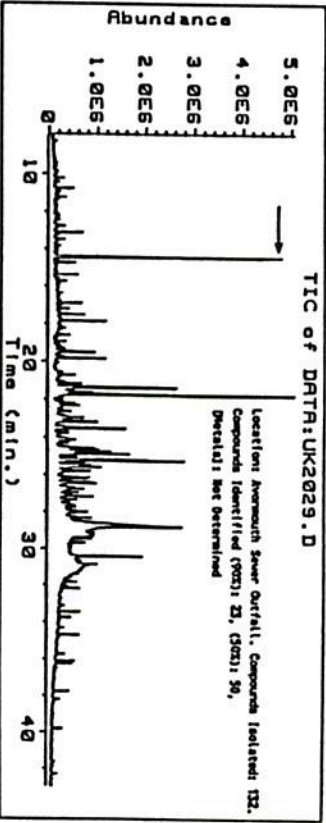
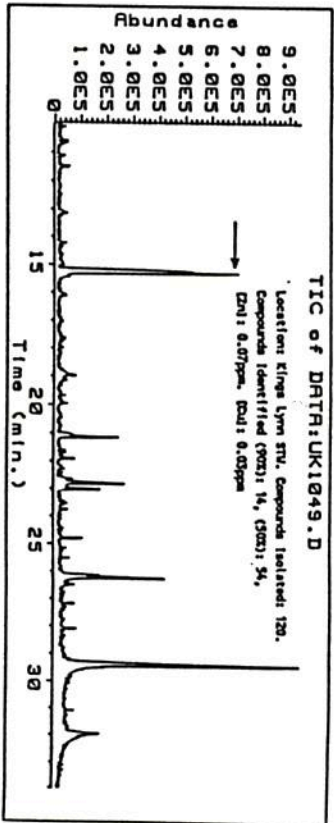
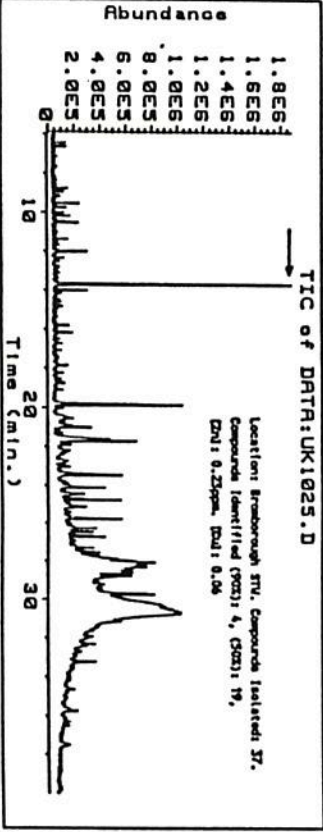
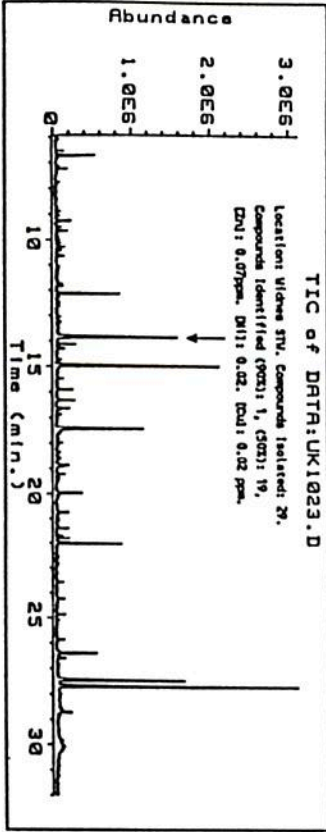
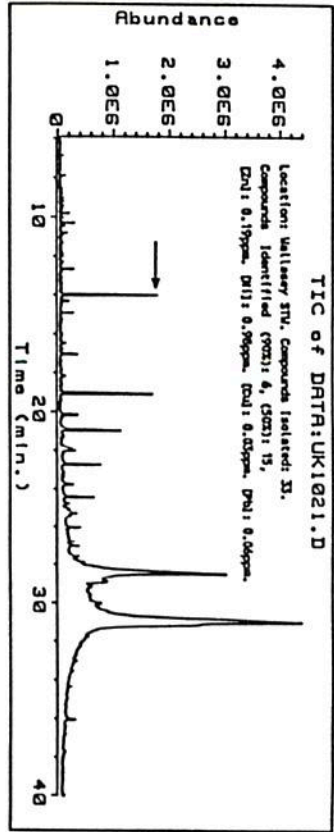


Figure 1: Total ion chromatograms obtained from the analysis of sewage effluents from various locations. The arrow identifies the peak due to the internal standard added at 200ppb. Numbers