Environmental Effects of Organic and Inorganic Contaminants in Sewage Sludge

Proceedings of a Workshop held at Stevenage,
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Commission of the European Communities

ENVIRONMENTAL EFFECTS OF ORGANIC AND INORGANIC CONTAMINANTS IN SEWAGE SLUDGE

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F O R E W O R D

This publication constitutes the Proceedings of two Seminars held at the Water Research Centre, Stevenage (United Kingdom) on May 25-26, 1982, under the auspices of the Commission of the European Communities, as part of the Concerted Action COST 68 ter "Treatment of Sewage Sludge". The Seminars were convened by Working Party 5 (Environmental Effects) of the Concerted Action to examine current knowledge concerning organic micropollutants (Seminar I) and inorganic contaminants (Seminar II) in sewage sludge utilised on agricultural land.

Continuing extension of sewage treatment in Europe is generating more sewage sludge and hence putting more pressure on disposal outlets. Agricultural land is a principal disposal route for sewage sludge and has the advantage that it involves the productive use of sludge to improve soil conditions and supply nutrients for crop growth. At the same time it is the route most sensitive to problems associated with organic and inorganic contaminants which may occur in sludge in higher concentrations than in soil. Considerable research effort is in progress within the Community to investigate the effects of these contaminants and to ensure that soil fertility, crops, livestock and the human foodchain are properly protected where sewage sludge is used on agricultural land. It was the aim of the Seminars to provide a forum for the exchange of recent research results and ideas on this subject.

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SESSION I - EFFECTS OF ORGANIC MICROPOLLUTANTS

OCCURRENCE, BEHAVIOUR AND FATE OF ORGANIC MICROPOLLUTANTS DURING WASTE WATER AND SLUDGE TREATMENT PROCESSES

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SUMMARY

The principal areas of concern when disposing of sewage sludges contaminated with organic micropollutants are outlined and the characteristics of the substances described. Some of the criteria which may be used to identify these substances are presented and the principal chemical classes listed. The sources of these substances and the routes by which they enter waste water treatment works are reviewed. Their fate and behaviour during conventional two stage waste water treatment is considered with emphasis upon their removal and biodegradation. It would appear that the majority of these materials are recalcitrant and liphophilic; they tend, therefore, to be closely associated with the sewage solids and it would seem probable that they are only biodegraded to a limited extent. As a consequence of this behaviour they are concentrated in the sewage sludges produced. Their behaviour during biological sludge treatment has been the subject of very limited study and it is not possible to draw conclusions from this limited information about the majority of these substances. The occurrence and range of concentrations of organic micropollutants in sewage sludges is reviewed as far as the available data permits. It is concluded that the principal area of concern is human health.

INTRODUCTION

There exists mounting evidence that long term exposure to low concentrations of certain organic chemicals can be an important factor in the development and manifestation of some chronic diseases. It is further believed that between 80% and 90% of cancer cases are of environmental origin, and, therefore, the extent to which the population is exposed to environmental chemicals has become of concern (1). When attempting to identify the contaminants of sewage sludge which could be of concern, it is useful to consider those which cause concern when present in waters. Since the principal reasons for concern are not dissimilar, in the case of sewage sludge, they are,

i) human health, particularly when sewage sludge is disposed to agricultural land or the sea;

ii) adverse effects on biological sludge treatment processes, anaerobic and aerobic digestion;

iii) effects upon the environment, particularly agricultural land and the sea.

The degree of concern will be influenced by the toxicity, persistence and bioavailability of the various substances. The persistence,or resistance to chemical and biological degradation is an important feature of any contaminant of sewage sludge. These substances will be essentially chemically stable in the sewage matrix and not readily amenable to biodegradation during aerobic biological sewage treatment. Their concentration in the sewage sludge will be dependent upon their concentration in the influent raw sewage to the waste water treatment works and their affinity for settleable sewage and bacterial solids.

SUBSTANCES OF CONCERN

It is impossible to identify all the organic substances which could be of concern when present in sewage sludge. There are two principal reasons for this,

i) the extremely large, and ever increasing number of compounds which are potentially involved;

ii) the paucity of information about their adverse effects on the environment and in particular their chronic effects on humans.

It is possible to produce a list of toxic elements of concern because their numbers are finite and as a consequence their environmental effects and toxicology have been studied over an extensive period. However, because of the factors above, the state of knowledge about organics is very limited.

The United States production of organic chemicals has grown from 4.5 x 10^6 tonnes in 1943 to 64.0 x 10^6 tonnes in 1972 (2). The rate of growth has been exponential, increasing by approximately 9% *per annum*, resulting in production doubling every 8 years. Not only the quantity, but also the total number of chemicals produced is growing. During 1974 there were reported to be 12,000 chemical compounds in use in the United States (3). In the United Kingdom, it has been suggested that the number of chemicals currently available could be between 10,000 to 20,000 and increasing by approximately 1,000 *per annum* (4). It would appear to be inevitable, therefore, that a proportion of these chemicals will enter waste water treatment plants, either as the result of their use by the consumer, the discharge of industrial effluents, by surface run-off or by volatilization and subsequent atmospheric deposition.

