

## Land Application of Biosolids: Human Health Risk Assessment Related to Microconstituents

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### Beneficial Use of Biosolids

Biosolids are generated during wastewater treatment processes and are extensively processed to meet the United States Environmental Protection Agency's (USEPA) 40 CFR Part 503 regulations promulgated in 1993, which dictate acceptable pollutant concentrations, pathogen levels, and material stability (as indicated by vector attraction reduction).

It is estimated that ~ 7.2 m dry tons of biosolids are generated in the US annually and approximately 55% (~ 3.9 m dry tons) are applied to soil for agronomic, silviculture or land restoration purposes; the remaining 45% are disposed of in municipal solid waste landfills, surface disposal units, or incineration facilities (USEPA, 2010). With an average agronomic rate of application of 10 U.S. dry tons of biosolids per acre (rates for land restoration at brownfields and mined lands may be higher) only 390,000 acres across the entire US receive annual biosolids application. The biosolids-amended land is ~ 0.12% of total harvestable acreage in the US (314,964,000 acres harvested according to the 2012 Agricultural Census). Thus, a very small proportion of cultivated land receives biosolids application annually. In addition, the majority of the biosolids are applied to forage and row crops used for animal feed or grains and a small amount is used for fertilizing horticultural or vegetable crops.



Figure 1: Kumar, K., Metropolitan Water Reclamation District of Greater Chicago, Land Application of Biosolids: Human Health Risk Assessment Related to Emerging Contaminants, presentation 01-MAR-2017.

Land application of biosolids results in enhancement of soil health by improving physical, chemical, and biological properties of soil, nutrient recycling, carbon sequestration, and increasing crop productivity by the addition of organic matter to soils. Biosolids have been used on farms and other lands across North America and other parts of the world for the several decades.

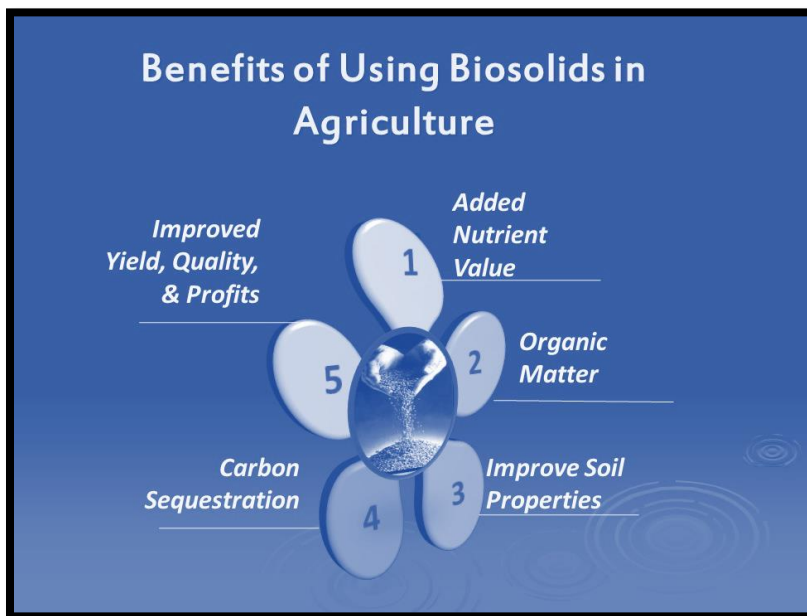


Figure 2: Kumar, K., Metropolitan Water Reclamation District of Greater Chicago, Land Application of Biosolids: Human Health Risk Assessment Related to Emerging Contaminants, presentation 01-MAR-2017.

### Microconstituents in Biosolids

As a result of our modern lifestyle and widespread use of organic chemicals in many applications, large amounts of chemical residues from industries, agriculture, and homes are being continuously released in the environment, some of which may find their way into municipal wastewater. A few examples of microconstituents that may be found in the environment are pharmaceuticals, personal care and consumer products (PPCPs), pesticides, cleaning materials, chemicals used in building materials, additives in foods and drinks, chemicals used for printing, and chemicals used in the manufacturing of housewares, electronic goods, transportation, sports, laboratory, and educational materials. Unlike heavy metals, sources of these chemicals, especially from PPCPs manufacturing and use, in municipal wastewater are diverse, and source control programs that proved effective for heavy metals, are generally not effective in reducing the levels of microconstituents reaching water resource recovery facilities (WRRFs). Although many microconstituents that reach the WRRFs are destroyed through wastewater treatment and sewage sludge processing, some recalcitrant

microconstituents and their metabolites may pass through the treatment process intact and may end up in the effluent or biosolids. Lipophilic (fat soluble) microconstituents show high affinities for organic carbon and preferentially partition into biosolids during solids separation and are inherently less bioavailable than the hydrophilic (water soluble) which may reach the aquatic environment via effluent discharges to receiving streams.

