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ORGANIC POLLUTANTS IN MUNICIPAL SLUDGE – HEALTH RISKS

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ABSTRACT

The chemical industry during the last twenty-five years has demonstrated a steady growth. It is estimated that at least 700 new chemical compounds are manufactured each year, and the precise chemical formulation of these is well-guarded, classified information. More than two million chemicals have been listed for use, mostly during the last thirty years [1]. While these substances have had great benefit to the society, disposal of waste chemicals and their by-products, both before and after use, is a major problem. Cleansing agents, cosmetics (containing solvents), deodorizers, disinfectants (containing naphthalene, chlorophenols, etc.), paints, pesticides, and other toxic substances are discharged into the environment without discrimination. This variety of discharged chemicals creates a significant burden on the human population–either directly or indirectly via accumulation in the food chain. A wide spectrum of different compounds have been identified in water and wastewater, many of them are toxic to mammals; the compounds range from simple hydrocarbons to complex chlorinated polynuclear compounds. The objectives of this article are to study contamination of municipal sludge by refractory organics and discuss the potential health implications caused by them.

ORGANIC TOXICANTS IN SLUDGE

In the past, the domestic wastewater contained urine, feces, food wastes, paper products, and detergents. But these days, in addition to these usual contaminants, the wastewater contains refractory and high molecular weight organics.

Note: Any opinions or conclusions that are expressed in this article are those of the authors and do not necessarily reflect the official views and policies of the U.S. Environmental Protection Agency, nor does mention of trade names or commerical products constitute endorsement or recommendation for use.

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Sludges concentrate the organic and inorganic constituents of wastestreams. The U. S. Environmental Protection Agency (EPA) initiated a comprehensive study of priority pollutants, covering sixty different treatment plants [2]. Many organic compounds were measured. The health effects caused by these agents have been discussed by various authors [3-6]. Among halogenated compounds, vinyl chloride and chloroethane were 3.6 and 1.5 ppm respectively. Toluene was as high as 1.9 ppm, phthalate acid esters [bis(2-ethylhexyl) phthalate] was 3.9 ppm, phenanthrene and anthracene were 0.41 ppm each. Jacobs and Zabik extended the characterization of specific refractory organics in sludge [7]. Farrell and Salotto found organochloro-insecticides in varying amounts in sewage sludges, among these aldrin and chlordane were about 16 ppm (dry weight) and others ranged from 0.24 to 0.91 ppm [8]. In the Netherlands the dieldrin concentration in sludge was 7.5 ppm (dry weight) [9]. The excessive amount was probably contributed by a wool textile industry. PCBs in the Midwest ranged from 240 to 1,700 ppm, with an average of 765 ppm [10].

Jacobs and Zabik analyzed sewage sludges from 204 municipal treatment plants in Michigan [7]. An analytical search was made for seventy-three organic chemicals. At least fifty-three compounds were detected in < 1 percent to 95 percent of the sludges analyzed. To illustrate, a total of 236 samples were tested for biphenyls, 33 percent of the samples were positive, the concentration varied from 0.04 to 1,730 mg/kg (ppm) dry weight, with an average of 89.6 mg/kg. The average concentration of hexachlorobenzene was 468 mg/kg. The concentration of various phthalates varied from 552 to 1,250 mg/kg. Approximately 84 percent of the samples tested contained bis(2-ethylhexyl)phthalate the concentration varied from 0.4 to 58,300 mg/kg.

Erickson and Pellizari identified many chlorinated compounds in sewage sludges [11]. Nicholls *et al.* detected polynuclear hydrocarbon in municipal sewage sludge in England [12]. Researchers also found these compounds in sludges in Czechoslovakia and Germany [13, 14].

The Municipality of Metropolitan Seattle (Metro) undertook a systematic program to monitor the characteristics of its municipal sludges [15]. The analytical tests were made for eleven organic chemicals. Of the eleven compounds sought, only PCBs were found in detectable amounts. Most of the time the concentration of PCBs was less than 2 mg/kg, dry weight. This is very low as compared to the Midwest study [10], because the Seattle municipal treatment system does not receive highly industrialized waste streams.

