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Survey of Organic Wastewater Contaminants in Biosolids Destined for Land Application†

CHAD A. KINNEY,‡
EDWARD T. FURLONG,*,§
STEVEN D. ZAUGG,§
MARK R. BURKHARDT,§
STEPHEN L. WERNER,§
JEFFERY D. CAHILL,§ AND
GRETCHEN R. JORGENSEN§

Department of Chemistry, Eastern Washington University, Cheney, Washington 99004-2440, and National Water Quality Laboratory, U.S. Geological Survey, P.O. Box 25046, Denver Federal Center, Denver, Colorado 80225-0046

In this study, the presence, composition, and concentrations of organic wastewater contaminants (OWCs) were determined in solid materials produced during wastewater treatment. This study was undertaken to evaluate the potential of these solids, collectively referred to as biosolids, as a source of OWCs to soil and water in contact with soil. Nine different biosolid products, produced by municipal wastewater treatment plants in seven different states, were analyzed for 87 different OWCs. Fifty-five of the OWCs were detected in at least one biosolid product. The 87 different OWCs represent a diverse cross section of emerging organic contaminants that enter wastewater treatment plants and may be discharged without being completely metabolized or degraded. A minimum of 30 and a maximum of 45 OWCs were detected in any one biosolid. The biosolids used in this study are produced by several production methods, and the plants they originate from have differing population demographics, yet the percent composition of total OWC content, and of the most common OWCs, typically did not vary greatly between the biosolids tested. The summed OWC content ranged from 64 to 1811 mg/kg dry weight. Six biosolids were collected twice, 3−18 months apart, and the total OWC content of each biosolid varied by less than a factor of 2. These results indicate that the biosolids investigated in this study have OWC compositions and concentrations that are more similar than different and that biosolids are highly enriched in OWCs (as mass-normalized concentrations) when compared to effluents or effluent-impacted water. These results demonstrate the need to better describe the composition and fate of OWCs in biosolids since about 50% of biosolids are land applied and thus become a potentially ubiquitous nonpoint source of OWCs into the environment.

Introduction

As part of normal wastewater treatment, solids, commonly referred to as sewage sludge or biosolids, are produced and subsequently disposed. Biosolids, the term used herein to describe solids produced during wastewater treatment, are organic carbon-rich material that have a beneficial use and must meet U.S. Environmental Protection Agency (USEPA) and, where applicable, local regulations for nutrient, metal, and pathogen content to be suitable for land application (*1*). In the United States, the daily per capita volume of wastewater produced is about 450 L, which contains about 240 mg/L of suspended solids (which are themselves approximately 80% organic matter) (*2*) or the equivalent of about 86 g of organic wastewater solids per person per day. It is estimated that the average wastewater treatment plant (WWTP) produces 240 kg dry weight of solids per million liters of wastewater treated (*3*).

Biosolids have been recognized as a useful soil amendment and source of nitrogen, phosphorus, organic matter, and other nutrients, which can enhance soil physical properties as well as plant yield (*4*, *5*). Because of these soil improvement qualities and the need to dispose of a continuous supply of biosolids, WWTPs produce different forms of biosolid products for agricultural, landscape, and home use. The USEPA estimates that 50% of all biosolids produced in the United States are land applied, with the remainder incinerated or disposed of in landfills (*1*). Among European countries surveyed, an average of 37% of biosolids are land applied on agricultural soils, totaling 2.39×10^6 dry tons per year (*6*). In many cases biosolids are composted with plant material prior to land application. Although land application of biosolids is predominantly on agricultural soil, biosolids also are commonly used in large-scale landscaping, home landscaping and gardens, remediation of abandoned mining sites, and soil-surface revegetation (*7*−*9*).

Organic wastewater contaminants (OWCs) are organic compounds produced to offer improvements in industrial, medical, and household products and applications (*10*) and usually are found in human- and agriculture-derived wastewater. Compounds that can be classified as OWCs include pharmaceuticals, hormones, detergent metabolites, fragrances, plasticizers, and pesticides (*11*). Many OWCs enter and leave WWTPs unaltered or incompletely removed and subsequently have been identified in the environment (*10*−*13*). The average wastewater residence time inside of a typical WWTP ranges between <1 h and a few days, which is shorter than the degradation half-lives of many OWCs (*14*, *15*). Although the average solid retention time, i.e., the duration of solids in the wastewater treatment process, in most WWTPs ranges from a few days to 30 days, depending on the specific treatment process, and generally is greater than that of wastewater, it is still shorter than the degradation half-lives of many OWCs (*2*, *15*). Research to date (2006) primarily has focused on aquatic environments susceptible to wastewater discharge (*11*, *16*, *17*), although less is known about the presence and fate of OWCs associated with biosolids.

