

## Public Health Concerns About Chemical Constituents in Treated Wastewater and Sludge

There are many chemical constituents that enter the municipal waste stream that are of potential concern for human health. These substances include organic chemicals, inorganic trace elements (such as cadmium and lead), and nitrogen. Conventional agricultural practices, such as the use of commercial fertilizers, also have the potential to introduce additional chemical constituents to soil. However, this report is not attempting a comparison between public health effects of conventional agricultural inputs and those derived from municipal wastewater or sludge. The degree to which constituents from municipal wastewater present a risk to human health depend on their concentration in reclaimed water and treated sludge and the fate and transfer of these chemicals from the wastewater/sludge sources to human receptors via various exposure pathways. The chemical composition of sewage and the degree to which chemical concentrations are reduced in effluent and sludge by secondary and advanced treatment were discussed in Chapters 2 and 3. The degree to which chemical concentrations of these contaminants in reclaimed wastewater and sludge are further reduced through natural processes in the environment and their availability to food plants were the subjects of Chapter 4. Trans-mission of toxic contaminants to humans from agricultural use of reclaimed wastewater and sludge is covered in this chapter.

The U.S. Environmental Protection Agency (EPA) identified Priority Pollutants in regulations that deal with municipal and industrial wastewater (EPA, 1984) due to their toxicity to humans and the aquatic environment. These Priority Pollutants are divided into four classes: (1) heavy metals (oftentimes referred to as trace elements or trace metals) and cyanide, (2) volatile organic compounds, (3) semivolatile organic compounds, and (4) pesticides and poly-chlorinated biphenyls (PCBs). In addition, nontoxic organic compounds in wastewater can be transformed into potentially toxic chlorinated organic compounds, such as trihalomethanes, when chlorine is used for disinfection purposes (National Research Council, 1980).

For the purpose of this review, non-metallic trace elements, such as selenium, are grouped together with the heavy metals under the more general designation of "trace elements." The following discussion first considers the fate of organic compounds from sludge and wastewater applications to soil. The uptake of these chemicals into plants and animals that go into the human food chain is then examined. Finally, the potential for adverse health effects from trace elements in sludge and wastewater via these same pathways is evaluated.

## FATE OF AND EXPOSURE TO ORGANIC CHEMICALS

Principal pathways of human exposure to sludge- and effluent-borne toxic organic compounds from land application to cropland include (Dean and Suess, 1985):

- Uptake by plant roots, transfer to edible portions of plants, and consumption by humans.
- Direct contact of edible plant parts with sludge or reclaimed water applied by spraying, and consequent consumption by humans.
- Direct contact by children who play on sludge/wastewater-treated soil and inadvertently ingest small amounts of soil.
- Uptake by plants used as animal feed, animal ingestion causing transfer to animal food products, and consumption of the animal food products by humans.
- Direct ingestion of soil and/or sludge by grazing animals, transfer to animal food products, and consumption of animal food products by humans.

Human exposure to toxic organic chemicals through incidental ingestion of sludge, effluents, or treated soil (pathways 2 and 3 above) is not considered in detail in this report. Chapter 7 discusses EPA's risk analysis, which evaluated all exposure pathways.

### Behavior of Toxic Organics in the Soil

It has been suggested that most toxic organic compounds are present in sludge at concentrations less than 10.0 mg/kg (Jacobs et al., 1987). Therefore, when sludges are applied to soil at agronomic rates and mixed with the surface, concentrations of toxic organics within the top 15 cm of soil normally will not exceed 0.10 mg/kg. In one survey, the level of toxic organics in sludge-amended soils was considered to be similar to or lower than background pesticide soil concentrations of 0.01 to 1.0 mg/kg (Naylor and Loehr 1982). They are further reduced by microbial decomposition.

In theory, there are a number of environmental processes that, when added to the soil, can interrupt the entry of toxic organic chemicals into the food chain. Organic chemicals from wastewater or sewage sludge may be destroyed directly after land application by biodegradation and chemical- and photo-oxidation. Organic compounds may also be volatilized, immobilized onto solid particles by sorption processes, or transported (leached) unaltered through the soil column to reach the ground water. In more complex mechanisms, sorbed organics may subsequently be chemically or photochemically degraded, microbially decomposed, or desorbed.

A considerable body of research has been performed on the behavior of organic pesticides in soil. Both laboratory and field experiments suggest that during land treatment, most pesticide residues are adsorbed by soil particles and remain sorbed on surface soils until degraded by microorganisms or volatilized (Cork and Krueger, 1991). The relative degree of intrinsic bio-degradability of toxic organics on the EPA Priority Pollutants list was illustrated by Tabak et al. (1981) in laboratory studies. They collected data on biodegradability and microbial acclimation of 96 compounds using bacterial inoculum from domestic wastewater and synthetic bacterial growth medium. Their overall results are summarized in Table 6.1. Significant bio-degradation was found for phenolic compounds, phthalate esters, naphthalenes, and nitrogenous organics; variable results were found for monocyclic and

polycyclic aromatic compounds, PCBs, halogenated ethers, and halogenated aliphatics; and no significant biodegradation was found for organochlorine pesticides.

Extrapolation of laboratory findings like these to predicting behavior in the field has two important constraints: (1) the rate of biodegradation in soil is probably less than that in laboratory medium and (2) the low concentration of the organics in reclaimed wastewater applied to crops may not support the level of microbial activity necessary for biodegradation in the soil environment.

Literature on the microbial decomposition of toxic organics in soil is diverse (Alexander, 1994). The degradation of petroleum hydrocarbons (a mixture of aliphatic, aromatic, and asphaltic compounds) in soils has been reviewed by Atlas (1981). Factors which appear to be important in encouraging high decomposition rates of petroleum hydrocarbons are temperature, concentrations, adequate supply of essential nutrients, and availability of oxygen. There is little evidence for significant leaching of petroleum organic compounds from the upper soil layers. Experiments with the high-rate application of sludge containing high levels of petroleum hydrocarbons onto land have shown a 77 percent degradation rate near the surface after one year, with most of the degraded compounds being n-alkanes (Lin, 1980). It was concluded that sludge land disposal would not result in petroleum hydrocarbon buildup in the soil. Also, in studies of other organic substances in wastewaters used for irrigation, Dodolina et al. (1976) found that acetaldehyde, crotonaldehyde, benzaldehyde, cyclohexanone, cyclohexanol, and di-chloroethane disappeared from soil within ten days.

The behavior of PCBs in soil has been comprehensively reviewed by Griffin and Chian (1980), who concluded that PCBs are strongly adsorbed by soil particulates. Influential factors affecting adsorption are the nature of the soil surface, the soil organic matter content, and the chlorine content and/or hydrophobicity of the individual PCB isomers. Adsorption increases with increasing organic matter content of the soil, with increasing chlorine content, and with increasing hydrophobicity of the PCB molecules. One study that examined percolation of PCB-containing solution through soil columns showed that less than 0.05 percent of one isomer was leached in the worst case. Fairbanks and O'Connor (1984) have shown that PCBs remain tightly adsorbed to sludge-amended soil, with minimal transport by soil water.