In general, wastewater influents contain microconstituents in concentrations ranging from nano-g/L to micro-g/L, in effluent from non-detect to nano-g/L, and in biosolids the concentrations vary from micro-g/kg to mg/kg.

### Dissipation of Microconstituents after Biosolids Application to Soils

Dilution, mineralization, and strong binding with soil matrices are the three pathways which reduce the bioavailability of microconstituents when biosolids are land applied. In general, there is 100 to 200-fold dilutions of biosolids-borne microconstituents in soil when biosolids are applied at an agronomic rate of 5 to 10 t/ac or 10,000 to 20,000 lbs/ac and incorporated in 6-inch surface layer of soil weighing approximately 2 million-lbs. Mineralization of microconstituents and their binding to soil matrices may be considered as detoxification or decontamination because the bound fraction is often unavailable for plant uptake, leaching, and microbial metabolism. Several abiotic processes resulting from interactions between microconstituents and soil matrices, including hydrophobic partitioning, covalent bonding, ligand exchange, migration to and entrapment into micro-sites, and ionic bonding, may determine the magnitude and strength of bound residues of microconstituents in the soil matrix. The relative prevalence of these mechanisms is influenced by the characteristics of the microconstituents and soil matrices, their concentrations, and the duration of exposure (aging) in the soil. In general, these three processes reduce the bioavailable fraction of microconstituents in soil to very small concentrations.

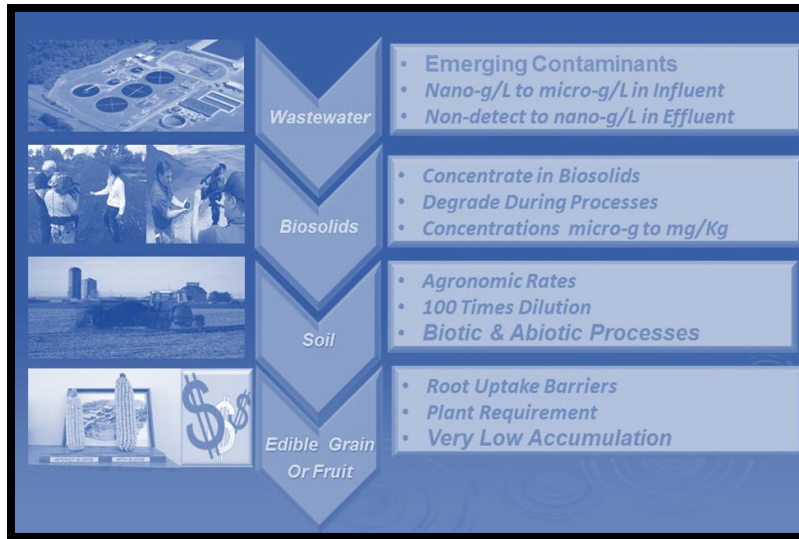


Figure 3: Kumar, K., Metropolitan Water Reclamation District of Greater Chicago, Land Application of Biosolids: Human Health Risk Assessment Related to Emerging Contaminants, presentation 01-MAR-2017.

### Exposure to Microconstituents via Food Crops Grown in Biosolids Amended Soils

The transpiration of water is the main driving mechanism for uptake and transport of microconstituents in plants with properties of microconstituents playing a vital role in determining their bioaccumulation in edible portions of plants (Kumar and Gupta, 2016). In general, review of published data on plant uptake of microconstituents from manure or other by-products amended soils and the State-of-the-science review conducted by WERF (Higgins et al., 2010) on trace organics in biosolids-amended soils show that:

- microconstituent bioaccumulation in edible parts from actual field studies <from pot studies <from hydroponic studies.
- microconstituents were not detected in most of the grains of row crops grown under field conditions.
- The potential for microconstituents to enter edible parts of vegetables and fruit crops was generally low under normal farming conditions when biosolids were land applied following typical agronomic practices.