of isolated and identified compounds are given in each case, together with metal concentrations determined by ICPAES

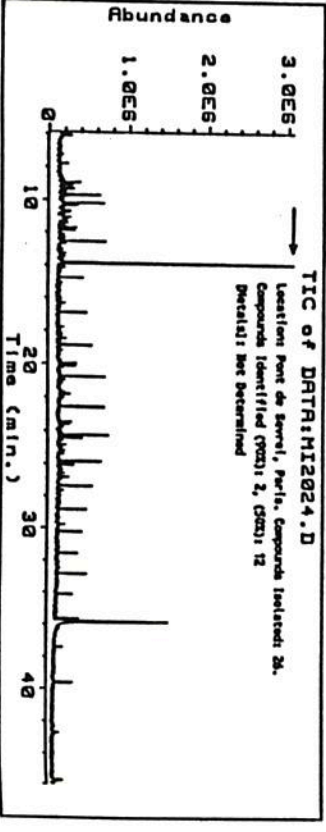
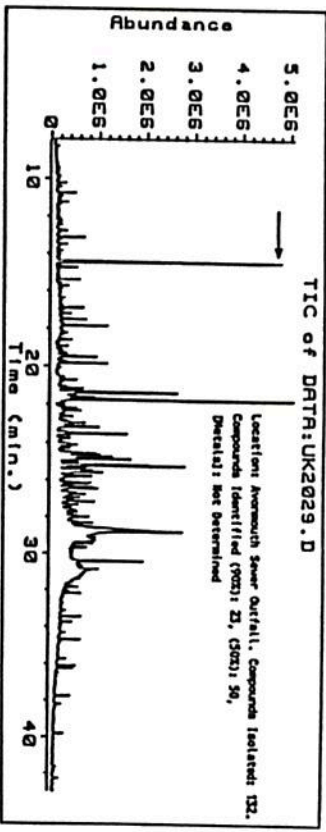
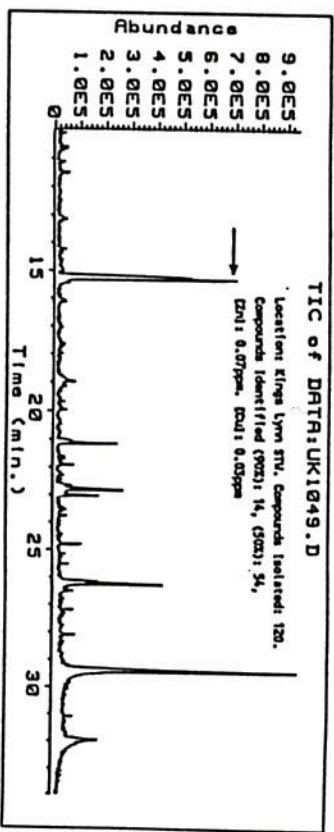
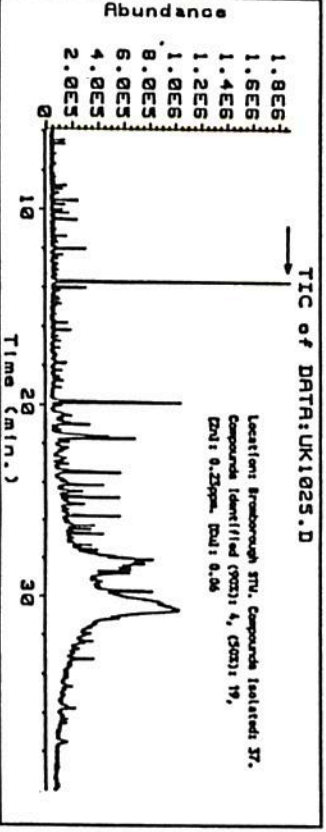
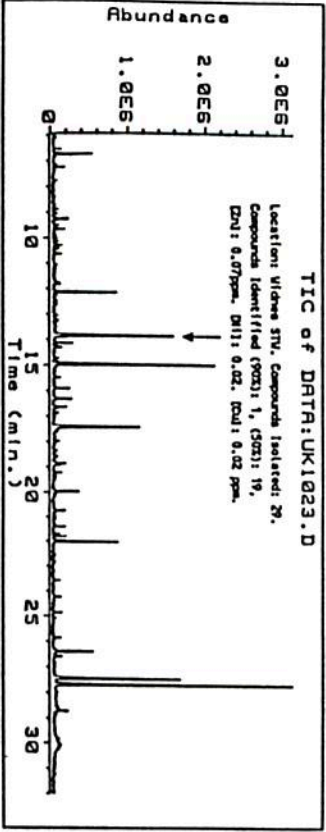
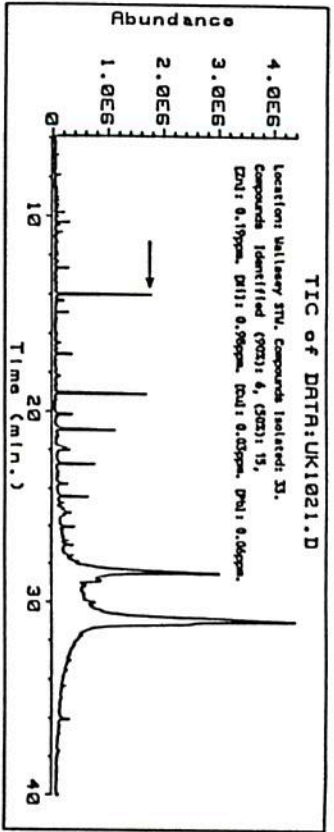