Studies involving land treatment of municipal wastewater provide some insight into the fate of toxic organic compounds from wastewater in soils. Land treatment of wastewater is considered to be an alternative form of secondary treatment, and is not performed for crop irrigation purposes. Generally, land treatment systems apply settled wastewater (primary effluent) to vegetated slopes using sprinklers or perforated pipes. The water flows over the sloped surface to collection ditches at the bottom of the slope where the effluent is discharged to a surface water. Biodegradable organic compounds, such as organic nitrogen and ammonia, are oxidized by soil bacteria. Percolation into the soil is negligible. Overland flow treatment slopes are selected for their relative impermeability; also, particulate material tends to seal the soil rapidly. Some water evaporates, but most of the wastewater is collected as surface runoff (Metcalf and Eddy, 1991). Wastewater flows in a very thin sheet across the vegetated slope. The U.S. Army Corps of Engineers Cold Regions Research and Engineering Laboratory in New

TABLE 6.1 Biodegradability of Priority Pollutant Organic Compounds

Test Compound Class	Number of compounds tested	Percent Degradation <sup>a</sup>
Phenols	11	82 D
Phthalate Esters	6	67 D
Naphthalenes	4	100 D
Monocyclic Aromatics	12	42 D, 50 T
Polycyclic Aromatics	7	50 A, 50 N
Polychlorinated Biphenyls (PCBs)	7	71 N
Halogenated Ethers	6	50 N, 50 D
Nitrogenous Organics	6	67 D, 33 N
Halogenated Aliphatics	23	26 D, 57 A or B, 17 C or N
Organochlorine Pesticides	17	100 N

<sup>a</sup>D-Significant degradation with rapid adaptation; A-significant degradation with gradual adaptation; T-significant degradation with gradual adaptation followed by a deadaptive process (toxicity); B-slow to moderate biodegradative activity, concomitant with significant rate of volatilization; C-very slow biodegradative activity, with long adaptation period needed; N-not significantly degraded under the conditions of test method.

SOURCE: Condensed from Tabak et al., 1981.

Hampshire operated a prototype overland flow wastewater land treatment system and found greater than 94 percent removal of each of 13 trace organics by volatilization and adsorption processes (Jenkins et al. 1983), with removal efficiencies decreasing as application rates increased and temperature decreased. With the possible exception of PCBs, biodegradation prevented contaminant buildup in the surface soil.

### Uptake of Toxic Organics by Plants

The following discussion summarizes information presented by O'Connor et al. (1991) from a recent review of the literature dealing with plant uptake of toxic organics. The results reported in the O'Connor et al. review are a mixture of laboratory and field data on plant uptake of organic compounds, both from sludge and from additions of pure chemical.

#### *Phthalate Esters*

Phthalate esters, which are the most common toxic organic compounds in sludges, present little risk because plants serve as effective detoxifying barriers to these chemicals and prevent their accumulation in the food chain (Aranda et al. 1989). Also, phthalates added to soils do not persist and are rapidly removed by volatilization and microbial decomposition (Dorney et al., 1985).

*Polynuclear Aromatic Hydrocarbons (PAHs),*

PAHs (also called "polycyclic aromatics") are produced by incomplete combustion. They are among the most common toxic organics in sludges (EPA, 1990). In the National Sewage Sludge Survey (NSSS), the PAH benzo(a)pyrene was found just at detection levels in 3 percent of the sludges analyzed from 209 municipal treatment plants. An earlier sludge survey of 40 municipal treatment plants conducted in the late 1970s by the EPA (EPA, 1982) found ben-zo(a)pyrene in 21 percent of the sludges with a mean level of 0.1 mg/kg (Kuchenrither and Carr, 1991). Some PAHs, particularly higher molecular weight species, are long-lived in soils (Bossert and Bartha, 1986). However, in long-term field studies (20-30 yrs), no evidence was found of elevated PAH concentrations in the above-ground portions of several crop species grown in PAH-contaminated soils. Air-borne sources of PAHs were regarded as the main origin of plant contamination in sludge-amended and control treatments alike (Witte et al., 1988; Kampe 1989; Wild et al., 1990). Overall, O'Connor et al. (1991) concluded that minimal PAH contamination occurred when sludge was used prudently for agriculture. The transfer of PAHs from soil was minimal for root crops, and essentially zero for above-ground crops.

*Polychlorinated Biphenyls (PCBs).*

Two studies found the median total PCB content in municipal sludges to be less than 0.5 mg/kg total PCBs (Jacobs et al. 1987; Mumma et al. 1988). The NSSS confirms this finding (EPA, 1990); it was found in the survey that the median concentration was 0.2 mg/kg. More highly chlorinated PCB congeners, which dominate in municipal sludges, tend to be more persistent, more strongly sorbed, less volatile and less bioavailable than the less chlorinated PCB species. O'Connor et al. (1990) conducted field studies and greenhouse pot studies using sludge-borne PCBs and found that the PCB levels in crops were below Limits of Detection (LOD), typically 0.20 µg/kg; bioconcentration factors were usually less than 0.02 based on dry weights of soil and crop and LOD. There was no statistically significant evidence of PCB uptake from sludge-borne PCBs in soils by aboveground parts of plants. Carrots were the only crop to contain detectable residues of PCBs, mostly in the carrot peel, which the authors noted can be easily removed with normal culinary practices of washing and peeling. No PCB vapor contamination of crops grown in soils amended with sludge-borne PCBs was detected.

Surface incorporation of PCB-contaminated sludges reduces PCB volatilization (Strek et al. 1981). Low inputs of PCBs to soil-plant systems, combined with low bioavailability, suggest negligible impact on plants grown in sludge-amended soils (Webber et al., 1994; O'Connor et al., 1991; Chaney, 1993). Witte et al. (1988) reported that repeated applications of municipal sludge over a period of 30 years and totaling 130 metric tons per ha, resulted in slight increases in soil PCB concentration, but no detectable (LOD=0.05 µg/kg dry weight) PCB residue in a variety of crops, including carrot. In another study, trace levels of PCBs from municipal sludge that was applied at a rate of about 2 metric tons/ha for two years to an old field resulted in no detectable PCBs in plant samples (Davis et al. 1981).

Adding PCBs (Arochlor 1254) to soils to produce concentrations from 50-100 mg PCBs/kg dry soil resulted in substantial uptake by carrot root with very little translocation to the aboveground plant parts (Iwata, et al., 1974). Computations from the data presented showed PCB concentrations in

carrot root ranging from 2.7 to 15.3 mg/kg with concentrations decreasing as the degree of PCB chlorination increased. Ninety-seven percent of the PCBs in the carrot root were in the peel. Although the above study showed substantial uptake by carrot root, it is unlikely that concentrations of this order of magnitude would ever occur in sludge-amended soils because regulations prohibit concentrations of PCBs in sludge greater than 50 mg/kg (EPA 1993a). Applying a sludge containing 50 mg PCBs/kg to a typical soil at a rate of 10 tons/ha would produce a concentration of 0.25 mg PCB/kg soil.

The less-chlorinated PCBs have greater potential to be taken up by the plant, but these are also much more volatile and biodegradable, and thus are less common in sludge. The more highly chlorinated PCBs are absorbed less by plants (Fries and Marrow 1981). Lee et al. (1980) were unable to detect PCBs in carrots grown in soil containing 0.23 mg PCB/kg of soil derived from an application of 224 metric tons/ha of sludge containing 0.93 mg PCB/kg of sludge. Naylor and Mondy (1984) have obtained similar results with potatoes. Over 1,400 samples of soil and crop tissue were analyzed in a greenhouse and field study for the Madison Metropolitan Sewage District (Gan and Berthoex, 1994). No PCB translocation into either corn grain or corn stover samples occurred. It was concluded that little, if any, PCB will be translocated from contaminated soil to plant tops. Apparently, PCB exposure via the plant/soil pathway is mini-mal.

Webber et al. (1994) determined concentrations of PCBs in corn, cabbage and carrots grown on coal refuse amended with sewage sludge at rates of 785; 1,570; and 3,360 tons per ha. Concentrations of PCBs in the treated coal refuse ranged from 1.3 to 3.7 mg/kg and were not related to treatment. The PCB concentrations in all tissues were less than 0.3 mg/kg; concentrations in carrot peels were greatest, followed in decreasing order by carrot tops; cabbage wrapper, inner leaves, and carrot core; corn ear leaf, stover, and corn grain. In general, the concentrations of PCBs in the plant tissues were independent of the PCB concentration in soil. Webber et al. (1994), Witte et al. (1988), and O'Connor et al. (1991), conclude that PCBs in municipal sludges represent no significant risk to crops or crop consumers.