Prosser and Sibley (2015a,b) conducted an extensive review of the literature for studies that reported residues of microconstituents in the

edible tissues of plants grown on biosolids-amended soils, then used the data to estimate daily intake (EDI) of PPCPs by an adult or a toddler. These EDIs were then compared with acceptable daily intake (ADI) to determine whether PPCPs posed a hazard to human health via the ingestion of contaminated food. The ADI value of pharmaceuticals is the amount of these PPCPs that can be consumed daily over a person's lifespan without causing any adverse effect. These authors computed ADI for pharmaceutical compounds by dividing the lowest daily

therapeutic dose for an adult (mg/day) by a safety factor of 1,000 and dividing by an additional factor of 10 was applied if the PPCPs belonged to an 'endocrine disruptors' group. ADI values for other microconstituents were computed by applying a safety factor of 300 to the no observable adverse effect level (NOAEL). Prosser and Sibley's assessment, using the above-mentioned conservative approach, indicates that consumption of trace concentrations of microconstituents via crops grown on biosolids-amended soils represents a *de minimis* risk to human health.

### Quantitative Human Health Risk Analysis for Microconstituents

Recently, the Northwest Biosolids (NW Biosolids 2015) conducted a quantitative exposure assessment for land application of biosolids using the general risk assessment methodology outlined by the USEPA. The following scenarios of exposure to microconstituents in biosolids from dermal contact and incidental ingestion were evaluated:

- Child exposed while playing in a home garden or lawn fertilized with Class A biosolids compost.
- Adult gardener exposed while working in a home garden fertilized with Class A biosolids compost.
- Occupational worker exposed while applying Class B biosolids to agricultural land.

- Adult hiker exposed while hiking in a forested area fertilized with Class B biosolids.

In addition to risk analysis, they also conducted a series of comparative risk calculations and exposure comparisons to facilitate the communication of risk results; for example, exposure to microconstituents in biosolids were measured against exposure to the same microconstituents from the common use of PPCPs containing those microconstituents (see graphics below). For example, it may take hundreds or thousands of years of exposure to

personal care products, (ii) Pesticides/fungicides/herbicides, (iii) Brominated flame retardants, (iv) Surfactants, (v) Plasticizers, and (vi) Perfluorochemicals.

### Pharmaceuticals and personal care products (PPCPs)

**Antibiotics and Drugs:** It may take thousands of years of exposure to some antibiotics and over the counter drugs from land applied biosolids for the equivalent daily single dose of these compounds taken orally. Research from many studies show that exposure or hazard of this category of

