

EVALUATING EXPOSURE RISK TO TRACE ORGANIC CHEMICALS IN BIOSOLIDS

Due to widespread use of TOrCs in manufacturing of personal care and consumer products, research finds that the greatest human exposure is in the household environment and not via land application of biosolids.

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AS a result of our modern lifestyle and widespread use of organic chemicals in personal care and consumer products (PCCPs), large amounts of chemical residues from industries and homes are being continuously released into the wastewater stream, some of which may find their way into the environment. Unlike heavy metals, sources of trace organic chemicals

(TOrCs), especially from manufacturing and use of PCCPs, in the wastewater stream are diverse, and source control programs that proved effective for heavy metals are futile in reducing the levels of TOrCs reaching a wastewater treatment plant (WTTP). Although most TOrCs that reach the WTTPs are destroyed through wastewater treatment and sludge processing, some recalcitrant TOrCs and their metabolites may pass through the treatment process intact. Lipophilic (fat soluble) TOrCs show high affinities for organic carbon and preferentially partition into biosolids during solids separation and are inherently less bioavailable than the hydrophilic (water soluble) TOrCs in the soil-water environment.

Our dependence on chemicals has significantly increased over the past 30 to 40 years. Recent innovations in chemical processes associated with technological advances in manufacturing have brought hundreds of thousands of new PCCPs into the market, which have improved the quality of our daily lives. However, excessive use of chemicals in the PCCPs has caused pollution of the environment and many pollutants have been detected in our food (e.g., fish, meat, milk). In addition, detectable levels of many TOrCs have been reported in breast milk, body fat and serum of humans. This begs an important question: What is the major exposure pathway by which humans are exposed to TOrCs?

Some reports suggested that dietary exposure is the most likely route of human exposure but others opined that many TOrCs may enter the terrestrial environment via land application of biosolids and then find their way into the



food chain. Thus, a better understanding of the major sources of TOrCs is critical to devise effective strategies for minimizing human exposure. However, the question remains: Is land application of biosolids a

significant source of commonly detected TOrCs in humans? This question can be answered by evaluating the extent of biosolids land application in the country and comparing the levels of TOrCs found in biosolids with other matrices that have the potential to result in human exposure.

COMMERCIALLY USED CHEMICALS

There are over 87,000 commercially used chemicals in the U.S., with an additional 2,000 introduced to the market annually. Over 3,300 of these chemicals (excluding polymers) are high production volume (HPV) chemicals with annual production and/or importation volumes exceeding one million pounds. These chemicals are important ingredients in beneficial and essential PCCPs but become pollutants of concern when they enter the environment because some of these chemicals may exhibit toxicity or produce subtle effects impacting the well-being of species within an ecosystem.

Most TOrCs found in the environment can be divided into seven categories:

- Personal care products
- Pesticides/fungicides/herbicides
- Brominated flame retardants
- Surfactants
- Plasticizers
- Hormones and steroids
- Perfluorochemicals

Common sources of these TOrCs are summarized in Table 1. Their significance in the environment is as follows:

Table 1. Commonly used trace organic compounds, their use, examples of personal care, consumer and household products containing these compounds