Figure 1: Total ion chromatograms obtained from the analysis of sewage effluents from various locations. The arrow identifies the peak due to the internal standard added at 200ppb. Numbers of isolated and identified compounds are given in each case, together with metal concentrations determined by ICPAES

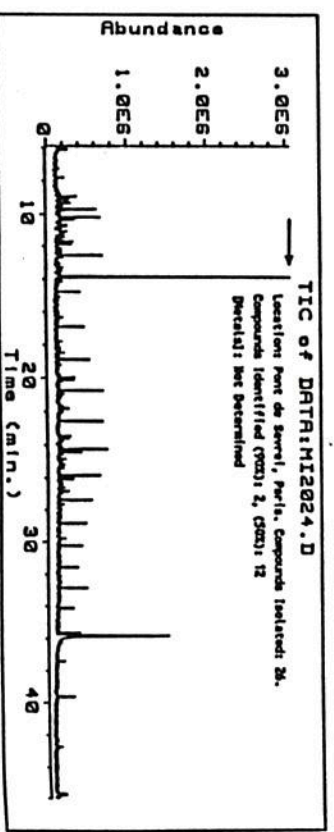
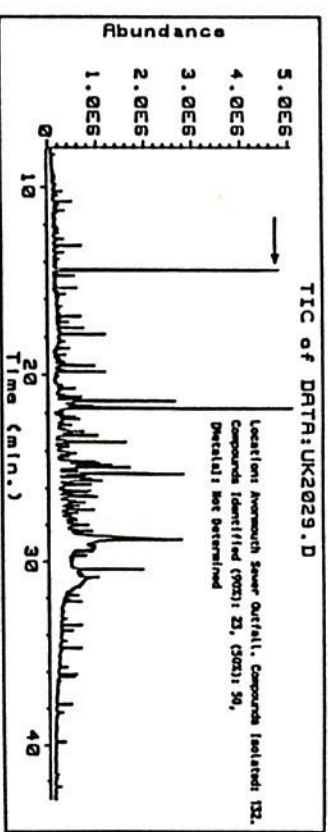