The profile of organic and inorganic constituents in sludge is influenced by industrial wastestreams. The Muskegon Study particularly points out this fact [16]. The sludge had 44 mg/kg of 3,3-dichlorobenzidine–a suspected carcinogen–which was probably discharged from a dye factory. This was not detected in other studies.

Naylor and Loehr tabulated twenty-four organic toxicants predominantly

found in sewage sludges [17]. These compounds were detected in several plants and had a medium concentration of 0.2 mg/l (wet weight) or greater.

Giger *et al.* recently reported that anaerobically treated sewage sludge contained extraordinarily high concentration (0.45 to 2.53 g/kg) of 4-nonylphenol (NP), a metabolite of nonionic surfactants of the nonylphenol polyethyoxylate type [18]. Its concentrations in aerobically treated sludge varied from 0.08 to 0.5 g/kg. The concentrations (4-nonylphenol) in sewage sludge is higher than contaminants such as PCBs (0.35 to 23.0 mg/kg), and polycyclic aromatic hydrocarbons (20 to 30 mg/kg). 4-nonylphenol is not among 129 priority pollutants. Varma *et al.* have measured NP in raw sewage and sludge [19].

The review of literature presented here shows that organic toxicants have been repeatedly detected in municipal sludges. The concentrations and amount of each chemical varied. Apparently, they depend on the type of unit process employed at the treatment plant, as well as the diversity and hydraulic flow from industrial wastestreams. However, the presence of refractory organics in sludge suggests that sludges used for land application should be routinely monitored for hazardous substances.

SURROGATE POLLUTANTS

A wide array of organic compounds have been found in municipal sludges. Some of these are routinely identified and quantified, while there are many more unknown chemicals for which no search is made.

It may be impossible to completely analyze the chemistry of municipal sludges. Considering the diversity of the chemicals, a "short list" of key chemicals to serve as surrogate indicators of persistent chemicals is presented in Table 1. One should keep in mind that no list can be complete and applicable in all cases, because the chemical profile of sludges is influenced by industrial discharges, and there are site-specific chemicals to be considered. The rationale used for the selection of surrogate chemicals is described below.

Polyaromatic hydrocarbons (PAHs) are organic compounds composed of two or more fused rings and are well documented carcinogens. Benzo(a)pyrene [B(a)P], phenanthrene and benzo(a)anthracene, are the most studied PAHs. PAHs are formed during incomplete combustion, and are often regarded as anthropogenic environmental contaminants. PAHs enter into the aquatic environment through many pathways including industrial wastestreams.

B(a)P produces dose response tumorigenesis in mammals. The acute LD_{50} dose in rats when administered subcutaneously is 50 mg/kg. The lowest recorded letal dose (LD_{LO}) for benzo(a)anthracene is 10 mg/kg. The acute LD_{50} dose of phenanthrene is 700 mg/kg. Recently, the World Health Organization (WHO) proposed a limit of 0.01 mg/l for B(a)P in drinking water. The U. S. EPA has no specific limit for B(a)P in drinking water.