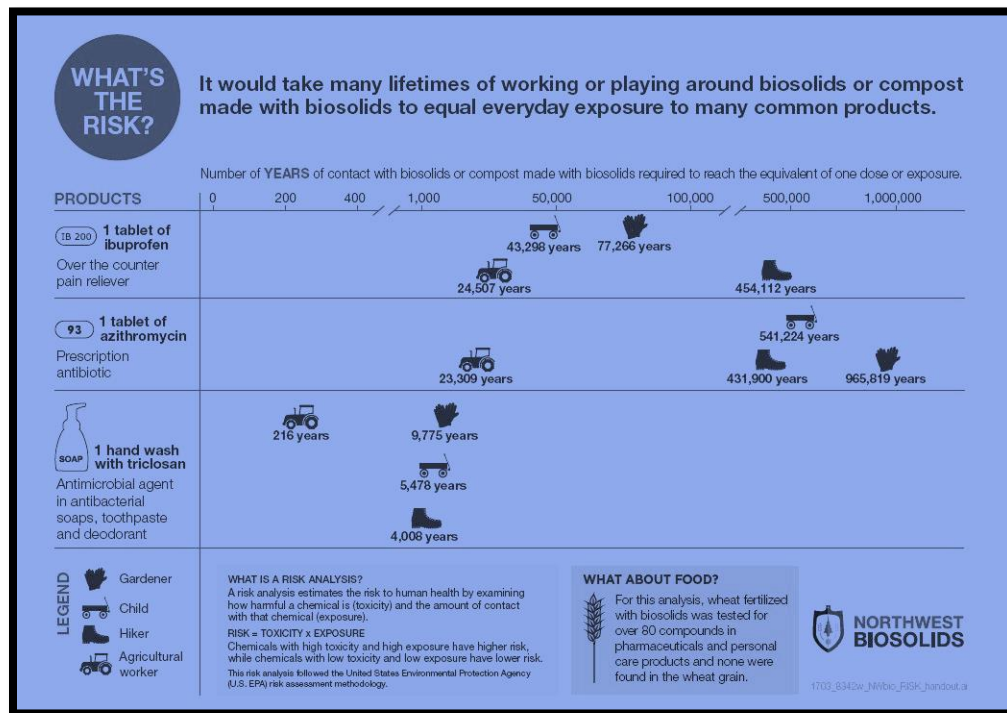


Figure 4: Source: Northwest Biosolids, What's the Risk? 2016. Reprinted with permission.

land applied biosolids to be equivalent to 1 tablet of Ibuprofen, or 1 tablet of the antibiotic azithromycin, or to be equivalent to triclosan from a single hand wash with anti-microbial soap. The results indicated that exposure to microconstituents via land applied biosolids is unlikely to result in any adverse health effects.

### Relative Exposure of Microconstituents from Biosolids and from Other Direct Exposure Pathways

Most microconstituents found in the biosolids and terrestrial environment can be divided into six categories: (i) Pharmaceuticals (antibiotics, hormones, steroids and other drugs) and

microconstituents is minimum from land applied biosolids or composted biosolids and entails minimum risk to human health.

**Antimicrobials:** Antimicrobial compounds triclocarban (TCC) and triclosan (TCS) are commonly added to a wide variety of personal care products (PCPs). TCC and TCS enter the WRRF via routine domestic activities and discharges from hospitals and nursing homes where these chemicals are heavily used as antiseptics. Land application of biosolids can introduce TCC and TCS into the environment. However, risks of human exposure to TCC and TCS via land application of biosolids are minimal because these compounds are tightly bound to

the biosolids matrix and are not taken up by the crops (Xia et al., 2010; Higgins et al., 2011). Dermal absorption and oral ingestion from PCPs are considered to be the major pathways for human exposure because the levels of TCC and/or TCS in antibacterial soaps and toothpastes are much higher (e.g., >6,000 ppm in bar soaps and 3,000 ppm in some toothpastes) than the levels generally observed in biosolids. Detectable levels of TCC have been observed in urine samples of people up to 72 hours after showering or bathing with antibacterial soaps, showing dermal absorption of TCC from bar soaps even after a single use. FDA banned these compounds in 2016 from consumer products. Many companies stopped adding these compounds to anti-microbial soaps after 2013 when FDA announced the rulemaking efforts, and concentrations of these compounds in biosolids have gone down significantly in the last 3 years.

**Hormones and Steroids:** Hormones and steroids are released into the environment from animal and human excrement. In the U.S., about 49 tons of hormones are excreted annually into the environment by farm animals alone. Humans excrete natural hormones 17 $\beta$ -estradiol (E2), estrone (E1), and testosterone, which enter the wastewater stream via domestic discharges. Unlike animal excrement, human excreta may also include a synthetic estrogen 17 $\alpha$ -ethynylestradiol (EE2), which is used in oral contraceptives. It has been observed that large proportions (>98%) of E1 and E2 entering the WRRF are removed during the activated sludge process. Only 90 percent of EE2 is removed because it degrades slowly during the activated sludge process, and traces can be detected in biosolids. Trace levels of natural and synthetic hormones and steroids could be released into the soil after land application of biosolids. However, these contaminants are easily degraded by commonly occurring microbial populations in agricultural soils with half-lives of the hormones ranging from only 1 to 10 d (Higgins et al., 2010).