Name	Common Use	Common Products
Triclocarban, Triclosan	Antimicrobial	Bar soaps, body washes, cleansing lotions, toothpastes, toothbrushes, mouthwashes, deodorants, hand sanitizers, dish detergents and wipes, cutting boards, kitchen utensils, bed sheets, socks, garbage bags
AHTN, HHCB	Synthetic musk	Cosmetics, deodorants, body perfumes, shampoos, laundry softeners, detergents, air fresheners
Herbicides, fungicides, insecticides	Weed, fungus, and insect control	Roach and ant killer sprays, lice and scabies treating shampoos and lotions, lawn care products
PBDEs	Flame retardant	Home furnishings, upholstery, sofas, mattresses and chairs, toys, baby cribs, car seats, strollers, textiles, electronics (computers, TVs), building materials (drywall, insulation), plastics used in car and aircraft components
Surfactants	Emulsifiers	Shampoos, soaps, contraceptives, detergents, cleaners, paints, metalworking fluids, asphalt emulsions, dust control agents, herbicides
Phthalates	Plasticizer	Vinyl curtains, upholstery, floor tiles, cleaning materials, food containers and wrappers, lotions, shampoos, moisturizers, perfumes, hair sprays, eye shadow liners, nail polishes, liquid soaps, hair sprays, children's feeding bottles, toys (squeeze), teething, polymer coating in oral medications, catheters, IV tubing, blood transfusion devices
Bisphenol A	Plasticizer	Electronic equipment, electrical laminates for printed circuit boards, digital media (CDs, DVDs), automobiles, sports safety equipment, reusable food and drink containers, lunch boxes, baby and water bottles, composites, paints, adhesives, protective liners in metal cans
PFOA, PFOS	Nonstick coating	Cookware, shampoos, lotions, eye make-up, facial moisturizer, lipstick, dental floss, nail polishes, shaving cream, film and denture cleaners, fire-fighting foams, soil, stain and grease resistant coatings on carpets, textiles, paper, and leather, semiconductors, insecticides, wax, polishes, paint, varnishes, protective coatings on medical devices and food-contact packaging, fast food and candy wrappers, pizza boxes, microwavable popcorn bags

PERSONAL CARE PRODUCTS

Antimicrobials: The antimicrobial compounds triclocarban (TCC) and triclosan (TCS) are HPV chemicals commonly added to a wide variety of PCCPs. TCC and TCS enter the WTPP via routine domestic activities and discharges from hospitals and nursing homes where these chemicals are heavily used as antiseptics. A portion of TCS is biologically degraded during the wastewater treatment process, while the majority of the remaining TCS and most of the TCC are removed from the water by partitioning into biosolids. Only a small fraction (parts per billion levels) exit the WTPP in the final effluent. Both TCC and TCS have been suspected of causing endocrine disruption and altering growth and development in animals. The presence of TCC and TCS in the environment may also result in the development of antimicrobial resistance in bacteria.

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 be-

cause these compounds are tightly bound to the biosolids matrix and are not taken up by the crops (Xia *et al.*, 2010). Dermal absorption and oral ingestion from PCCPs 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 (Table 2). 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.

Synthetic Musks: 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 PCCPs. 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 PCCPs has been considered to be the 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 WTPP is eliminated during the wastewater treatment processes and anaerobic digestion of biosolids. A small fraction may exit the WTPP 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 in agricultural soils.

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. Trace levels of these chemicals are also frequently detected in agricultural soils and in municipal biosolids (Table 2). 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. Interestingly, the addition of organic matter due to the land application of biosolids may actual-



ly 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 (Nigg *et al.*, 1990).

BROMINATED FLAME RETARDANTS

Polybrominated flame retardants (PBDEs) are widely used to retard the flammability of many consumer and in-

dustrial products. PBDEs are primarily indoor pollutants and are generally found at high levels in dust and air in homes and at the workplace. Concentrations of PBDEs detected in house dust are much higher than the levels generally reported in biosolids (Table 2). In addition to routine domestic activities and industrial input, the PBDEs may also enter the wastewater steam via leachate from municipal solid waste landfills because the vast majority of consumer products containing PBDEs are ultimately disposed of in 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 (Hites *et al.*, 2004). 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). 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 (i.e., nursing mothers have so much PBDEs accumulated in their bodies that it gets in the breast milk, exposing infants to PBDEs as soon as breast feeding begins) (Jones-Otazo *et al.*, 2005).

SURFACTANTS

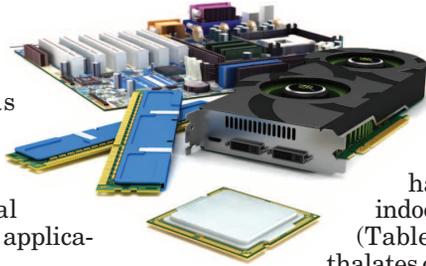
Surfactants, like alkylphenol ethoxylates (APEs), are added as emulsifiers in PCCPs. Some APEs, especially nonoxynol-9, have spermicidal properties and are used in contraceptives. APEs are also used as antioxidants in the polymer and food industries. These contaminants are ubiquitous in the environment and their levels in surface waters are increasing. APEs and their degradation products have received considerable attention due to their endocrine disruption effects in the envi-



ronment. 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 NP in the environment have been known since 1938, but it is still widely used in consumer and personal care products.