Table 1. List of Hazardous Pollutants for Routine Monitoring

Description	*Water µg/l Range of Conc.*	*Tumorigenesis TD_{Lo}[a]*	*Chronic Health Effects*	*Aquatic Toxicity*	*Acute Oral Rat Toxicity LD_{50} mg/kg[a]*	*Sludge Conc. mg/kg Dry Weight[b]*	*FDA Market Basket Study mg/kg Body Weight*
Benzene	1-953	52 gm/kg/52W-I	Carcinogenicity Leukemia	LC_{50} 5,100 µg/l	4894	0.53-7.1	
Benzo(a)pyrene	1-490	160 mg/kg/6dic	Carcinogenicity	—	SUB. 50 ipr. LD_{Lo}=500 (mouse)	0-82 µg/l	
Benzo(a)anthracene	1-1,500	Sub 2mg/kg	Carcinogenicity	87% mort. in bluegill at 1,000 µg/l	inb. mouse LD_{Lo}=10		
Bis(2-ethylhexyl)-phthalate	47,000	216 gm/kg/2yC	Liver Cancer	3 ug/l(Daphnia Magna)	31,000	4.1-273	
Chloroform	1-366	70 gm/kg/78w-c	Carcinogenicity	1240 mg/l	1194	0.00118-564	
DDE (Metabolite of DDT)	—	9700 mg/kg/78 w-c	Bioaccumulation in tissue— Carcinogenicity	1050 µg/l	880		0.0607
Heptachlor	—	403 mg/kg/80w-c	Neurotoxic Teratogenic	0.0038 µg/l	40		0.0077
Hexachlorobutadiene	—	15 gm/kg/2y-c	—	—	90	0.52-8	0.0024
Lindane	—	25 gm/kg/73w-c (Mouse)	Carcinogenicity	0.080 µg/l	76	0.00059-12.51	
Methylene Chloride	1-10,500	—	Neurotoxic	LC_{50} 193 mg/l	2524	0.06-30	
4-Nonylphenol	—	—	—	EC_{50} (fish) 0.2 mg/l	1620	80-2,530[18]	
Phenanthrene	1-10,000	skin mouse 7 mg/kg	Carcinogenicity	600 µg/l for polychaete worm	700 (mouse)	0.89-44	
PCB's	—	—	Carcinogenicity	0.014 µg/l	Varies 325 +	0.435-2390	
Toxaphene	—	30 gm/kg/80n-c	Carcinogenicity Liver Damage	0.013 µg/l	40		0.1071
Tetrachlorethylene	1-2800	195 gm/kg/50W-I	Carcinogenicity	40 mg/l	8850	0.024-42	
Vinyl Chloride	8-62,000	10 gm/kg/52W-I	Liver Angiosarcoma Carcinogenicity	—	500	3-110	

[a] See [37].
[b] See [17].

Abbreviation Legend: Sub. = Subcutaneous; ipr. = Intra peritoneal; inv. = Intravenous; w = Weeks; c = Continuous; I = Intermittent; LD_{Lo} = Lowest Recorded Lethal Dose.

Chloroform is formed in sewage effluents by the interaction of aqueous chlorine with ubiquitous precursors, including industrial wastes. The acute LD_{50} dose for chloroform in rats is 31,000 mg/kg. It has been shown that chloroform is a carcinogen-mutagen.

DDT was banned in the United States in 1975. Its biological half life (t1/2) in soil is about seven to ten years. DDE is one of its metabolic residues, hence it is included in Table 1. Also listed in this table are several other toxic insecticides-pesticides such as, heptachlor and lindane. The acute LD_{50} doses are 40, and 76 mg/kg, respectively.

Acute oral toxicity of these compounds is listed in the table. More of these are proven carcinogens. Listed in the table are also the lowest published toxic doses (TD_{LO}) for induction of tumorigenesis.

UPTAKES OF ORGANICS BY PLANTS

If sludges are used for nutritive application on land, the specific organic compounds in high concentration may be phytotoxic. Kenagal, in a review of data on more than 130,000 chemicals, found only 0.17 percent chemicals that killed seeds or seedlings at a concentration of 0.1 to 0.99 ppm [20]. However, at lower concentrations, the toxicants will biomagnify in crops. The rate of bioavailability of refractory organics depends upon the chemical composition of the compound, its concentration and vegetative species grown. The translocation of organic chemicals from the sludge-soil matrix has not been studied extensively. In general, lipophilic, non-polar and non-volatile compounds are absorbed by the soil and by the roots of the plants. Small polar molecules penetrate through the roots and are translocated into the plant [20].

Several studies have shown that peak accumulation of organics occurred in the first few days after sludge application followed by dramatic reduction after ten to fifteen days [21-24]. Hexachlorobenzene (HCB) is used as a seed disinfectant and soil conditioner, as well as a plasticizer in PVC. It is absorbed from soil by various types of plants, accumulates in fatty tissues, and is not readily biodegraded. It has been detected in carrots, lettuce, grass, and hay. HCB is not very toxic in rats. The LD_{50} varies between 1,000 to 3,000 mg/kg.