At present (2006) little is known about the potential effects of OWCs present in wastewater effluent or biosolids disposed of in surface water or on soil, respectively. However, potential concerns about the environmental presence of OWCs include adverse physiological effects, increased rates of cancer, and reproductive impairment in humans and other animals as well as antibiotic resistance among pathogenic bacteria (*18*−*25*). Impacts on human health of chronic exposure to subtherapeutic concentrations of pharmaceuticals and the potential for OWCs to effect terrestrial and aquatic organisms also are poorly understood (*10*).

Monitoring the concentration of OWCs in WWTP liquid influent and effluent has shown that some OWCs, namely

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* Corresponding author phone: (303)236-3941; fax: (303)236-3499; e-mail: efurlong@usgs.gov.
‡ Eastern Washington University.
§ U.S. Geological Survey.

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pharmaceuticals, are removed from the liquid phase during treatment, with efficiencies ranging from 10 to 100%, averaging 60% (*13*, *26*). The mechanisms of OWC removal, biodegradation, or sorption on activated sludge are not well understood (*19*). Because WWTP influent is segregated into two components, biosolids and liquid effluent, a large fraction of the total OWCs entering WWTPs ultimately could reside in biosolids. Those contaminants that have a large octanol−water partitioning coefficient might partition preferentially into the organic-rich biosolids phase (40−85% organic matter on a dry-wt basis) during wastewater treatment (*27*−*30*). Partitioning of OWCs into the microstructure of biosolids and a focus on discharge of OWCs in effluents could imply, albeit incorrectly, higher OWC removal efficiencies during treatment. It is more likely that biosolid-sequestered OWCs are degraded much more slowly as a result of slow diffusion from microstructures (*31*, *32*).

The research described herein was designed to determine the range of compositions and concentrations of OWCs present in biosolid products produced under different treatments and from those results estimate the possible impacts of land applications of biosolids. Nine biosolids from seven states produced by different processes were analyzed to describe representative ranges of OWC compositions and concentrations in biosolids from wastewater treatment plants in the United States. To accomplish this, OWCs were extracted from the biosolids using accelerated solvent extraction and measured by liquid chromatography−mass spectrometry or gas chromatography−mass spectrometry.

Methods

Biosolids. The biosolids, and the characteristics of the WWTPs that produced them, are described in Table 1. Biosolid products A−F are intended for commercial, homeowner, and municipal use and are available to the public for purchase or free of charge. Products G−I are used in agriculture. All samples were collected directly from the producer, by purchase, or from publicly available repositories. Except for biosolid D, E, and I, two separate samples of each biosolid were collected 3 to 18 months apart and analyzed. Each sample was composited from the available bulk material (bag or mound) in order to collect a more representative sample. All samples were analyzed in triplicate.

Extraction and Sample Preparation. The OWCs were extracted from the biosolids using accelerated solvent extraction (ASE; Dionex-200, Dionex Corp., Sunnyvale, CA, U.S.A.). The number and types of OWCs evaluated in this study were similar to those investigated by Kolpin et al. (*11*). Two different ASE methods were used to extract polar and nonpolar OWCs.

The polar organic compounds were extracted using a method similar to those described elsewhere for sediment and soil (*33*, *34*). About 13 g of biosolid was extracted using ASE for five static cycles (10-min each) at 130 °C and 10 300 kPa using an acetonitrile/water mixture (70:30, v/v). One milliliter of the total extract was filtered using a 0.20-μm syringe filter into a high-performance liquid chromatography (HPLC) vial, and the acetonitrile evaporated under nitrogen. The sample was reconstituted to about 0.95 mL using a 10 mM aqueous ammonium formate buffer; a nicotinamide 2,4,5,6 d_4 solution was added as an injection internal standard, and the final volume of the extract was brought to about 1 mL.