Although most reports show PCB uptake by crops grown on sludge-amended soil to be negligible, Baker et al. (1980) found that PCB concentration in vegetables grown on garden soils were significantly higher than that of the control soil. However, concentrations of PCBs in the sludge used for that study averaged 479 mg/kg and would be banned from land application by present-day federal regulations.

### *Chlorinated Pesticides*

Some important chlorinated pesticides include aldrin, chlordane, DDD, DDE, DDT, endrin, dieldrin, heptachlor, lindane, and toxaphene. While the use of these pesticides are banned, there are many other approved pesticides that are applied to food crops in the course of normal agricultural operations. The use of approved pesticides is regulated by governmental agencies, and pesticide residues are closely monitored by food processors to prevent con-tamination of processed food products. Although no longer used in the United States, many of the banned pesticides persist in the environment. Their presence in treated sludge or reclaimed wastewater that is applied to agricultural land used for food crop production may be viewed by food processors as an additional uncontrollable pesticide burden, and they have expressed concern about their product quality being compromised when treated sludge and wastewater are used in food crop production (National Food Processors

Association, 1992). Residues of aldrin and dieldrin were detected in 3 and 4 percent, respectively, of the 209 sludges surveyed in the NSSS, at concentrations of 0.002 mg/kg and at LOD, respectively (Kuchenrither and Carr, 1991). Studies in the late 1980s reported median concentrations in sludges for the pesticide compounds listed above to be very much less than 1 mg/kg dry weight and, frequently, below detection limits (0.05 mg/kg) (EPA, 1990). Application rates of sludges with such median concentrations would result in concentrations in plant growth media of less than 0.01 mg/kg. Pot and field studies (Baxter et al., 1983; Kampe, 1989; Singh, 1983; Witte et al., 1988), some of which spanned 30 years, found that sludge additions failed to measurably increase the level of chlorinated pesticides and hydrocarbons above background soil levels. In a study involving the application of municipal sludge to coal refuse at a rate of 3360 tons/ha, Webber et al. (1994) observed sludge concentrations of organochlorine pesticides ranging from less than 0.015 mg/kg (heptachlor, aldrin, pp'-DDT, lindane, hexachlorobutadiene, and hexachlorobenzene) to 0.217 mg/kg (pp'-DDE). Other pesticides and their concentrations (in mg/kg) include alpha and gamma chlordane (0.08 and 0.125), dieldrin (0.042), and pp'-DDD (0.091). Levels of the pesticides in corn, cabbage, and carrot tissues grown on the sludge-treated coal refuse were less than the method's detection limit of 0.1 mg/kg dry weight. Based on the information presented, it seems reasonable to conclude that it is highly unlikely that pesticides in sludge applied to land will harm crops or their consumers.

### *Disinfection Products*

In United States, the treatment and discharge of municipal wastewater are governed by regulations derived from federal legislative mandates in P.L. 92-500 as well as state statutes. The current regulations consider disinfection as an important element of wastewater treatment when necessary to protect public health. While several options are available, chlorination is by far the most commonly used disinfection process for wastewater effluents.

Rook (1976) discovered that chlorine used in disinfection reacts with naturally occurring humic substances in the water to form trihalomethanes (THMs), the most prevalent species among them are chloroform, bromodichloromethane, dibromochloromethane, and bromoform. Subsequent surveys of municipal water supplies in the United States found that THMs were log-normally distributed with median concentrations less than 30 µg/l. Bellar et al. (1974) found that chloroform also formed during the chlorination of wastewater effluents. Jolley (1975), Glaze and Henderson (1975), and Glaze et al. (1978) evaluated other chlorine-containing compounds that also form, including chlorophenols, chlorobenzoic and chlorophenylacetic acids, and chlorinated purines and pyrimidines.

Byproducts of water and wastewater chlorination are subjects of continuous investigations worldwide (Johnson and Jensen, 1986; Badaway, 1992). Based on their concentrations in chlorinated wastewater effluents and their fate and transport characteristics, it seems unlikely that they will persist in the receiving soil or become bioaccumulated to any great extent (Jolley, 1978; Howard, 1989). Although actual data on the persistence of these byproducts on food crops are lacking, their presence should not limit the use of reclaimed water for crop irrigation.

### *Acid-Extractable Organic Compounds*

Some important acid-extractable organic compounds include phenols, pentachlorophenol (PCP), and 2,4-dinitrophenol (DNP). These compounds' toxicity to plants is pH-dependent. While toxicity and soil sorption are greatest in acid soils, compound degradability and mobility in soil are likewise greatest in high pH conditions. Studies of PCP and DNP in high pH soils found soil degradation to be rapid. O'Connor et al. (1991) concluded that when soils amended with sludges containing median concentrations of phenol (from NSSS, EPA 1993b) are managed according to sound agronomic practices, neither DNP or PCP would be expected to persist in soils long enough to impact plants, and little plant contamination would be expected.

#### *Chlorinated Dibenzo-p-dioxins (CDDs) and Dibenzofurans (CDFs).*

Like PAHs, dioxins are generally formed by combustion of organic solids, and they have been reported in sludge quality surveys. In a literature review of studies of plants exposed to dioxin-contaminated soils, Kew et al. (1989) found that root uptake and translocation of tetrachlorodibenzodioxin (TCDD) was minimal, but that significant contamination may occur by volatilization of TCDD from soil and absorption by foliage. Facchetti et al. (1986) concluded that volatilization would be the dominant process of transfer from soil to plant, if it were to occur. Nevertheless, O'Connor et al. (1991) concluded that contamination of plants with either CDDs or CDFs would be unlikely due to their very low bioavailability. Even if present at high initial concentrations, dioxins in sludge would be diluted at least 100-fold at agronomic sludge application rates, and the organic matter with the sludge would increase soil sorption capacity and likely reduce volatilization.

#### *Volatile Aromatic Compounds (VOC)*

Although volatile aromatic compounds (e.g., toluene, benzene, xylene) in municipal wastewater are largely lost by volatilization in the sewer or at the treatment plant, they have been detected in sludges (Jacobs et al., 1987). Benzene was detected in 93 percent of the municipal sludge samples analyzed in the 40 Cities Survey (EPA, 1982) with a mean concentration of 1.8 mg/kg; however, some fraction of these sludges were untreated, and therefore not eligible for land application under current regulations. Benzene was detected in less than 2 percent of the sludges from 209 treatment plants in the NSSS in a range of concentrations from 0.01 to 0.2 mg/kg. When sludges are applied to soils, volatile aromatics are rapidly lost due to volatilization. Jin and O'Connor (1990) and concluded that volatile aromatics do not persist long enough in sludge-treated soils to represent a problem for agriculture under normal aerobic soil conditions. However, anaerobic conditions resulting from both bacterial depletion of oxygen due to high organic carbon content and lack of reaeration due to water-logging can promote temporary volatile aromatic retention in soil (Jin and O'Connor 1990). This source of volatile organics should not pose a problem since crops, except for rice, are not grown in waterlogged soils. Some investigators have reported contamination and bioconcentration of toxic volatile aromatic compounds in plants (Facchetti et al., 1986). O'Connor et al. (1991) suggested that further research was necessary to study possible plant contamination by sorption of chemical vapors from volatile aromatic compounds and other volatile organic compounds (e.g., toxaphene, and



TCDD).

### *Generalizations Regarding Uptake of Organics by Plants*

Plant bioconcentration factors for most toxic organics are small and are often not significantly different from zero. Most toxic organics occur in sludge at low concentrations and when the sludge is applied to soil, their concentrations are further diluted. Furthermore, toxic organic compounds that are not destroyed by biodegradation, chemical oxidation, or photolysis are so strongly sorbed to the sludge-soil particulate matrix as to have low bioavailability to plants. Many of the organic compounds are chemically or biologically degraded or volatilized from the soil during the cropping season. Finally, because the fraction of sludge-borne toxic organics that does remain in soil has low bioavailability, absorption by crops is negligible. In some cases, volatile toxic organics may contaminate plant tissue through absorption of volatilized compounds; however, management practices, such as incorporation of sludge with soil and application of sludge before plant sprouting, will substantially reduce any plant exposure to VOCs.