**Synthetic Musk:** Polycyclic and aromatic nitro musks are commonly used in perfumes. It was later discovered that aromatic nitro musks were unstable in light and alkaline media, so polycyclic musks (PCMs) gained popularity and are widely used in air fresheners and other PCPs. PCMs have been shown to be toxic to biota and

are linked to endocrine disruption and increased breast cancer in humans. Measurable levels of PCMs have been detected in human blood, breast milk and newborn babies. Use of fragranced PCPs has been considered a major source of human exposure. HHCB (Glaxolide) and AHTN (Tonalide), the most commonly used PCMs, are ubiquitous in the environment and have been detected in biosolids and house dust at similar levels. PCMs enter the wastewater stream via domestic activities and indoor dust. A large portion of the PCMs entering the WRRF are eliminated during the wastewater treatment processes and anaerobic digestion of biosolids. A small fraction may exit the WRRF in the final effluent and the remainder is partitioned into biosolids. Irrigation with effluent and fertilization with biosolids may introduce PCMs to agricultural soils. While HHCB has been shown to degrade fairly rapidly in soil, AHTN tends to persist in the environment with a half-life of greater than 180 days (Higgins et al., 2010). AHTN, being highly lipophilic, is strongly bound to the biosolids and soil matrices and is not expected to be mobile or bioavailable in agricultural soils. Reducing direct exposure may be more important to protect human health.

### Pesticides/Fungicides/Herbicides

Toxic and persistent chemicals are present in pesticides/fungicides/herbicides commonly used in and around the house, and are frequently detected in high concentrations in indoor air and house dust (Hundal et al, 2011). Trace levels of these chemicals are also frequently detected in agricultural soils and in municipal biosolids. Although land application of biosolids may potentially add trace levels of these contaminants to the soil, the levels coming from biosolids are negligible in comparison to the soil background levels resulting from regular use of these chemicals for crop production. The addition of organic matter due to the land application of biosolids may reduce the bioavailability of these chemicals in soil. The most significant human exposure pathways for pesticides/herbicides are ingestion of house dust and inhalation of indoor air or exposure from sites where these chemicals have been recently applied (Nigg et al., 1990).

### Brominated Flame Retardants

Polychlorinated flame retardants (PBDEs) are widely used to retard the flammability of many

consumer and industrial products. PBDEs are primarily indoor pollutants and are generally found at high levels in dust and air in the homes and at the workplace. Concentrations of PBDEs detected in house dust are much higher than the levels generally reported in biosolids. In addition to routine domestic activities and industrial input, the PBDEs may also enter the wastewater stream via leachate from municipal solid waste landfills because the vast majority of consumer products containing PBDEs are ultimately disposed of in the landfills. PBDEs are similar to polychlorinated biphenyls (PCBs) and have been shown to be persistent in the environment. They are ubiquitous in soil, water and air and are widely found in people and wildlife. The presence of PBDEs in the environment and humans is of serious concern because some PBDEs are potent endocrine disruptors. During the wastewater treatment process, PBDEs preferentially partition into biosolids due to their lipophilic nature. It has been shown that levels of PBDEs in soils increase after application of biosolids. However, the land application of biosolids is not considered a major exposure pathway because PBDEs have strong affinity for soil organic matter and tend to accumulate in the biosolids incorporation zone (6- to 8-inch surface layer). The PBDEs in the land applied biosolids are not taken up by crops and have minimal risks of translocation in the food chain (Xia et al., 2010; Hale et al., 2012). Risk assessment studies show that diet and ingestion of house dust are the major sources of PBDEs exposure to adults and children, but mother's milk is the major source of exposure to infants due to high body burdens of nursing mothers in North America.

## Surfactants

Surfactants, like alkylphenol ethoxylates (APEs), are added as emulsifiers in PCCPs. Some APEs, especially nonoxynol-9, have spermicidal properties and are used in contraceptives. APEs are also used as antioxidants in the polymer and food industries. These contaminants are ubiquitous in the environment and their levels in the surface waters are increasing. APEs and their degradation products have received considerable attention due to their endocrine disruption effects in the environment. Nonylphenols (NPs), the raw material for making APEs, as well as their degradation product 4-nonylphenol 4-NP, have been implicated in fish feminization in rivers. Interestingly, the estrogenic

effects of NPs in the environment have been known since 1938, but NPs are still widely used in consumer and personal products. A variety of APEs enter the WRRF via routine domestic activities and industrial discharges. APEs are generally degraded into shorter chain NPs like 4-NP during the wastewater treatment process, a small fraction of which may exit the WRRF in the final effluent. But the majority is removed by partitioning into biosolids. Some surfactants in biosolids may occur in concentration greater than found in house dust. However, human exposure to house dust is much greater than to biosolids. Also, NPs and 4-NP degrade rapidly (half-life = 3 to 30 d) in agricultural soil after land application of biosolids (Xia et al., 2010). There has been no report showing deleterious effects of APEs on human or environmental health following land application of biosolids.