Nonpolar compounds were extracted using an ASE method outlined in ref *35*. About 1 g of biosolid was first extracted at 120 °C with water/isopropyl alcohol (IPA) (50:50, v/v) to extract the bulk of the heat-labile compounds. Moderate temperature was used to avoid hydrolysis and thermal degradation of some compounds (*36*). The same cell then was re-extracted with water/IPA (20:80, v/v) at 200

TABLE 1. Wastewater Treatment Plant Characteristics and Produced Biosolid Product Descriptions and Preparation Techniques

product	location of WWTP	population served ($\times 10^3$ persons)	volume of wastewater (MGD)	treated biosolid produced (dry tons per day)	treatment	preparation	applied form	organic carbon (mg/g)
A	WI	1100	160	164	secondary activated sludge and chlorine disinfection	rotary kiln dried (25−35 min)	beady granule	367
B	CO	1500	160	74	secondary activated sludge and chlorine disinfection	composted (15−20 days) + plant material (32% wet wt)	compost	259
C	TX	N/A[b]	85.4	45	secondary activated sludge and chlorine disinfection	composted (30 days active windows, 3−6 mos. curing) + yard waste (47% wet wt)	compost	205
D	WA	1400	248	0.027	secondary activated sludge and chlorine disinfection	composted with saw dust	compost	380
E	WA	10	1.2	0.61	secondary activated sludge and chlorine disinfection	composted with yard waste	compost	278
F	KS	80	8.9	4.2	secondary activated sludge and chlorine disinfection	air dried biosolid and turned (9 months-1 year)	dry cake	99
G[a]	AZ	320	24.8	26	secondary activated sludge and chlorine disinfection	biosolid (~2% solid material)	suspended solid	311
H[a]	AZ	320	24.8	26	secondary activated sludge and chlorine disinfection	dewatered biosolid + polymer (8−10% solid material)	wet cake	296
I	IA	67	6.5	4.2	secondary activated sludge and chlorine disinfection	thermophilic and mesophilic digestion + polymer (~20% solid material)	wet cake	319

[a] Products G and H were produced by the same wastewater treatment plant. [b] Data not available.

°C into a second collection vial to obtain the more hydrophobic, thermally stable compounds, with 3 mL of pentane added to the vial to minimize problems associated with collecting a hot extract. All extractions were at 13 800 kPa for three 10-min static cycles. When cool, the nonpolar OWCs extract underwent a concentration and cleanup step by using 20-mL solid-phase extraction (SPE) cartridges containing 1 g of modified polystyrene-divinylbenzene (PSDVB) phase (Waters Corp., Milford, MA). The bulk of the water then was removed from the loaded PSDVB SPE phase by vacuum for a few minutes after extract loading had been completed. The SPE cartridge was further dried by applying 2 L/min nitrogen for 15 min. About 4 g of sodium sulfate was placed in the 6-mL barrel of a 1-g Florisil SPE cartridge (International Sorbent Technologies, Mid Glamorgan, U.K.), and the combined sodium sulfate/Florisil cartridges were attached below the loaded PSDVB SPE extraction cartridge prior to elution. The combined SPE stack then was eluted with three 10-mL aliquots of dichloromethane/diethyl ether (80:20). The collected eluent was evaporated under a gentle stream of nitrogen to a final volume of about 1 mL. Internal standards were added prior to transferring the extract to a 2-mL autosampler vial.

Instrumental Analysis. The polar pharmaceutical OWCs were identified and quantified using external calibration by reversed-phase high-performance liquid chromatography coupled with electrospray ionization/quadrupole mass spectrometry (HPLC/ESI/MS) in the positive ion mode (Hewlett-Packard/Agilent Model Series 1100 LC/MSD quadrupole mass spectrometer, Hewlett-Packard/Agilent, Palo Alto, CA, U.S.A.), similar to methods described elsewhere (*37*). Samples were analyzed using selected-ion monitoring to improve sensitivity and to minimize the effect of coeluting chemical interferences, and at least two ions were monitored for each compound.

Nonpolar OWCs were quantified using external calibration on an Agilent Technologies Model 5973 gas chromatograph/mass spectrometer (GC/MS) as described by Burkhardt et al. (*35*). Compounds were separated using a 30-m by 0.25-mm inside diameter fused-silica capillary column coated with a 0.50-μm bonded film of 5% polyphenylmethylsilicone. The mass spectrometer (MS) was operated using electron-impact ionization mode (70 electron volts) and full-scan mode from 45−450 mass/charge ratio (m/z) for 30 min. The mass range scanned was increased from 45 to 550 m/z for the last 10 min to detect brominated flame retardants.