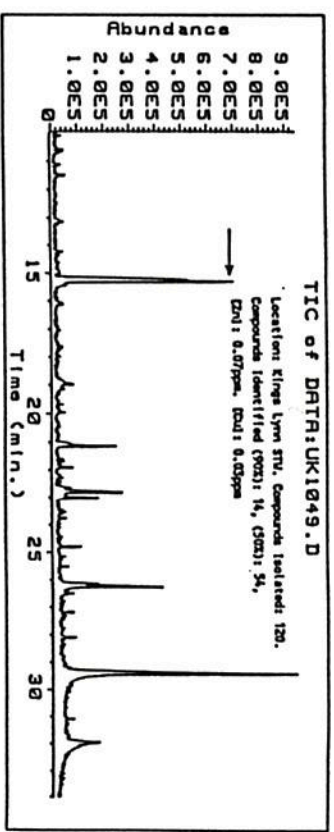
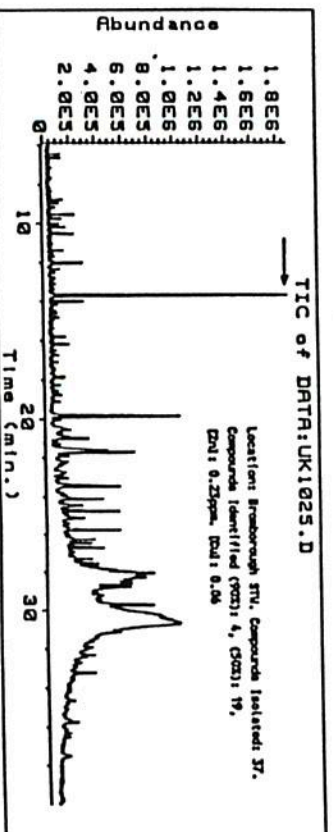
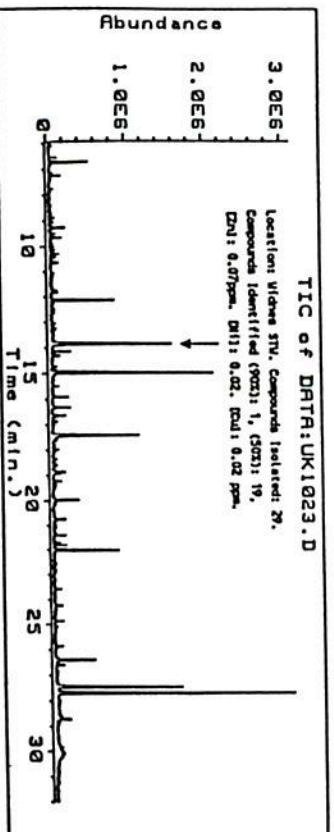
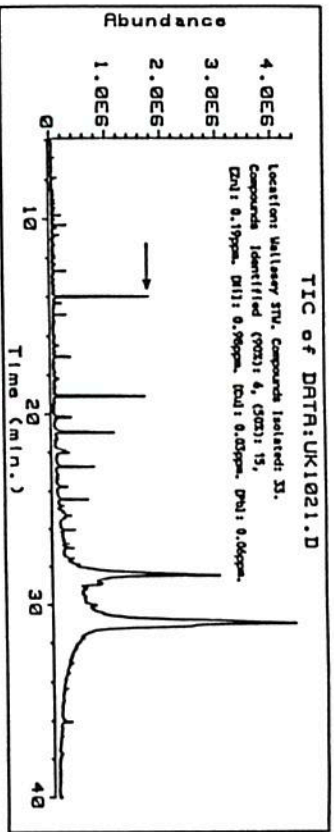


Figure 1: Total ion chromatograms obtained from the analysis of sewage effluents from various locations. The arrow identifies the peak due to the internal standard added at 200ppb. Numbers of isolated and identified compounds are given in each