Land application of sewage sludge containing trace levels of PCBs did not yield detectable levels in plant samples [25], but levels ranging from 50 to 100 ppm dry soil resulted in 3 to 50 percent soil concentration in carrots [26]. Maximum concentration of PCBs (97%) was found in carrot peels, hence insignificant amounts were in the plants *per se.* Similarly, Chou *et al.* concluded that polybrominated biphenyls (PBBs) are not significantly translocated by plants [27].

The uptake of HCB by lettuce was studied [28]. The concentration of HCB in soil varied from 0.007 to 0.16 ppm. The residues in lettuce ranged from 0.005 to 0.12 ppm. The allowable tolerance level in food in West Germany is

Table 2. Uptake of Pesticides by Various Plants

	Concentration – ppm			
Residue	*Roots*	*Grains*	*Soil*	*Crop*
Aldrin	0.29	< 0.01	0.83	Maize
	0.58	0.36	0.59	Potato
Dieldrin	4.16	< 0.01	0.63	Maize
	2.26	1.16	0.40	Potato

0.005 ppm. In another study, HCB was 0.035 ppm in lettuce [29]. It was lower in cress and parsley. Many of the uses of aldrin and dieldrin have been banned in the United States, however it is still used for termite control, dipping of non-food roots and moth proofing. Some of the typical earlier studies are summarized in Table 2. The purpose of this table is to illustrate that organics are biomagnified in the food chain. The table also illustrates that the concentration of the compound(s) is different in various parts of the plants. Chlordane has also been measured in different plants. In carrots it was as high as 1.53 ppm [30].

Demirijian *et al.* studied the fate of organic priority pollutants in sludge and wastewater irrigation [31]. Municipal sludge from Muskegon, Michigan, was applied at a rate of 40,333 kg/ha (dry weight). At the end of the study most of the organic compounds were not detectable in the soil. They were probably degraded by the microorganisms. Only 2,2-dichloroazobenzene was detected. This was perhaps an intermediate metabolite of 2-chloroaniline. Also, corn grain grown on a test area did not indicate a substantial build-up of organic chemicals, when compared to a background area.

In general, root crops tend to accumulate higher concentrations of organic compounds than leafy vegetables, and B(a)P concentrations are higher in radish roots than in foliage. This study shows that organic compounds are not readily translocated in plants [32].

The Food and Drug Administration (FDA) has been analyzing food for heavy metals and organic chemicals. A typical "market basket" study represents the recommended two week diet of an adult person. All food items are divided into twelve composite groups for analysis. In 1978, a total of 11,357 residues, represented by sixty-one different pesticides, metal, herbicides, fungicides, and industrial chemicals, were reported [33]. Of these, thirty-six chemicals ranged from 0.0007 μg/kg (PCNB) to 0.1437 μg/kg (chlorpropham). The cumulative intake of organic chemicals was 47.35 μg/day [33]. Based upon the limited data available it is fair to assume that the health risks of organic toxicants can be avoided or at least minimized by routine monitoring of municipal sludges and plants grown via land application.

ANIMAL FEED STUDIES

It is known that cattle ingest soil as a part of grazing. It is estimated that in certain cases animals may consume soils up to 10 percent of the dry matter consumed. Surface soil may be transferred to baled hay by machines.

Decker *et al.* found significant amounts (2 to 6.5%) of compost in feces of cattle [34]. Studies show a significant increase of toxicants in milk and fatty tissues of grazing cows. Baxter *et al.*, experimenting with varying ratios of sewage sludge, found that fatty tissues were the only tissue that showed a significant increase of refractory organics [35]. However, the authors concluded that "if sewage sludge is incorporated into soil and utilized as an agricultural fertilizer, there would appear to be little hazard that heavy metals or persistent organics would be elevated in cattle tissues above levels that would be considered normal."