Available data indicate that potentially harmful toxic organic pollutants do not enter edible portions of plants that are irrigated with treated municipal wastewater. Irrigation of vegetables in test plots with wastewaters has shown no accumulation of PAHs, especially benzo(a)pyrene (Il'nitskii et al. 1974). In a study of aldehydes and other organics at agricultural land treatment sites, Dodolina et al. (1976) found no uptake of acetaldehyde, crotonaldehyde, and benzaldehyde in the aboveground portions of potatoes and corn. Cyclohexanone and cyclohexanol could be found in corn plants four days after irrigation, but not later. Di-chloroethane was taken up by beets and cereals, but was metabolized and absent within about two weeks after irrigation. Although these compounds are found in crops and soils, they appear to be metabolized at a rate sufficient to prevent their occurrence in the harvested product.

### **Uptake of Toxic Organics by Animals**

Toxic organic compounds present in plant tissues (and soil in the case of pastured animals) may be incorporated into animal tissues. However, the low levels of toxic organic compounds to be expected in the aboveground portions of plants growing at land application sites pose little hazard to animals feeding upon them. Under certain site-specific conditions, however, high concentrations of particular organic compounds in the sludge may cause problems.

In considering the pathways for exposure of humans to toxic organic compounds in sludge, Chaney (1985) suggests that direct ingestion of the sludge-soil mixture by animals is the only reasonable route by which toxic organic chemicals have been traced directly from sludge to animal products. However, the transfer of toxic organic compounds to the human food chain via this pathway is still considered to be negligible.

## **FATE OF AND EXPOSURE TO TRACE ELEMENTS IN SLUDGE**

The availability of inorganic chemicals or trace elements for uptake by plants (and thus entry into the human food chain) or transport to ground water is limited by the extent to which these elements remain free in the soil solution. The binding of chemicals to the soil substrate is controlled by the chemical processes of complexation to organic matter, adsorption, and precipitation. Adsorption occurs on organic matter, hydrous oxides of iron and manganese, clays, and other soil minerals. Precipitation reactions include the formation of sparingly soluble oxides, hydroxides, carbonates, phosphates, and sulfides, etc. Mercury may leave the soil through volatilization. As a result of these processes, only small amounts of the trace elements remain free in the soil solution where they would be available for absorption by plant roots. These processes are strongly affected by soil pH, with cation levels decreasing and anion levels increasing in the soil solution with increasing pH (Logan and Chaney, 1983).

Chaney (1980) introduced the concept of the "soil-plant barrier" for consideration of potential toxicity to the food chain if trace elements are applied to soils. After a trace element enters the root cells, translocation to other plant organs (tubers, shoots, leaves, fruits, seeds), depends on the properties of the specific element and the plant. Important plant processes are membrane surfaces, organic chelators, and cells specialized for pumping materials into the xylem, through which it reaches the shoot. One group of metals are so insoluble or so strongly adsorbed to soil or plant roots that they are not translocated into edible plant parts regardless of quantities present in the soil. Elements like trivalent chromium, mercury, and lead are examples. Lead is so insoluble inside the plant root and mercury is so strongly bound inside the fibrous plant roots that harmful levels of these elements are not found in edible plant parts (Berthet et al., 1984; Naylor and Mondy, 1984). Mercury can be transferred through volatilization from the soil surface to plant foliage, but this route is not very relevant to sludges because they are low in mercury.

Copper and zinc belong to another group of elements that are translocated to vegetative parts of the plant. However, before they reach levels in crops that could be potentially harmful to consumers, crop growth is so severely stunted that they are not harvestable. Phytotoxicity thus prevents excessive plant concentration of contaminants to levels harmful to animals, and the food chain is protected (see Chapter 4).

There are exceptions to protection by the soil-plant barrier. These exceptions have been the focus of intense research in agriculture. Livestock have been injured by forage grown on soils with excessive geochemically-derived selenium (Mayland, 1994) or molybdenum (Mills and Davis, 1987) for centuries. Cadmium is absorbed by crops and can reach levels that are dangerous to humans if a high percentage of the consumer's diet is derived from crops grown on cadmium-contaminated soil over an extended time period. Human disease from soil cadmium occurred in Japan where mining wastes polluted paddy rice fields, and farm families consumed the rice grown on these paddies for 40 or more years (Kobayashi, 1978; Nomiyama, 1986). It should be noted that the diets of the Japanese families were low in calcium and zinc. Calcium, iron, and zinc play major roles in interfering with cadmium absorption in the human intestine (Fox, 1988), and when sufficiently present in the human diet, high cadmium foods will not always produce health effects.

Selenium toxicity to humans has been documented in China (Combs and Combs, 1986); however, the source of the selenium was not municipal sewage sludge. Municipal sewage sludges are not high in selenium. Similarly for livestock, soil molybdenum is a potentially toxic element, but no cases have been reported of molybdenum toxicity to animals from consumption of forage grown on sludge-amended soils. In pot studies, where clover was grown on alkaline soils containing up to 16 kg

of molybdenum per ha, concentrations in the plant tissue reached levels that could be harmful to animals if the clover were to make up a substantial portion of the diet for an extended period of time (Davis, 1981).

Burau et al. (1987) collected five years of field data on the impact of treated municipal wastewater on trace element concentrations in soil and vegetables. They concluded that there was no significant difference in concentration between food crops irrigated with treated domestic wastewater and wellwater. They did observe, however, that commercial fertilizer application did result in increased plant tissue levels of some metals, such as cadmium, zinc, and man-ganese. Levels in plants, however, were well below those considered harmful.

The uptake of trace elements by plants has been reviewed by Logan and Chaney (1983). Important factors affecting uptake rate include: trace element properties, soil properties, the immediate environment (especially pH) of the roots, plant crop species, and plant crop cultivar (variety or strain). As an example of species effects, leafy vegetables, especially Swiss chard, are much better cadmium accumulators than most other plants. Cultivars of maize (corn) and wheat have been shown to vary in their rates of cadmium accumulation.

### **Uptake of Trace Elements by Animals**

As with toxic organics, animals can be exposed to trace elements through sludge residuals adhering to plants, sludge on the soil surface or mixed into the soil, or trace elements absorbed and translocated by plants. All three routes can operate on grazing land, but only the third is relevant when animals are given feed grown on sludge-amended soils.

At Werribee Farm in Melbourne, Australia, cattle are grazed on wastewater-irrigated pastures and show higher organ levels of cadmium and chromium than farm cattle grazed on nonirrigated pastures, although elevated levels were within the range common to cattle in general (Croxford, 1978). Organ levels of lead, however, did not increase, in spite of lead increases in both soil and pasture plants. Dowdy, et al. (1983) fed silage grown on sludge-amended soils to goats and sheep for a period of three years. The silage contained up to 5.3 mg cadmium/kg, a concentration 10 or more times greater than normal silage. Cadmium and zinc were elevated in the kidney and liver but not in the muscle tissue. Concentrations of copper, nickel, chro-mium, lead and zinc in organs of animals fed silage from sludge-amended soils did not differ from animals fed a similar diet of forage from lands not receiving sludge. The concentrations of cadmium in the milk of goats that were fed the treated silage did not differ from controls (Dowdy et al., 1983). Telford et al. (1984) also reported that the concentration of cadmium in milk from goats fed silage from sludge-amended soils over a period of 135 days did not differ from the controls. Silage from the sludge-amended soils contained up to 3.8 mg cadmium/kg; control silage contained 0.14 mg cadmium/kg. Concentrations of cadmium in the livers of adult goats fed silage from the sludge-amended soils were significantly greater than the controls.