case, together with metal concentrations determined by ICPAES

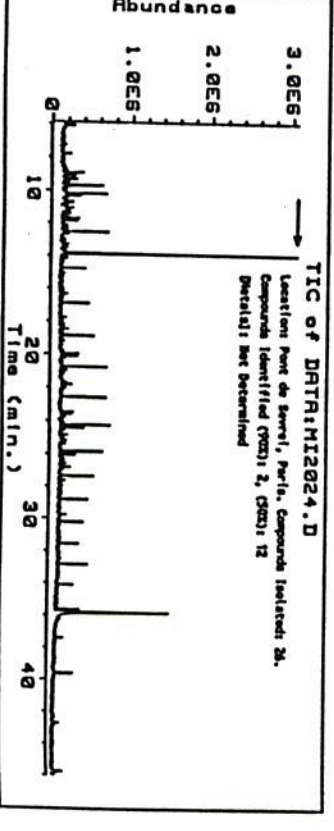
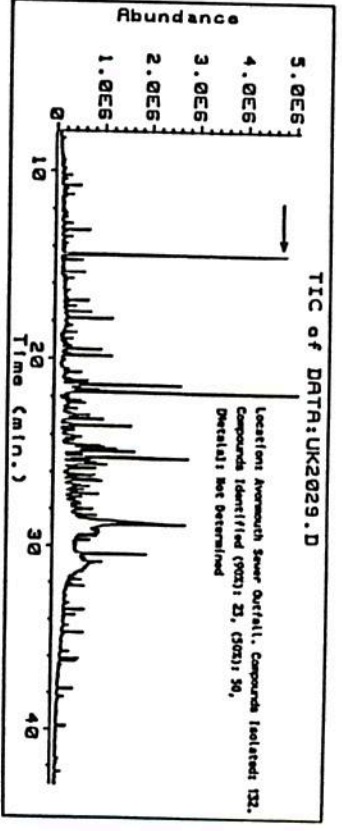
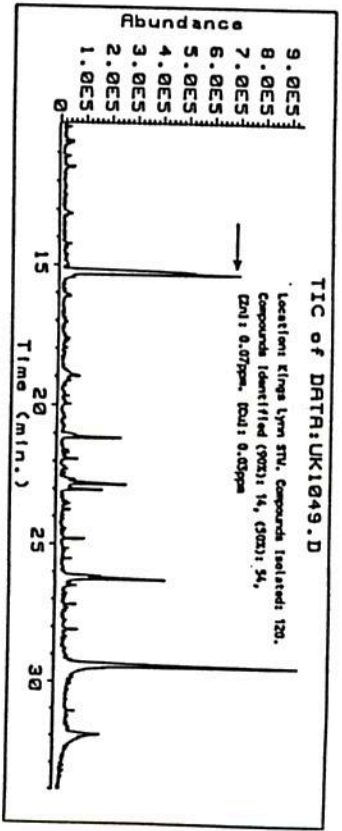
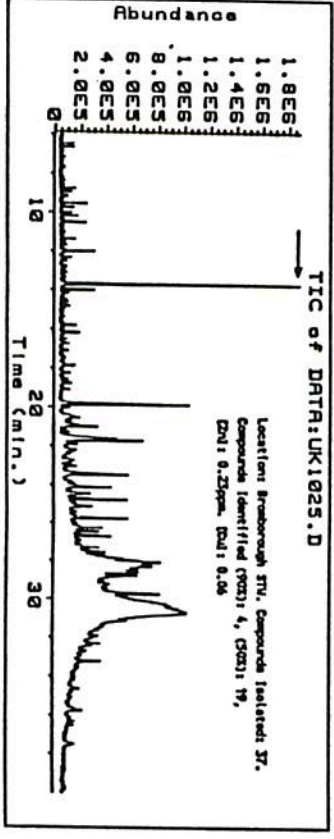
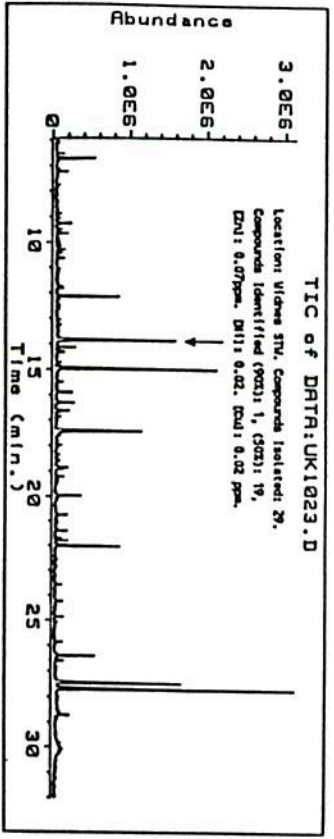
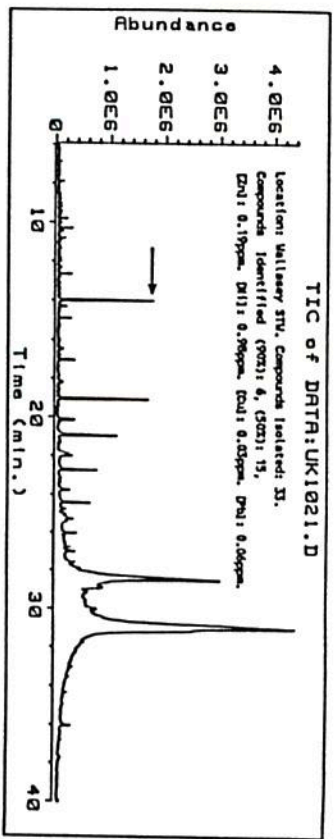


Figure 1: Total ion chromatograms obtained from the analysis of sewage effluents