Two lists of substances are widely accepted to include the principal compounds of concern, they are the European Economic Community list, both black and grey (5), and the United States Environmental Protection Agency Priority Pollutants list (6). The criteria used in the formulation of both lists are similar and it is, therefore, not surprising that many groups of substances are common to both. The USEPA list is however somewhat more detailed.

The EEC lists include substances or classes of substances which cause particular concern because they are known or suspected to adversely effect

a) human health and welfare,
b) aquatic life,
c) waste water treatment processes.

It should be noted that "in selecting priorities some consideration must be given to the likelihood of harmful substances reaching a living organism, whether animal or man in sufficient concentration for a sufficient period for harm to occur" (7).

In the United States compounds with adverse effects on humans and the natural environment were included in the Priority Pollutants list based on the following criteria,

a) frequency of occurrence of the compound in water,
b) the availability of chemical standards for the quantification of the material of concern,
c) the amount of production of the substance,
d) chemical stability of the substance.

In general these lists include the following classes of organic compounds, polychlorinated biphenyls(PCB), phenols, chlorinated hydrocarbon solvents, polynuclear aromatic hydrocarbons (PAH), organophosphorus compounds, organotin compounds, phthalate esters, petroleum hydrocarbons and organochlorine insecticides.

In addition to the above concern has been expressed on occasion about surfactants, detergent builders, polybrominated biphenyls, and non-phosphorus fire retardents (including pentabromotoluene).

SOURCES AND OCCURRENCE

In areas where industrial effluents are discharged to sewage treatment works, many synthetic organic compounds may be present in the raw sewage. Pollutants commonly occurring under such circumstances include aliphatic and aromatic hydrocarbons, petroleum hydrocarbons, benzenes, phenols, PAH, halogenated (particularly chlorinated) aliphatic, alicyclic and aromatic compounds, organochlorine pesticides, PCB and phthalate esters (8-16). Table 1 includes a summary of the major sources of these pollutants. In addition to industrial effluents as sources of organic micropollutants in waste water, it is evident that domestic sewage contributes a low-level background concentration of trace organic contaminants to raw sewage (1, 35, 36) either through usage or as the result of the contamination of products during manufacture.

A significant proportion of organic contaminants carried by sewers derives from urban run-off, consisting of stormwater from roads, motorways and paved areas. A great variety of organic compounds may be found in urban run-off, including aliphatic and aromatic hydrocarbons, PAH, fatty acids, ketones, phthalate ester plasticisers and other more polar compounds (1,11,21, 22). Solvent-extractable organics are dominated by petroleum hydrocarbons which are considered to arise mainly from motor fuels and oils, tyres and bitumen in road surfaces (1,22). This source of organic contaminants has been shown to be responsible for a significant petroleum hydrocarbon load to the environment. Run-off may contain mg l^{-1} concentrations of total hydrocarbons (21) and during heavy rainstorms, urban run-off may increase raw sewage PAH concentrations by up to 100-fold over dry weather conditions (11, 37). Leachates from solid waste dumps and landfill sites may contain a wide range of organic contaminants (38,39). While run-off from agricultural land contains significant concentrations of pesticides, although only a small

Table 1. Major sources of organic contaminants in waste water

Chemical class	Sources	Reference
Aliphatic and aromatic hydrocarbons (including benzenes and petroleum hydrocarbons).	Petrochemical industry wastes	17,18
	Heavy/fine chemicals industry wastes	19
	Industrial solvent wastes	19
	Plastics, resins, synthetic fibres rubbers and paints production	19
	Coke oven and coal gasification plant effluents	20
	Urban run-off	21,22
	Disposal of oil and lubricating wastes	17
Polynuclear aromatic hydrocarbons	Urban run-off	23
	Petrochemical industry wastes	24
	Various high temperature pyrolytic processes	11
	Bitumen production	25
	Electrolytic aluminium smelting	25
	Coal-tar coated distribution pipes	26
Halogenated aliphatic and aromatic hydrocarbons	Disinfection of water and waste water	3,27
	Heavy/fine chemicals industry wastes	28-31
	Industrial solvent wastes and dry cleaning wastes	28-30,32
	Plastics, resins, synthetic fibres, rubbers and paints production	28-32
	Heat-transfer agents	28,29,31
	Aerosol propellants	29,30
	Fumigants	29-31
Organochlorine pesticides	Agricultural run-off	33,34
	Domestic usage	35
	Pesticide production	36
	Carpet mothproofing	37-39
	Timber treatment	40
Polychlorinated biphenyls	Capacitor and transformer manufacture	41
	Disposal of hydraulic fluids and lubricants	42
	Waste carbonless copy paper recycling	42
	Heat transfer fluids	43
	Investment casting industries	41
	PCB production	44
Phthalate esters.	Plastics, resins, synthetic fibres, rubbers and paint production	32
	Heavy/fine chemicals industry wastes	32
	Synthetic polymer distribution pipes	1,45

proportion of the applied compounds may be lost in this manner (34,46,47). Under some circumstances both these sources may contribute to the contamination of sewage sludge although not to the same degree as urban run-off.