A variety of APEs enter the WTTP 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 WTTP in the final effluent. But the majority is removed by partitioning into biosolids. Some surfactants in biosolids may occur in concentrations greater than found in house dust (Table 2). However, human exposure to house dust is much greater than biosolids. Also, NPs and 4-NP degrade rapidly (half-life = 3 to

30 days) in agricultural soil after land application of biosolids. 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 (Table 2). 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.

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 days (Higgins *et al.*, 2010). Land application of biosolids is not considered to be a significant source of phthalate exposure in humans.

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 (from less than 10 (detection limit) to 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), nearly 95 percent of Americans have high levels of BPA (>0.1µg/L urine) in their system.

BPA has been suspected to have adverse effects on fetal and infant brain development. 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 days. Thus, land application of biosolids is not a significant pathway for human exposure to BPA.

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 hor-



mones 17 β -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 α -ethynylestradiol (EE2), which is used in oral contraceptives. It has been observed that large proportions (>98%) of E1 and E2 entering the WTPP 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 days (Higgins *et al.*, 2010).

PERFLUOROCHEMICALS

Perfluorochemicals (PFCs), 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 PCCPs and eventually enter the wastewater stream. PFOA and PFOS were the most prominent PFCs detected in indoor air, house dust and biosolids (Table 2). They are also detected in low concentrations in the blood of wildlife and humans around the world. Both PFOA and PFOS have been shown to be toxic and persistent in the environment (Delinsky *et al.*, 2010).

Exposure of PFOA and PFOS to developing fetus and infants is of particular concern because they are found in breast milk and have the ability to cross the placenta barrier. 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 μ g/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

Table 2. Levels of trace organic contaminants in indoor air, house dust and biosolids

Chemical	Concentration		
	Indoor Air (ng/m ³)	House Dust (μ g/g)	Biosolids (μ g/g)
PCCPs¹-Antimicrobials			
Triclosan (TCS)	na	0.03 - 1.83 ²	nd - 133
Triclocarban (TCC)	na	na	0.18 - 441
PCCPs¹-Synthetic Musk³			
AHTN	nd - 107	0.9 - 3.1	0.08 - 27
HHCb	15 - 299	0.7 - 11.4	0.8 - 3.5
Pesticides⁴			
4'-4'-DDD	1 - 3.5	0.2 - 0.718	0.001 - 84.1
4'-4'-DDE	1 - 5.1	0.2 - 0.738	0.001 - 564
4'-4'-DDT	1 - 30	0.3 - 9.61	0.06 - 135
Chlordane	1 - 144	0.3 - 20.57	0.46 - 12
Chlorpyrifos	1 - 92	0.2 - 3.20	nd - 0.529
Diazinon	1 - 550	0.2 - 51.0	nd - 0.151
Dieldrin	2 - 3.0	0.4 - 4.89	0.01 - 53
Heptachlor	1 - 71	0.2 - 0.549	0.05 - 0.55
Lindane	2 - 110	0.4 - 1.04	<0.01 - 70
Methyl parathion	2 - 92	0.3 - 0.992	nd - 0.07
Methoxychlor		0.5 - 12.9	nd - 0.33
Pentachlorophenol	1 - 34	0.3 - 7.96	0.17 - 8490
Permethrin (sum of cis & trans)	1 - 9.1	0.3 - 159.9	nd - 163
Trifluralin	1 - 23	na	nd - 0.235
2,3-Dibromo-1-propanol	1 - 200	0.2 - 42.8	na
4-tert-Butylphenol	3.4 - 290	0.2 - 1.12	na
Brominated Flame Retardants			
PBDEs (sum of 47, 99, and 100)	0.001 - 1.6 ⁵	0.3 - 35.76	0.14 - 10.1
Tetrabromobisphenol (TBBPA)	na	0.01 - 1.48 ⁶	nd - 0.6
Surfactants			
4-Nonylphenol	21 - 420	1 - 8.68	450 - 2530
Nonylphenol ethoxylates (sum of mono & di) ⁷	4 - 99	0.2 - 64.9	25.7 - 135
Nonylphenol ethoxycarboxylate	nd - 18	0.3 - 9.45	6.1 - 168.4
Octylphenol ethoxylates (sum of mono & di) ⁷	8 - 170	0.2 - 4.11	6.7 - 12.6
Plasticizers⁸			
Diethyl phthalate (DEP)	130 - 4300	4 - 111	0.09 - 3780
Di-n-butyl-phthalate (DBP)	52 - 1100	24 - 352	0.07 - 3210
Benzyl butyl phthalate (BBP)	31 - 480	3.8 - 1310	0.05 - 12800
Di(2-ethylhexyl) phthalate (DEHP)	59 - 1000	16.7 - 7700	0.42 - 58300
Di(2-ethylhexyl) adipate (DEHA)	3 - 66	0.9 - 391	nd - 0.45
Diisobutyl phthalate (DIBP)	11 - 990	1 - 39.1	0.08 - 0.346
Bisphenol A (BPA)	208	0.2 - 17.6	0.1 - 4.6
Perfluorochemicals^{9,10}			
PFOS	nd	nd - 18.1	nd - 5.38
PFOA	nd - 2.6	nd - 9.8	nd - 4.78