## Plasticizers

**Phthalates:** Phthalates are synthetic chemicals of increasing concern because of their endocrine disruption effects. Phthalates are commonly added to plastics to increase their flexibility and transparency. They are used to soften polyvinyl chloride (PVC). Most widely used phthalates are di(2-ethylhexyl) phthalate (DEHP), benzyl butyl phthalate (BBP), diethyl phthalate (DEP) and di-n-butyl phthalate (DBP). Phthalates are used in industrial applications as well as in PCCPs, children's toys and feeding bottles, pharmaceutical applications, and medical devices. Phthalates are not chemically bonded to the plastics or PVC and can be easily released in the environment via volatilization or leaching. They could also migrate into food from plastic containers. Many phthalates, especially DEHP and BBP, are ubiquitously present in air, water, soil and biosolids. High concentrations of phthalates have been observed in indoor air and house dust. Exposure to phthalates could occur through direct use (PCCPs, medical devices, etc.) or indirectly via environmental contamination. In the general population, oral intake is considered to be the main route of exposure because phthalates can easily migrate into food and beverages from the containers and wrappers. Levels of phthalates have been shown to be higher in young children as compared to other age groups. In addition to mother's milk and canned food, ingestion of house dust, inhalation of indoor air, and dermal absorption are other significant routes of exposure to phthalates in

young children who spend most of their time indoors, play close to the floor, and have frequent hand to mouth contact. In 2008, Congress permanently banned three types of phthalates: DEHP, DBP, and BBP in any amount greater than 0.1 percent (computed for each phthalate individually) in children's toys, and any child care article that is designed or intended by the manufacturer to facilitate sleep or the feeding of children age 3 and younger, or to help children age 3 and younger with sucking or teething. Congress has also banned on an interim basis three additional phthalates DINP (diisononyl phthalate), DIDP (diisodecyl phthalate), and DnOP (dioctyl phthalate) and directed the U.S. Consumer Product Safety Commission to convene a chronic hazard advisory panel on phthalates (CPSIA, 2008). However, other phthalates are still being used in children's toys. Phthalates enter the wastewater stream via industrial and domestic discharges due to their widespread use. Land application of biosolids could introduce phthalates into the soil environment. However, they are not persistent in the soils and are degraded fairly quickly with half-lives ranging from 20 to 25 d (Higgins et al., 2010). Land application of biosolids is not considered to be a significant source of phthalate exposure in humans (Hundal et al., 2011).

**Bisphenol A:** Bisphenol A (BPA) is a chemical intermediate used to make epoxy resins and polycarbonates. Free BPA (loose individual molecules), which has a much higher exposure potential than the BPA bound into resin or polycarbonates, is found in high concentrations (8 to 17 g/kg) in carbonless copy paper and thermal paper widely used for credit card and cash register receipts. On average, 0.2 to 0.6 µg BPA could be transferred to fingers upon contact with the paper, and the amount transferred could increase by ten times in cases of wet or greasy fingers (Biedermann et al., 2010). BPA is an endocrine disruptor compound and its estrogenic effect has been known since the 1930s. Considerable levels of BPA have been observed in indoor air and house dust. Levels observed in house dust are greater than the levels detected in biosolids. Measurable levels of BPA (<10 – 646.5 ppb) have also been reported to migrate into food and beverages due to leaching from plastic packaging and BPA lined cans and plastic containers. According to the Center for Disease Control and Prevention (CDC,

2010), nearly 95 percent of Americans have high levels of BPA (>0.1 µg/L urine) in their system. Concerns over the harmful effects to infants were heightened by the fact that infants and children are expected to have the highest daily intake of BPA via release from baby bottles, pacifiers, ingestion of house dust, and inhalation of indoor air that contain considerable amounts of BPA. Use of BPA in baby bottles and toys has been banned in many countries including several states in the U.S. to minimize exposure to infants. BPA could be released into the soil via land application of biosolids. However, BPA has been shown to easily degrade under field conditions with an average half-life ranging from 1 to 10 d. Thus, land application of biosolids is not a significant pathway for human exposure to BPA.