Studies of the accumulation of trace elements in cattle grazed on sludge-amended pastures have revealed elevated levels in liver and kidney, but not in muscle tissue (Bertrand et al. 1981a, Baxter et al. 1983). Sheep grazing on sludge-amended pasture had no statistically significant increases in tissue levels of cadmium (Hogue et al. 1984). Bertrand et al. (1981b) observed no increases in metal concentration of cattle fed sorghum grown on sludge-amended soil nor did increases occur in tissues of

mice or guinea pigs fed lettuce and Swiss chard grown on sludge-amended soil (Chaney et al., 1978a,b). Other studies, however, have shown significant increases of cadmium in kidney and liver, but not in muscle tissue of animals fed sludge-fertilized crops (Hansen and Hinesly, 1979; Lisk et al., 1982; Hinesly et al., 1984; Bray et al., 1985; Hogue et al., 1984). Trace element levels and disease conditions of cattle grazing on land reclaimed using Chicago sludge have been observed by Fitzgerald et al. (1985) for up to eight years; they concluded that little risk to man or animals is associated with land application of anaerobically digested wastewater sludge. All cattle remained healthy and no pathological changes could be attributed to sludge.

Based on these studies, it appears that trace elements either do not accumulate or accumulate in very small quantities in animal muscle tissue. However, cadmium and other metals do accumulate in the liver and kidneys of animals.

The current state of knowledge allows the following generalizations to be made: Elements potentially harmful to consumers which may accumulate in crops include cadmium (humans, animals), molybdenum (animals), and selenium (animals). Cadmium accumulates in the liver and kidney of animals fed crops grown on sludge-amended soils. Although levels of cadmium in foods grown on sludge-amended soil are elevated, there are no documented cases of human or animal poisoning from this source.

### **NONSPECIFIC HEALTH EFFECTS OF SLUDGE AND WASTEWATER**

Although a large number of organic and inorganic constituents of sewage have been identified in the Priority Pollutants list and additions as discussed above, some of the residual fraction that is unidentified may have nonspecific human health effects. Investigations of non-specific or general health effects normally consist of research on non-human organisms as explained below.

Nonspecific toxicological testing of whole reclaimed water using the Ames Salmonella Microsome Mutagen Assay and the Mammalian Cell Transformation Assay have been used to indicate the potential of mutagenic, cytotoxic, and carcinogenic effects on bacterial and mammalian cells (Nellor et al., 1984). Clevinger et al. (1983) performed bioassays on five sludges using the Ames test and found that none had significant mutagenic effects. In another large study where concentrated reclaimed wastewater was given to several species of test animals for two years, no acute or chronic health or developmental effects were found (Lauer, 1993). The latter study is of particular interest because of its thoroughness. The National Research Council (1982) concluded that the "ultimate" evaluation of health effects of reclaimed wastewater used for drinking water purposes must come from studies on whole animals. Furthermore, because of the difficulty of reliable interpretation of results of bioassay studies using single compounds isolated from wastewater, test animals should be exposed to concentrates of the whole reclaimed water.

Accordingly, in a two-year animal effects testing study of reclaimed wastewater for potable reuse, the Denver Water Department compared reclaimed wastewater with Denver municipal drinking water in tests administered to animals as drinking water (Lauer et al., 1990). Matched groups of Fischer 344 rats and B<sub>6</sub>C<sub>3</sub>F<sub>1</sub> mice were used to test for acute and chronic (including carcinogenic) health effects over the 104-week study, and Sprague-Dawley rats were used for a two-generation reproductive toxicity study. The results of chronic toxicity/carcinogenic studies of both rats and mice showed no significant differences in clinical pathology (hematology, clinical chemistry, and urinalysis)

and gross pathology. Similarly, no significant difference were found in the formation of neoplasms between reclaimed water-fed mice and rats and controls fed both treated drinking water and distilled water. Finally, there were no demonstrated effects from the ingestion of reclaimed water on reproductive performance, fetal development, offspring survival or growth during the two-year reproductive study (Lauer, 1993).

A number of studies have been conducted on the effects of feeding sewage sludges to animals either directly or where animals have ingested sewage sludge that was sprayed on for-age. Where cattle were fed up to 12 percent of their diet from sludge for 94 days, or 6 percent of their diet for 141 days, no adverse health effects were noted (Keinholz et al., 1979; Bertrand et al., 1981a). No adverse effects were noted when baby pigs were raised on a diet consisting of 5 percent sludge for a period of one month (Firth and Johnson, 1955). Rats and Japanese quail fed diets with 30 percent sludge for 2 weeks showed no adverse health effects (Cheeke and Meyer, 1973). Baby chicks consuming a diet consisting of 10 percent sludge for about one month showed no adverse health effects (Firth and Johnson, 1955); where 20 percent of the diet of birds came from sludge, however, body weight gain and liver vitamin A were found to be reduced (Keinholz, 1980). In a four-year study, where 7 percent of the dry matter of the diet of breeding ewes consisted of sludge sterilized by gamma irradiation, Smith et al. (1985) reported no accumulation of hazardous levels of toxic elements and little, if any, evidence of toxicity. Johnson et al. (1981) fed Hereford steers a diet that included 11.5 percent sewage sludge and observed retention of dietary cadmium, mercury and lead to be 0.09 percent, 0.06 percent and 0.3 percent, respectively. Concentrations of these elements in the liver and kidneys of steers were found to increase from 5- to 20-fold following sludge ingestion. The authors' data showed that less than 0.3 percent of the sludge-fed cadmium, mercury, and lead were retained by the cattle. These data indicated that cattle are a moderately effective screen against entry of toxic elements from sludge into the human food chain. Damron et al. (1982) substituted 7 percent sludge in the diet of white Leghorn hens for a period of 84 days and observed no effect on bird performance; egg production was unaffected and no increase in cadmium was found in the eggs.

Keinholz (1980) cited a study of PCBs in cabbage grown on sludge-amended soil that was reportedly linked to degenerative changes in liver and thyroid of sheep (Haschek et al., 1979, cited by Keinholz, 1980). However, the study involved cabbage grown in sludge that contained 17 mg/kg PCBs, an unusually high concentration and about twice the maximum observed in the NSSS. Adding this sludge to soil at an agronomic rate would result in a PCB concentration in soil of less than 0.1 mg/kg in the surface 20 centimeters. Therefore, it is unlikely that sludge containing even this high a concentration of PCBs would be harmful if applied to agricultural land according to present-day regulations and guidelines.

Hansen et al. (1976) studied young swine fed for 56 days on corn grown on sludge-fertilized land. Electroencephalograms, electrocardiograms, clinical chemistry, and histopathology were all normal. However, they did observe elevated levels of hepatic microsomal mixed function oxidase (MFO) activity. This increased MFO activity may have been caused by toxic organics and inorganic trace elements in the sludge, and the authors concluded that further study should be performed before such grain can be recommended as the major dietary component for animals over long periods.

An epidemiologic study on human exposure to pathogens in sludge compared health effects in 164 people living on 47 farms which received 2 to 10 tons of sludge per ha per year for three years to 130 people from 45 farms who formed a control group. Both study groups were from geologically matched areas of rural Ohio. Study participants answered monthly surveys and had annual tuberculin

testing and serological tests of quarterly blood samples. In addition, monthly surveys included questions about farm animals' health. It was found that there were no significant differences in the health of those living on farms where sludge was applied compared to the control group with respect to respiratory or digestive illness or other reported physiological symptoms. Similarly, no differences were reported between domestic animals from sludge-amended versus control farms (Brown et al., 1985).

### **SUMMARY**

A review of the literature for toxic organics and for inorganic trace elements in treated municipal wastewater effluents or treated sewage sludge indicates that most of these chemicals are either not transferred from soil to plant tissues or that translocation to edible tissues does not reach levels harmful to consumers under normal agricultural conditions.