It is estimated that a cow, in extreme cases, may consume soil up to 500 kg/year (1.4 kg/day). If sludge contains 109 mg/kg of bis(2-ethylhexyl)-phthalate, and it is mixed with 15" top soil (2×10^6 kf/ha) at an application rate of 10 tons/ha. This will provide an average concentration of about 1.09 mg/kg of the compound. Hence, the cow will consume 545 mg per year, and in a normal span of five to ten years the maximum consumption (in ten years) will be about 5.4 grams. The LD_{50} for a rat is 3,100 mg/kg, therefore, the LD_{50} for a cow weighing 500 kg is 15.5 kg.[1] Hence, a cow can consume not more than 0.017 percent of LD_{50} dose in its entire life via soil eating or conversely it will take 5100 life spans to ingest an LD_{50} dose [36, 37].

Usually 10 percent of the ingested amount is retained in the system, but a recent study conducted on seven different refractory organics showed that the residual level at the end of a ninety-four day feeding trial, varied from less than 1 percent to 75 percent [38]. Assuming 40 percent retention, the cow will accumulate a total of 2.1 grams in a normal life span of ten years.

In recent years, there have been voluminous amounts of research on the health effects on animals of toxic materials in sludge. The animals included ranged from earthworms to cattle, however, most of these studies dealt with heavy metal composition in various organs. The heavy metal mostly referred to was cadmium. Haschek *et al.* fed sheep on cabbage grown on digested sewage sludge containing PCBs [39]. They found a correlation between elevated levels of PCBs in sheep's liver and sludge PCB contents. In addition, animals exhibited proliferation of the smooth endoplasmic reticulum and some degenerative liver changes. The way liver metabolizes PCBs and other refractory organics has been presented by various authors [40-43].

[1] The extrapolation has been made assuming a linear interspecies relationship. Risk assessment of human health from exposure to a complex mixture should be made on a proper interspecies conversion factor.

Refractory organics differ in half-life retention in animals. The PCB half-life in cows varies from forty to seventy days and milk contained a constant rate of PCBs and other chemicals compared to body fat burden. Serious injuries resulted to primates when mothers were fed on diet containing 2.5 ppm of PCBs during gestation and lactation [44].

Hensen fed young swine for fifty-six days on corn grown on sludge-fertilized land [45]. The results were essentially negative except elevated levels of hepatic microsomal mixed function oxidase (MFO) activity. Other liver enzymes were normal. It was theorized that probably the increased MFO activity was caused by toxic compounds. Telford et al. obtained similar results when sheep were fed corn grown on a soil-sludge matrix [46]. The sludge was used at 224 tons/ha. On the other hand, Lisk et al. did not find elevated MFO activity in swine [47]. Kienholz found that cabbage grown on sludge-amended soil caused degenerative changes in liver and thyroid of sheep [48]. It has also been suggested that many insecticides, pesticides such as DDT, its metabolite DDE, and PAHs may possess weak estrogenic activity [49]. Estrogens are hormones which promote normal and abnormal secondary sex characteristics and organs.

MUTAGENICITY-CARCINOGENICITY

It has become increasingly clear that mutagens and carcinogens are released in the environment. The feces of certain animals, the dog, cow, horse, and sheep, are known to cause chromosomal breakage, indicating that animals are discharging mutagens in their feces. Van Tassel *et al.* contend that mutagenicity of substances in feces may be increased by anaerobisis and the presence of bile and bile acids [50].

Hopke and Plewa determined the mutagenesis of five different municipal sludges in Illinois [51]. The authors used different extraction techniques, and tested each sludge using a variety of indicator organisms. Mutagenic activity was observed in the sludges from Chicago and Sauget. The sludges from primarily domestic sewage did not contain mutagens. Furthermore, *in situ* tests of sludge-amended soil (Chicago sludge) using the maize *wx* locus assay showed that "mutagens are available to the plant when sludge is used for soil amendment." Mutagenic activity was also observed in the secondary effluents in Illinois [52]. The maximum activity was 4,500-fold.