### Perfluorochemicals

Perfluorochemicals (PFCs) (more commonly being referred to as Per- and Polyfluoroalkyl Substances (PFASs)), especially perfluorooctane sulfonates (PFOS) and perfluorooctanoic acid (PFOA), have been used in industrial and consumer products since the 1950s. PFOA is also used in the production of Teflon and Gore-Tex. PFCs can be released into the environment from the manufacture of fluorinated chemicals and losses from PFCs-treated consumer products and eventually enter the wastewater stream. PFOA and PFOS were the most prominent PFCs detected in indoor air, house dust, and biosolids. They are also detected in low concentrations in the blood of wildlife and humans around the world. Exposure to PFOS and PFOA may result from the intake of contaminated food, including fish and water. The most significant human exposure results from ingestion of indoor air and house dust because the largest volume of PFCs (>2.5 million pounds in 2000) is used for indoor applications. Use of PFCs in food contact wrappers and boxes represent another potential source of oral exposure. PFOA is present in microwave popcorn bag paper at amounts as high as 300 µg/kg. According to the U.S. Food and Drug Administration, microwavable popcorn bags alone could account for about 20 percent of the PFOA levels measured in an individual consuming 10 bags of popcorn a year (US FDA, 2012; Egeghy and Laober, 2011; Trudel et al., 2008). These compounds were banned in US food packing papers in 2016. Ingestion of house dust and inhalation of indoor air are the major pathways for PFOS and PFOA exposure to

toddlers and children because they spend greater than 90 percent of their time indoors, exhibit the highest hand-to-mouth frequency and may ingest 100 to 200 mg of dust per day (Trudel et al., 2008; Langer et al., 2010; Hundal et al., 2011). Land application of biosolids may release trace levels of PFCs into the agricultural soils but it doesn't seem to be a major source of human exposure (Hundal et al., 2011; Blaine et al., 2013).

## Conclusions

Diet, lifestyle, ingestion of house dust and inhalation of indoor air are the major sources of microcontaminant exposure to humans (Hundal et al., 2011). Land application of biosolids may only account for minor exposure to some microcontaminants at the most. On an average, 7.2 million dry tons of biosolids are produced in the U.S. annually and only 55 percent are land applied. Less than 0.12 % of the nation's total cropland receives biosolids application. This leads to the logical conclusion that only a small fraction of the total population consumes biosolids-fertilized crops and resides in the vicinity of biosolids-fertilized farmland. Therefore, the land application of biosolids alone cannot account for a significant amount of human exposure to microcontaminants (USEPA, 1995).

Human exposure to biosolids-derived microconstituents would be expected to mainly occur via ingestion of biosolids fertilized soil, consumption of grains, produce, meat and dairy raised on biosolids-fertilized feed, fish from ponds adjacent to biosolids-fertilized fields, and ground or surface waters impacted by land application of biosolids. Both state and Federal biosolids land application regulations and management practices are designed to be very conservative and highly protective of human and environmental health. Strict adherence to these management practices and loading rate restrictions are protective because the biosolids-derived microconstituents have low bioavailability and are not very mobile in the soil profile. Lipophilic microconstituents like PBDEs are not generally taken up by the plants. Less lipophilic microconstituents tend to accumulate in vegetative parts of the plant and are generally not detected in grains, which further limit their translocation into the food chain. These arguments strongly suggest that land application of biosolids would not be a major pathway for human exposure to microconstituents.

Furthermore, experience with similar organic chemicals from Part 503 Risk Assessment shows that risk to humans is *de minimis*.

As a society, our exposure to microconstituents can be reduced by being smart consumers. Uses of antimicrobials in personal care products, excessive use of PBDEs and APEs in consumer products, and indiscriminate use of phthalates, BPA and PFCs in personal care and consumer products are unnecessary. Simply avoiding or minimizing use of such products can greatly reduce environmental contamination and human exposure. Also, regulatory agencies could help in reducing the environmental burden by banning unnecessary and indiscriminate use of microconstituents (recent bans on triclosan, triclocarban, and perfluorochemicals are good steps in the right direction) and by promoting non-toxic biodegradable alternatives.

## Further Reading

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## Additional Resources

- [National Biosolids Partnership](#)
- [U.S. Environmental Protection Agency](#)
- [Water Environment Federation](#)
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## Additional Notations:

This paper does not include a thorough assessment of impacts to other species.

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For further Biosolids information, please see <http://www.biosolids.org>.

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