Quality Assurance and Quality Control. Several quality-assurance and quality-control measures were used during the course of this study. At least one laboratory (burned Ottawa sand) spike and laboratory blank were evaluated for each set of extractions and quantifications. Both extraction and quantification methods included the addition of at least one method performance surrogate compound to all samples, spikes, and blanks. In addition, both quantification methods verified detections by determining multiple characteristic ions for each compound and comparing the ratios of these ions and chromatographic retention to authentic standards. Finally, separate samples from the first set of biosolids collected were fortified with known amounts of OWCs (matrix spikes) to assess compound recoveries from each of the sample matrixes (Table S1). Mean triplicate matrix spike recoveries of all compounds determined by GC/MS in this study ranged between 22 and 130%, with a median of 62% and a mean of 63%, corrected for ambient environmental concentrations. Recoveries of pharmaceuticals determined by liquid chromatography/mass spectrometry (LC/MS) ranged from 6 to 205%, with a median of 71% and a mean of 76%, corrected for ambient environmental concentrations. The greater variability associated with pharmaceutical quantification with the LC/MS reflects matrix effects from components that are coextracted during biosolid-sample processing, most likely caused by competition for charge during ionization (*38*). The reported compound concentrations in Table 2 were not recovery corrected. None of the compounds determined in this study were detected in the burned sand laboratory blanks determined with each set of 10 samples, thus indicating no laboratory contamination.

Results and Discussion

In total, 55 of the 87 OWCs were detected in one or more of the biosolids studied. The 55 OWCs detected for each biosolid appear in Table S1 as well as average concentrations, matrix spike recoveries for each compound in each biosolid, and the method detection level for each compound. The classes and individual organic contaminants detected in this study were similar to those detected in surface water by Kolpin et al. (*11*). Biosolid product I, a wet-cake biosolid, had the fewest number of OWCs detected, 30; a granular, kiln-dried biosolid, biosolid A, had the greatest number of detections, 45, in the second of two samples. In fact, 25 of the OWCs were detected in all nine biosolids (Table 2). Most notably there were several pharmaceuticals, detergent metabolites, sterols, synthetic fragrances, and polycyclic aromatic hydrocarbons (PAHs) that were detected in all nine biosolid products. It is evident that despite the variety of biosolid production techniques, the total number of OWCs detected in any one biosolid does not vary greatly between products, suggesting some compositional uniformity of OWCs in biosolid products. For six of the biosolids, a second set of samples was analyzed to gauge the seasonal variability in OWC content and concentration. The pharmaceutical concentration of the six biosolids did vary temporally between the two samples (Table S1); however, there is not a consistent difference, positive or negative, in pharmaceutical concentrations between any two temporally separated samples. These differences may be attributable to variations in the composition of pharmaceuticals entering a WWTP's influent stream or aspects of plant operations producing the biosolids analyzed. The same can be said for the nonpolar OWCs measured in this study (Table S1). The total number of detections does not vary much between the two temporally separated samples for the six biosolids, although there are slight differences among the exact compounds detected between the two samples. While some compositional and concentration differences do exist between the nine biosolids tested, which may reflect the different preparation technologies, it can be stated that the wastewater treatment processes and biosolid preparation methods for the biosolids studied are inadequate to completely remove 25 of the OWCs measured (Tables S1 and 2).

The most commonly detected compounds in these biosolids are pharmaceuticals (prescription and nonprescription), detergent metabolites, steroids, fragrances, and PAHs (Figure 1). Although the most commonly detected OWCs already represent a wide range of compound classes, there are several other classes of compounds detected in these biosolids, such as fire retardants, plasticizers, and disinfectants. Differences in OWC content between the two time-separated samples for each biosolid are minimal. The difference in total OWC content varies by less than a factor of 2 for the two sample sets, and the total number of detections for each biosolid varied by 7 or less between the two samples (Table 2). Variations in a WWTP's influent volume, which may be influenced by precipitation or other operating characteristics, and influent OWC content are likely to contribute to the observed differences between samples from the same treatment facility collected months apart.

Although absolute concentrations vary, the relative composition of compounds from different chemical classes is reasonably consistent among the 9 biosolids tested (Figure 1), so that, despite differences in source and preparation, these nine biosolid products can be described as more alike than different and may reflect the typical OWC class

TABLE 2. Carbon Normalized Concentrations, in μg/kg Organic Carbon, of Organic Wastewater Contaminants Detected in All Nine Biosolids and Their Physical–Chemical Properties.