With regards to human health concerns about the use of treated sludge on crops, the inorganic chemical considered to be of greatest concern is cadmium. This conclusion is consistent with the policy approach taken to minimize health risks due to chemicals from land application of sewage sludge.

Research on the bioavailability of toxic organic compounds to plants indicates that the risk to humans consuming food crops grown on soils amended with sludge is negligible. Toxic organic compounds are typically present at such low concentrations and/or are largely not bioavailable to plants, that they would accumulate only at very low concentrations, if at all, in edible portions of plants.

Few adverse health effects have been found in studies where treated sludge and treated effluent were fed directly to animals. No adverse human acute or chronic toxicity effects have been reported resulting from ingestion of food plants grown in soils amended by sludges or crops irrigated by reclaimed water.

### **REFERENCES**

- Alexander, M. 1994. *Biodegradation and Bioremediation*. San Diego, Calif.: Academic Press, 302 pp.
- Atlas, R. M. 1981. Microbial degradation of petroleum hydrocarbons: An environmental perspective. *Microbiol. Rev.* 45:180-209.
- Aranda, J. M., G. A. O'Connor, and G. A. Eiserman. 1989. Effects of sewage sludge on di-(2-ethylhexyl)phthalate uptake by plants. *J. Environ. Qual.* 18:45-50.
- Badaway, M. I. 1992. Trihalomethane in drinking water supplies and reused water. *Bull. Environ. Contam. Toxicol.* 48:157-162.
- Baker, E. L., Jr., P. J. Landrigan, C. J. Gluek, M. M. Zack, Jr., J. A. Lidde, V. W. Burse, W. J. Housworth, and L. L. Needham. 1980. Metabolic consequences of exposure to polychlorinated biphenyls (PCB) in sewage sludge. *Am. J. Epidem.* 112:553-560.
- Baxter, J. C., D. E. Johnson, and E. W. Kienholz. 1983. Heavy metals and persistent organics content in cattle exposed to sewage sludge. *J. Environ. Qual.* 12:316-319.
- Bellar, T. A., J. H. Lichtenberger, C. R. Kroner. 1974. The Occurrence of Organohalides in

- Chlorinated Drinking Waters. *J. Amer. Water Works Assoc.* 66:703-706.
- Berthet, B., C. Amiard-Triquet, C. Metayer, and J. C. Amiard. 1984. Etude des voies de transfert du plomb de l'environnement aux vegetaux cultives: Application a l'utilisation agricole de boues de station d'epuration. *Water Air Soil Poll.* 21:447-460.
- Bertrand, J. E., M. C. Lutrick, G. T. Edds, and R. L. West. 1981a. Metal residues in tissue, animal performance, and carcass quality with beef steers grazing on Pensacola Bahiagrass pastures treated with liquid digested sludge. *J. Animal Sci.* 53:146-163.
- Bertrand, J. E., M. C. Lutrick, G. T. Edds, and R. L. West. 1981b. Animal performance, carcass quality, and tissue residues with beef steers fed forage sorghum silages grown on soil treated with liquid digested sludge. *Proc. Soil Crop Sci. Soc. Florida* 40: 111-114.
- Bossert, I. D., and R. Bartha. 1986. Structure-biodegradability relationships of polycyclic aromatic hydrocarbons in soil. *Bull. Environ. Contam. Toxicol.* 37:490-495.
- Bray, B. J., R. H. Dowdy, R. D. Goodrich, and D. E. Pamp. 1985. Trace metal ac-cumulations in tissues of goats fed silage produced on sewage sludge-amended soil. *J. Environ. Qual.* 14:114-118.
- Brown, R. E. 1985. Demonstration of acceptable systems for land disposal of sewage sludge. Water Engineering Research Lab. EPA 600/Z-85-062. Cincinnati, Ohio: U.S. Environmental Protection Agency.
- Burau, R. G., B. Sheikh, R. P. Cort, R. C. Cooper, and D. Ririe. 1987. Reclaimed Water for Irrigation of Vegetables Eaten Raw. *California Agriculture*, July-August:4-7.
- Chaney, R. L. 1980. Health risks associated with toxic metals in municipal sludge. Pp. 59-83 in *Sludge—Health Risks of Land Application*. G. Bitton, B. L. Damron, G. T. Edds, and J. M. Davidson, eds. Ann Arbor, Mich.: Ann Arbor Science Publications.
- Chaney, R. L. 1985. Potential effects of sludge-borne heavy metals and toxic organics on soils, plants, and animals, and related regulatory guidelines. Pp. 1-56 in *Final Report of the Workshop on the International Transportation, Utilization or Disposal of Sewage Sludge Including Recommendations*. PNSP/85-01. Washington, D.C.: Pan American Health Organization.
- Chaney, R. L. 1993. Risks associated with the use of sewage sludge in agriculture. Pp. 7-31 in *Proc. 15th Federal Convention Australian Water and Wastewater Association (Gold Coast, Queensland, April 18-23)*. Volume 1. Australian Water and Wastewater Association (Queensland Branch), P.O. Box 5412 West End, Queensland, Australia 4012.
- Chaney, R. L., G. S. Stoewsand, A. K. Furr, C. A. Bache, and D. J. Lisk. 1978a. Elemental content of tissues of guinea pigs fed Swiss chard grown on municipal sewage sludge-amended soil. *J. Agr. Food Chem.* 26:994-995.
- Chaney, R. L., G. S. Stoewsand, A. K. Furr, C. A. Bache, and D. J. Lisk. 1978b. Cadmium deposition and hepatic microsomal induction in mice fed lettuce grown on municipal sludge-amended soil. *J. Agric. Food Chem.* 26:992-994.
- Cheeke, P. R., and R. O. Meyer. 1973. Evaluation of the nutritive value of activated sewage sludge with rats and Japanese quail. *Nutrition Reports International* 8:385-392.
- Clevinger, T. E., D. D. Hemphill, K. Roberts, and W. A. Mullins. 1983. Chemical composition and possible mutagenicity of municipal sludges. *Jour. WPCF* 55:1470-1475.
- Combs, G. F., Jr., and Combs, S. B. 1986. *The Role of Selenium in Nutrition*. New York: Academic Press, 532 p.
- Cork, D. J., and J. P. Krueger. 1991. Microbial transformations of herbicides and pesticides. *Advan.*