Unless indicated otherwise, air and dust data taken from Rudel et al. and biosolids data taken from Higgins et al. na = data not found; nd = not detected. ¹PCCPs = personal care and consumer products; ²Geens et al.; ³Air and dust data from Fromme et al.; ⁴Biosolids data from Harrison et al.; ⁵Wilford et al.; ⁶Stapleton et al.; ⁷La Guardia et al.; ⁸All biosolids data except BPA from Jacobs et al.; ⁹Air data from Shoeib et al.; ¹⁰Dust data from Kato et al.

year (Renner, 2006).

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/day of dust. 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.

SUMMARY

Diet, ingestion of house dust and inhalation of indoor air are the major

sources of human exposure to TOrCs. Land application of biosolids may only account for minor exposure to some TOrCs at the most. On an average, 7 million dry tons of biosolids are produced in the U.S. annually and only 60 percent is land applied. Less than one percent of the nation's total cropland receives biosolids application (USEPA, 2010). 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 human exposure to TOrCs.

Furthermore, human exposure to biosolids-derived TOrCs could only occur via ingestion of biosolids fertilized soil, consumption of grains, produce, meat and dairy raised on bio-solids-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 extremely 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 TOrCs have low bioavailability and are not very mobile in the soil profile. Lipophilic TOrCs like PBDEs are not taken up by the plants. Less lipophilic TOrCs 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 could not be a major pathway for human exposure to TOrCs.

Detection of high levels of many TOrCs like PBDEs, DDT and PFCs in polar bears in remote places such as Arctic Canada and Alaska rules out

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land application of biosolids as a major source of TOrCs exposure. The long range transport of TOrCs via air and water could account for wildlife exposure, but the extent of human exposure seems to suggest other sources. Human exposure to priority "legacy" pollutants such as PAHs, PCBs, dioxins and DDT, was primarily from food because dominant sources of these contaminants were outdoors. Recent research has challenged this paradigm for PBDEs, PFCs and plasticizers because extensive indoor deployment of these chemicals in PCCPs resulted in contaminated indoor air and dust. Undoubtedly, the general population's exposure to TOrCs occurs primarily in the home because people spend most of their time inside the home where they are constantly exposed to these contaminants (Nigg *et al.*, 1990).

As a society, our exposure to TOrCs can be reduced by being smart con-

sumers. Uses of antimicrobials in PC-CPs, excessive use of PBDEs and APEs in consumer products, and indiscriminate use of phthalates, BPA and PFCs in PCCPs are unnecessary. Simply avoiding or minimizing use of such products can greatly reduce environmental contamination and human exposure. Also, regulatory agencies could help to reduce the environmental burden by banning unnecessary and indiscriminate use of TOrCs and by promoting biodegradable alternatives. ■

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