from various locations. The arrow identifies the peak due to the internal standard added at 200ppb. Numbers of isolated and identified compounds are given in each case, together with metal concentrations determined by ICPAES

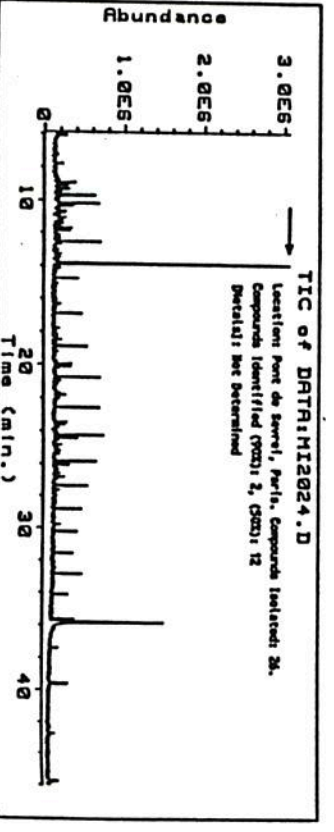
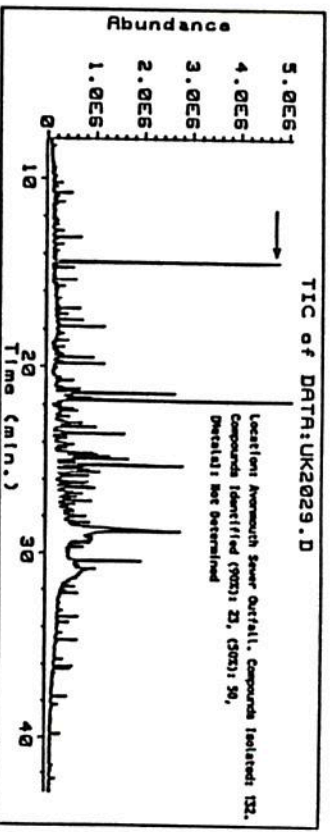
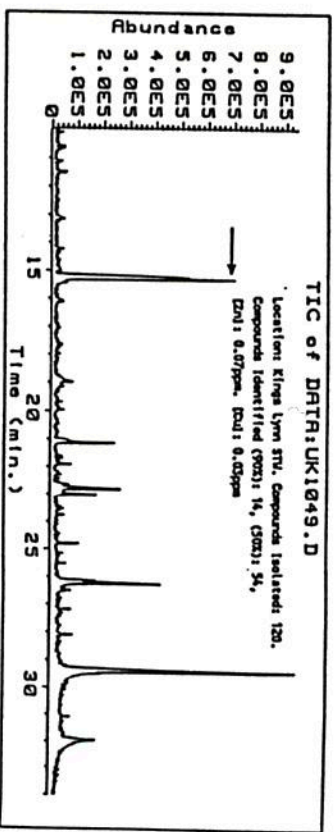
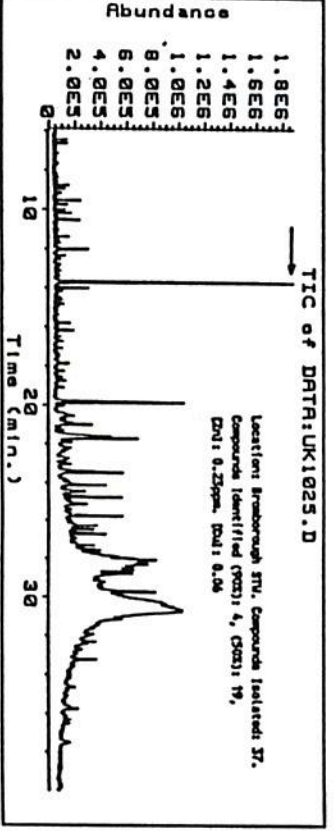