- Applied Microbiol. 36:1-66.
- Croxford, A. H. 1978. Melbourne, Australia, Wastewater System: Case Study. 1978 Winter Meeting, Chicago, Illinois. Amer. Soc. Agric. Eng. Paper No. 78-2576. St. Joseph, Michigan.
- Damron, B. L., H. R. Wilson, M. F. Hall, W. L. Johnson, O. Osuna, R. L. Suber, and G. T. Edds. 1982. Effects of feeding dried municipal sludge to broiler type chicks and laying hens. *Polut. Sci.* 61:1073-1081.
- Davis, R. D. 1981. Uptake of molybdenum and copper by forage crops growing on sludge-treated soils and its implication for the health of grazing animals. Pp. 194-197 in *Proc. Intl. Conf. on Heavy Metals in the Environment*, CEP Consultants, Edinburgh, Scotland.
- Davis, T. S., J. L. Pyle, J. H. Skillings, and N. D. Danielson. 1981. Uptake of poly-chlorobiphenyls present in trace amounts from dried municipal sewage sludge through an old field ecosystem. *Bull. Environ. Contam. Toxicol.* 27:689-694.
- Dean, R. B., and M. J. Seuss. 1985. The risk to health of chemicals in sewage sludge applied to land. *Waste Manage. Res.* 3:251-278.
- Dodolina, V. T., L. Y. Kutepov, and B. F. Zhirnov. 1976. Permissible quantities of organic substances in waste waters used for irrigation. *Vestn. S-kh. Nauki Moscow* 6:110-113.
- Dowdy, R. H., B. J. Bray, and R. D. Goodrich. 1983. Trace metal and mineral composition of milk and blood from goats fed silage produced on sludge-amended soil. *J. Environ. Qual.* 12:473-478.
- EPA. 1982. Fate of priority pollutants in publicly-owned treatment works. Vol. 1. EPA 440/1-82-3.3. Washington, D.C.: U.S. Environmental Protection Agency, Industrial Technology Division.
- EPA. 1984. Environmental Regulations and Technology. Use and Disposal of Municipal Wastewater. Washington, D.C.: U.S. Environmental Protection Agency.
- EPA. 1990. National Sewage Sludge Survey: Availability of information and data, and anticipated impacts on proposed regulations. Proposal Rule 40CFR Part 503. *Federal Register* 55:47210-47283. November 9, 1990.
- EPA. 1993a. Standards for the Use And Disposal of Sewage Sludge, Final Rule, 40 CFR Part 257, 403 and 503. *Federal Register* 58(32):9248-9415.
- EPA. 1993b. Technical Support Documents for 40 CFR Part 503. Land Application of Sewage Sludge, Vol. I-PB93-11075; Land Application of Sewage Sludge, Vol. II-PB93-110583, Appendices A-L; Pathogen and Vector Attraction Reduction in Sewage Sludge—PB93-110609; Human Health Risk Assessment for Use and Disposal of Sewage Sludge, Benefits of Regulation—PB93-111540; The Regulatory Impact Analysis—PB93-110625. Springfield: Virg.: National Technical Information Service.
- Facchetti, S., A. Balasso, C. Fichtner, G. Frare, A. Leoni, C. Mauri, and M. Vasconi. 1986. Studies on the absorption of TCDD by some plant species. *Chemosphere* 15:1387-1388.
- Fairbanks, B. C., and G. A. O'Connor. 1984. Effect of sewage sludge on the adsorption of polychlorinated biphenyls by three New Mexico soils. *J. Environ. Qual.* 13:297-300.
- Firth, J. A., and B. C. Johnson. 1955. Sewage sludge as a feed ingredient for swine and poultry. *Agri. and Food Chem.* 3:795-796.
- Fitzgerald, P. R., J. Petersen, and C. Lue-Hing. 1985. Heavy metals in tissues of cattle exposed to sludge-treated pastures for eight years. *Am. J. Vet. Res.* 46:703-707.



- Fries, G. F., and G. S. Marrow. 1981. Chlorobiphenyl movement from soil to soybean plants. *J. Agr. Food Chem.* 29:757-759.
- Fox, M. R. S. 1988. Nutritional Factors that may influence the bioavailability of cadmium. *J. Env. Quality* 17:175-180.
- Gan, D. R. and P. M. Berthoex. 1994. Disappearance and crop uptake of PCBs from sludge-amended farmland. *Water Environment Research* 66(1):54-69.
- Glaze, W. H. and J. E. Henderson. 1975. Formation of Organichlorine compounds from the Chlorination of a Municipal Secondary Effluent. *Jour. WPCF* 47:2511-2515.
- Glaze, W. H., J. E. Henderson, and G. Smith. 1978. Analysis of new chlorinated compounds by chlorination of municipal wastewater. pp. 139-159 in *Water Chlorination: Environmental Impact and Health Effects*. Vol 1. R. L. Jolley, ed. Ann Arbor, Mich.: Ann Arbor Publishers, 439pp.
- Griffin, R. A., and E. S. K. Chian. 1980. Attenuation of Water-Soluble Polychlorinated Biphenyls by Earth Materials. EPA-600/2-80-027. Cincinnati, Ohio: U.S. Environmental Protection Agency.
- Hansen, L. G., and T. D. Hinesly. 1979. Cadmium from soil amended with sewage sludge: Effects and residues in swine. *Environ. Health Perspect.* 28:51-57.
- Hansen, L. G., J. L. Dorner, C. S. Byerly, R. P. Tarara, and T. D. Hinesly. 1976. Effects of sewage sludge-fertilized corn fed to growing swine. *Amer. J. Vet. Res.* 37:711-714.
- Haschek, W. M., A. K. Furr, T. F. Parkinson, C. L. Heffron, J. T. Reid, C. A. Bache, P. C. Wszolek, W. H. Gutenmann, and D. J. Lisk. 1979. Element and polychlorinated biphenyl deposition and effects in sheep fed cabbage grown on municipal sewage sludge. *Cornell Vet.* 69:302-314.
- Hinesly, T. D., L. G. Hansen, and G. K. Dotson. 1984. Effects of Using Sewage Sludge on Agricultural and Disturbed Lands. EPA-600/2-83-113. Cincinnati, Ohio: U.S. Environmental Protection Agency.
- Hogue, D. E., J. J. Parrish, R. H. Foote, J. R. Stouffer, J. L. Anderson, G. S. Stoews and, J. N. Telford, C. A. Bache, W. H. Gutenmann, and D. J. Lisk. 1984. Toxicological studies with male sheep grazing on municipal sludge-amended soil. *J. Toxicol. Environ. Health* 14:153-161.
- Howard, P. H. 1989. *Handbook of Environmental Fate and Exposure Data for Organic Chemicals*, Volumes I and II. Chelsea, Mich.: Lewis Publishers.
- Il'nitskii, A. P., L. G. Solenova, and V. V. Ignatova. 1974. Sanitary and oncological assessment of agricultural use of sewage containing carcinogenic hydrocarbons. *ORNL- tr-2959. Kazanskii Med. Zh.* 2:80-81.
- Iwata, Y., F. A. Gunther, and W. E. Westlake. 1974. Uptake of a PCB (Arochlor 1254) from soil by carrots under field conditions. *Bull. Environ. Contam. Toxicol.* 11:523-528.
- Jacobs, L. W., G. A. O'Connor, M. A. Overcash, M. J. Zabik, and P. Rygiewicz. 1987. Effects of trace organics in sewage sludges on soil-plant systems and assessing their risks to humans. Pp. 107-183 in *Land Application of Sludge*, A. L. Page, T. J. Logan, and J. A. Ryan, eds. Chelsea, Mich.: Lewis Publishers.
- Jenkins, T. F., D. C. Leggett, L. V. Parker, J. L. Oliphant, C. J. Martel, B. T. Foley, and C. J. Diener. 1983. Assessment of the Treatability of Toxic Organics by Overland Flow. *CRREL Report 83-3*. Hanover, New Hampshire: U.S. Army Corps of Engineers.

- Jin, Y. and G. A. O'Connor. 1990. Behavior of toluene added to sludge-amended soils. *J. Environ. Qual.* 19:573-580.
- Johnson, D. E., E. W. Kienholz, J. C. Baxter, E. Spanger and G. M. Ward. 1981. Heavy metal retention in tissues of cattle fed high cadmium sewage sludge. *J. Anim. Sci.* 51: 108-114.
- Johnson, J. D. and J. N. Jensen. 1986. THM and TOX formation: routes, rates, and precursors. *J. Amer. Water Works Assoc.* 78:156-161.
- Jolley, R. L. 1975. Chlorine-containing Organic Constituents in Chlorinated Effluents. *J. WPCF* 47:601-618.
- Jolley, R. L., ed. 1978. *Water Chlorination: Environmental Impact and Health Effects*. Ann Arbor, Mich.: Ann Arbor Publishers.
- Kampe, W. 1989. Organic substances in soils and plants after intensive application of sewage sludge. Pp. 180-185 in *Sewage Sludge Treatment and Use: New Developments, Technological Aspects and Environmental Effects*. A. H. Dirkzwager and P. L'Hermite, eds. London: Elsevier Applied Sci. Publ.
- Kew, G. A., J. L. Schaum, P. White, and T. T. Evans. 1989. Review of plant uptake of 2,3,7,8-TCDD from soil and potential influences on bioavailability. *Chemosphere* 18: 1313-1318.