Rainwater has on numerous occasions been shown to contain many organic compounds including PAH, PCB, organochlorine insecticides, low molecular weight chlorinated hydrocarbons, plasticisers and industrial solvents (1,8, 11,27,48,49). It has been demonstrated that the average concentration of total organochlorine insecticides in the rainfall of the U.K. is about 150 ng l^{-1}, but ranges from 3 to 300 ng l^{-1} (50). Organic contaminants reach the atmosphere because of evaporation from sites on the earth's surface. The principal mode of entry to the environment of many organic compounds of high volatility such as industrial solvents is in fact by evaporative loss at sites of manufacture and use (27). Return to the earth's surface occurs either by particulate fall-out, rain, or vapour phase deposition (27). This contribution may be combined with the urban run-off and enter waste water treatment plants with combined sewerage systems.

REMOVAL OF CONTAMINANTS DURING WASTE WATER TREATMENT

Many organic substances of concern are non-polar and hence have a very low water solubility. Such compounds tend, therefore, to adsorb strongly on suspended particulate matter (51). This suggests that mechanical separation processes such as sedimentation will achieve substantial removal of these materials into the primary and secondary sludges. The limited information on the distribution of synthetic organic compounds in waste water treatment processes confirms this.

Many synthetic organic compounds, because of their non-polar and hydrophobic nature not only adsorb onto particulate matter, but also partition into non-polar fat and lipid material present in raw sewage. This component of the raw sewage including mineral oils, greases, waxes and surfactants, some of which in varying degrees are resistent to degradation, could potentially represent an important mechanism for the concentration and transport of these materials. However, this is difficult to assess, since very little research has been carried out in this area (48,52).

The limited information on the behaviour of organic micropollutants during waste water treatment is summarised in table 2. Whilst the reported removals exhibit a wide range, conventional two-stage sewage treatment would appear to remove 70 to 90% of these substances present in the influent

Table 2. Removal of Organic Pollutants in Waste Water Treatment Processes.

Compound	Process Type	Removal Efficiency %	Reference
Polynuclear Aromatic Hydrocarbons (PAH)	Primary sedimentation percolating filter	50	53
	Primary sedimentation percolating filter/ activated sludge	86	53
	Primary sedimentation/ activated sludge	99	53
Polychlorinated Biphenyls (PCB)	Primary sedimentation/ percolating filter	86	54
	Primary sedimentation/ activated sludge	71	13
	Primary sedimentation with various secondary and tertiary treatment processes	66	55
	Primary sedimentation	51	56
	Primary sedimentation pilot plant	58	57
Dieldrin	Primary sedimentation/ trickling filter	≤95	38
	Primary sedimentation	51	56
	Primary sedimentation/ equalisation lagoon	99	37
	Activated sludge pilot plant	0-63	58
Lindane	Primary sedimentation/ trickling filter	44	59
p,p'-DDE	Primary sedimentation	37	56
	Activated sludge pilot plant	0-79	58
Aldrin	Activated sludge pilot plant	0-41	58
2,4-D Alkyl Esters	Activated sludge pilot plant	0-100	58

sewage. Where data is available the removals during the primary sedimentation(mechanical) stage of conventional sewage treatment appear to be in the range of 40-60%.Removals during secondary biological treatment vary between 0 and 100% even for the same compound. The role which biodegradation may play in the removal of organic micropollutants has not been clearly established.

If a particular organic contaminant is present in raw sewage and it is not removed or only partially removed by primary sedimentation, then its subsequent removal could be significantly influenced by its amenability to biodegradation during aerobic biological sewage treatment. Solubility in water appears to be one of the factors limiting the biodegradation of sparingly soluble compounds such as PAH and PCB (48,60). The limiting factor for biodegradation appears to be the organic/water interface which is the major site of biodegradation of organic compounds (48,60).

PAH are biologically active molecules and are, therefore, able to enter into biochemical reactions. There is also evidence of their biosynthesis (61). There would appear to be no evidence for their biodegradation during secondary sewage treatment. However, bacteria have been isolated from soil which are able to aerobically biodegrade PAH (62,63);such bacteria are present in activated sludge. Significant biodegradation of the less chlorinated PCB in laboratory scale activated sludge simulations has been reported (64), but it is questionable whether this would occur in full-scale conventional plants. Industrial waste waters containing the organophosphorus insecticide parathion, have been successfully treated using modified biological sewage treatment, extended retention time, high biological solids and alkalinity control resulted in the complete biodegradation of parathion and its principal metabolite p-nitrophenol (65). The treatment of waste waters from the herbicide manufacturing industry has been reported after dilution with domestic sewage and oxidation in an aerated lagoon (66).

It is improbable that most organic micropollutants are biodegraded to a significant degree during conventional aerobic waste water treatment. Information about their possible inhibitory effects on the biological treatment process is apparently limited to 10 of the 114 USEPA priority pollutants, whilst information about effects on the sensitive nitrifying bacteria is limited to phenol and 2,4-dichlorophenol (6). Removal is principally by adsorption onto biological solids followed by sedimentation to form a secondary sludge. Thus when organic micropollutants are present in raw waste waters both primary and secondary sludges will be contaminated with them.