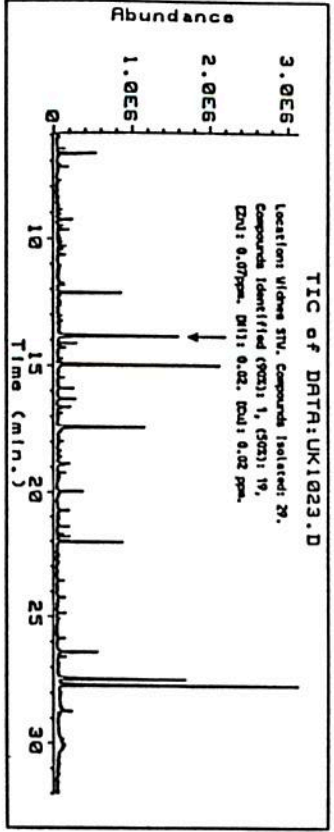
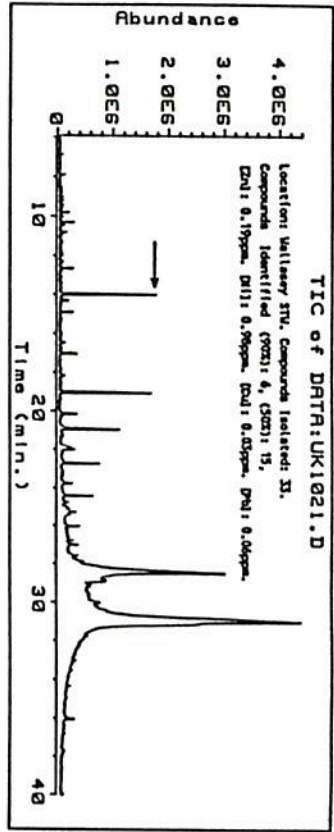


Figure 1: Total ion chromatograms obtained from the analysis of sewage effluents from various locations. The arrow identifies the peak due to the internal standard added at 200ppb. Numbers of isolated and identified compounds are given in each case, together with metal concentrations determined by ICPAES