Recently, Theis et al. detected weak mutagenic activity using the Ames assay on incinerated municipal sludges from several sources [53]. Four of ten ash samples tested were positive. In addition, one sample of dewatered sludge provided positive results. Parallel tests on non-incinerated samples were not performed. Chemical analysis showed presence of heavy metals, but refractory organics were not analyzed. Babish *et al.* tested dichloromethane extracts of sludge samples from thirty-four cities for mutagenicity using the Ames test [54]. Of the thirty-four samples, thirty-three showed a dose related increase in revertants in at least one of the five tester strains.

RISK ASSESSMENT

One of the problems in evaluating municipal sludges is to determine the level at which the sludge may be labeled as toxic to animals including man. This requires the translation of laboratory data on health effects to quantitative risk assessment. Risk assessment eventually leads to the formulation of regulations.

In multi-chemical complex mixture exposure some of the prime factors are:

1. the joint effect of two or more chemicals;
2. multiple health effects;
3. interspecies conversion coefficients; and
4. accountability of variations in human dietary habits, and mode of exposure.

In co-toxicity the question is not only whether the chemicals administered jointly caused more adverse health responses than each chemical administered singly. The question is also whether the response of one chemical is altered in any way (increased or decreased) by the presence of other chemicals. Also, it is possible that small doses of each fraction may be safe, but research has yet to determine if the sum total effect of multiple chemicals administered jointly is safe. Some chemicals may cause more than one type of adverse health response. Research on this model presumes that multi-chemical exposure may not be arithmetically additive, and it also attempts to improve on models employing the linear extrapolation of the results.

CONCLUSIONS

Municipal sludges can be contaminated by a wide variety of toxic organic compounds, which pose a threat to human health. Contaminated sludge can complicate human health by one or more of the following pathways:

- translocation of chemicals in plants;
- direct consumption of sludge-amended soil; and/or
- bioaccumulation in the food chain.

The movement and translocation of organic compounds is a complex phenomenon. Past studies indicate that generally root crops have higher concentration than leaves and fruits. The plant to soil weight ratios of pollutants, in most cases, was less than one. This shows that bioaccumulation is not significant. However, the data needs further verification on more chemicals before deriving a firm and binding conclusion.

Scientists seem to think that direct consumption of the soil by the animals can add measurable amounts of pollutants to the biological system. Although model estimates of soil intake show potential for minimal human exposure of organic pollutants, research data indicates the need for more study.

Mutagenicity studies have indicated the presence of mutagens in sludge. These studies can, at best, describe the toxic potencies of the sludges, and categorize them as high, low, or medium depending upon mutants per unit dose as compared to background levels. Such results are of semi-quantitative nature. Based upon these studies, conclusions *cannot* be made on whether land application of sludge is significant in including mutagenicity among humans.

To evaluate the health risk to humans, the following questions need answers.

- Is there a maximum acceptable mutagenicity of sludge for land application?
- What types of plants are most significant in translocating mutagens and carcinogens?
- Are sludge-grown plants genotoxic?
- At what rate do mutagens degrade in soil?

To fully study the biological effects from exposure to a complex mixture of chemicals, it is necessary to isolate and concentrate the toxic fraction(s) by chemical methodology before performing biological assays. The fractions and subfractions need chemical characterization. Several fractionation analytical techniques are available. Each has its strength and weakness but all the techniques have not been studied under parallel conditions in the laboratory to provide comparative data. It is not known what compounds, if any, are lost during the extraction procedure. Chemical profiles must be obtained before and after fractionation. By providing more information on the identity of toxicants, their frequency of occurrence, including removal and/or formation during treatment processes will greatly help in determining the acceptability of sludge for land application and selecting an environmentally acceptable method of disposal. The following research in this area is needed:

- Which fractionation technique is best?
- Which types of chemical compounds are lost in each fractionation method?
- Which fractions are synergistic or additive?
- Which bioassay(s) should be used for screening?

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