- Keinholz, E. W. 1980. Effects of toxic chemicals present in sewage sludge on animal health. Pp. 153-171 in *Sludge--Health Risks of Land Application*. G. Bitton, B. L. Damron, G. T. Edds, and J. M. Davidson, eds. Ann Arbor, Mich.: Ann Arbor Science Publishers.
- Keinholz, E., G. M. Ward, D. E. Johnson, J. Baxter, G. Braude, and G. Stern. 1979. Metropolitan Denver sludge fed to feedlot steers. *J. Anim. Sci.* 48:735-741.
- Kobayashi, J. 1978. Pollution by cadmium and the itai-itai disease in Japan. pp. 199-260 in *Toxicity of Heavy Metals in the Environment*. F. W. Oehme, ed. New York: Marcel Dekker.
- Kuchenrither, R. D. and S. Carr. 1991. A review of the National Sewage Sludge Survey Results. 62nd Annual Meeting, Water Pollution Control Assn., Springfield, MO. Water Environment Federation, Alexandria, Vir.
- Lauer, W. C., F. J. Johns, G. W. Wolfe, B. A. Meyers, L. W. Condie, and J. F. Borzelleca. 1990. Comprehensive health effects testing program for Denver's potable water reuse demonstration project. *J. Toxicol. & Environ. Health* 30:305-321.
- Lauer, W. C. 1993. Denver's direct potable water reuse demonstration project. Final report. Denver, Colo.: Denver Water Department.
- Lee, C. Y., W. F. Shipe, L. W. Naylor, C. A. Bache, P. C. Wszolek, W. H. Gutenmann, and D. J. Lisk. 1980. The effect of a domestic sewage sludge amendment to soil on heavy metals, vitamins, and flavor in vegetables. *Nutr. Rep. Int.* 21:733-738.
- Lin, D. 1980. Fate of petroleum hydrocarbons in sewage sludge after land disposal. *Bull. Environ. Contam. Toxicol.* 25:616-622.
- Lisk, D. J., R. D. Boyd, J. N. Telford, J. G. Babish, G. S. Stoewsand, C. A. Bache, and W. H. Gutenmann. 1982. Toxicological studies with swine fed corn grown on municipal sewage sludge-amended soil. *J. Animal Sci.* 55:613-619.
- Logan, T. J., and R. L. Chaney. 1983. Metals. Pp. 235-323 in *Utilization of Municipal Wastewater and Sludge on Land*. A. L. Page, T. L. Gleason, J. E. Smith, I. K. Iskandar, and L. E. Sommers, eds. Riverside: University of California.
- Mayland, H. F. 1994. Selenium in plant and animal nutrition. Pp. 29-45 in *Selenium in the Environment*. W. T. Frankenberger and S. Bensen, eds. New York: Marcel Dekker.
- Metcalf and Eddy, Inc. 1991. *Wastewater Engineering: Collection, Treatment, Disposal*. New York: McGraw-Hill Book.
- Mills, C. F. and G. K. Davis. 1987. Molybdenum. In *Trace Elements in Human and Animal Nutrition*. Vol. I. 5th Edition. Walter Mertz, ed. New York: Academic Press.
- Mumma, R. O., K. A. Rashid, D. C. Raupack, B. S. Shane, J. M. Scarlet-Kranz, C. A. Bache, W. H. Gutenmann, and D. J. Lisk. 1988. Mutagens, toxicants, and other constituents in small city sludges in New York State. *Arch. Environ. Contam. Toxicol.* 17:657-663.
- National Research Council. 1980. *Drinking Water and Health*, Vol. 2. Chapter 2: The chemistry of disinfectants in water: Reactions and products. pp. 139-250. Washington, D.C.: National Academy Press.
- National Research Council. 1982. *Quality Criteria for Water Reuse*. Washington, D.C.: National Academy Press.
- National Food Processors Association. 1992. Statement by the National Food Processors Association on the Use of Treated Municipal Sewage Sludge in the production of crops for human consumption. NFPA, 6363. Clark Ave., Dublin, CA 94568-3097. May 1, 1992.
- Naylor, L. M. and R. C. Loehr. 1982. Priority pollutants in municipal sewage sludge. Part I.

- Biocycle, July/August:18-27.
- Naylor, L. M., and N. I. Mondy. 1984. Metals and PCBs in potatoes grown in sludge amended soils. Amer. Soc. Agric. Engin. Technical Paper No. NAR-84-211. St. Joseph, Michigan.
- Nellor, M. H., R. B. Baird, and J. R. Smyth. 1984. Health Effects Study—Final Report. Whittier, Calif.: County Sanitation Districts of Los Angeles County.
- Nomiyama, K. 1986. The chronic toxicity of cadmium: Influence of environmental and other variables. Pp. 101-134 in Cadmium. E.C. Fowles, ed. New York: Springer-Verlag.
- O'Connor, G. A., D. Kiehl, G. A. Eiceman, and J. A. Ryan. 1990. Plant uptake of sludge-borne PCBs. *J. Environ. Qual.* 19:113-118.
- O'Connor, G. A., R. L. Chaney, and J. A. Ryan. 1991. Bioavailability to plants of sludge-borne toxic organics. *Rev. Environ. Contam. Toxicol.* 121:129-155.
- Rook, J. J. 1976. Formation of Haloforms During Chlorination of Natural Waters. *Water Treatment Examinations* 23:234-243.
- Singh, D. 1983. The effect of land application of sewage sludge in concentration of certain sludge-associated toxic chemicals in Michigan soils and crops. Report: March 1983. Lansing, Mich.: Toxic Substances Div., Michigan Dept. of Agriculture.
- Smith, G. S., D. M. Hallford, and J. B. Watkins, III. 1985. Toxicological effects of gamma-irradiated sewage sludge solids fed as seven percent diet to sheep for four years. *J. Anim. Sci.* 61:931-941.
- Strek, H. J., and J. B. Weber. 1982. Behavior of polychlorinated biphenyls (PCBs) in soils and plants. *Environ. Pollut.* A28:291-312.
- Tabak, M. H., S. A. Quave, C. I. Mashni, and E. F. Barth. 1981. Biodegradability studies with organic priority pollutant compounds. *Jour. WPCF* 53:1503-1518.
- Telford, J. N., J. G. Babish, B. E. Johnson, M. L. Thonney, W. B. Currie, C.A. Bache, W. H. Gutermann, and D. J. Lisk. 1984. Toxicologic studies with pregnant goats fed grass-legume silage grown on municipal sludge-amended soil. *Arch. Environ. Contam. Toxicol.* 13:631-640.
- Webber, M. D., and J. D. Goodin. 1989. Persistence of volatile organic contaminants (VOC) in sludge treated soils. In: *Alternative Uses for Sewage Sludge—an International Workshop Organized by the Water Resources Center and Co-sponsored by the European Commission and the Univ. of York*, 5-7 September, 1989, York, U.K.
- Webber, M. D., R. I. Pietz, T. C. Granato, and M. L. Swoboda. 1994. Plant uptake of PCBs and other organic contaminants from sludge-treated coal refuse. *J. Environ. Quality* 23:1019-1026.
- Wild, S. R., K. S. Waterhouse, S. P. McGrath, and K. C. Jones. 1990. Organic contaminants in an agricultural soil with a known history of sewage sludge amendments: Polynuclear aromatic hydrocarbons. *Environ. Sci. Technol.* 24:1706-1711.
- Witte, H., T. Langenohl, and G. Offenbacher. 1988. Investigation of the entry of organic pollutants into soils and plants through the use of sewage sludge in agriculture. Part A. Organic pollutant load in sewage sludge. Part B. Impact of the application of sewage sludge on organic matter contents in soils and plants. *Korresponding Abwasser* 13:118-136.