target OWC	use	log K_{ow}[a]	water solubility (mg/L)[a]	vapor pressure (mmHg)[a]	biosolid A	B	C	D	E	F	G	H	I	median of all biosolids
carbamazapine	antiepileptic	2.45	17.7	1.84×10^{-7}	140	180	15	58	68	51	150	64	1200	68
diphenhydramine	antihistamine	3.27	3.06×10^{3}	3.7×10^{-5}	2900	340	550	32	54	810	150	170	22000	340
fluoxetine	antidepressant	4.05	60.3	2.52×10^{-5}	480	350	130	140	100	2800	370	830	4700	370
d-limonene	fragrance	4.57	13.8	1.98	520	705	390	282	255	2120	744	630	3340	630
tonalide (AHTN)	fragrance	5.70	1.25	5.12×10^{-4}	11000	11600	5200	8950	281	43900	15900	16700	1340000	11600
galaxolide (HHCB)	fragrance	5.90	1.75	5.45×10^{-4}	3900	12300	5070	2460	47	21900	3210	3150	554000	3900
indole	fragrance	2.14	3.560×10^{3}	0.0122	20700	27000	19600	4210	38200	19400	4290	7000	21300	19600
4-tert-octylphenol	detergent metabolite	5.28	5	4.78×10^{-4}	4140	1270	895	4030	4210	2710	5370	4080	3500	4030
para-nonylphenol-total	detergent metabolite	5.92	5×10^{3}	9.42×10^{-5}	261000	190000	68700	491000	2180	229000	434000	473000	1520000	261000
nonylphenol, monoethoxy-total	detergent metabolite	4.17[b]			44600	17200	5320	10800	3960	23900	25800	21600	79400	21600
nonylphenol, dithoxy-total	detergent metabolite	4.21[b]			89000	7010	2850	3250	3000	44600	19800	15500	793	7010
bisphenol A	fire retardant	3.32	120	3.91×10^{-7}	1680	5540	6180	9030	4690	3550	1620	1090	14400	4690
3-beta-coprostanol	steroid	8.82	2.03×10^{-4}	5.47×10^{-10}	221000	50200	72800	47400	8100	126000	355000	325000	1460000	126000
cholesterol	steroid	8.74	0.095	7.79×10^{-10}	402000	66900	86200	157000	19100	236000	386000	333000	209000	209000
beta-sitosterol	steroid	9.65	1.3×10^{-5}	3.77×10^{-10}	110000	50800	131000	200000	112000	257000	138000	112000	554000	131000
stigmastanol	steroid				9310	6760	17400	2760	3140	113000	44600	40500	243000	17400
phenanthrene	PAH	4.52	1.15	1.21×10^{-4}	1090	367	176	342	183	535	159	166	5430	342
anthracene	PAH	4.50	0.0434	6.53×10^{-6}	324	253	56	139	115	359	92	74	1000	139
fluoranthene	PAH	5.20	0.26	9.22×10^{-6}	1090	1200	744	387	2740	1150	166	166	2980	1090
pyrene	PAH	5.00	0.135	4.5×10^{-6}	1310	1420	1120	508	43	1110	186	169	2320	1110
phenol	disinfectant	1.50	8.28×10^{4}	0.35	2180	1970	2900	587	1650	1240	4940	4480	19600	2180
triclosan	disinfectant	4.53	10	6.45×10^{-7}	7270	10200	5360	1170	3000	12100	11100	11400	32900	10200
diethylhexyl phthalate	plasticizer	7.88	0.27	1.42×10^{-7}	31700	12700	7890	4561	3460	3840	27200	21400	10500	10500
para-cresol	preservative	1.97	2.15×10^{4}	0.11	8610	2460	4620	763	989	1520	4900	4400	15600	4400
skatol	fecal indicator	2.60	498	5.55×10^{-3}	5310	2510	4020	1610	1330	7000	986	806	16200	2510

[a] Physical–chemical properties were found from the following online data bases: http://www.syrres.com/ese and http://logkow.cisti.nrc.ca/logkow/search.html. [b] Log K_{ow} values from ref *69*.

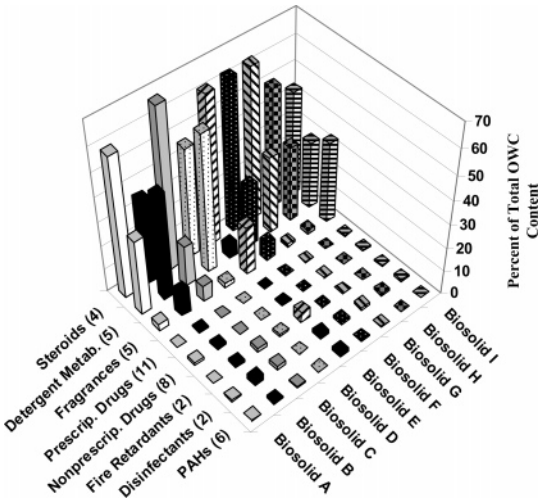

FIGURE 1. The average percentage of the total organic wastewater contaminant content for various classes of organic compounds in each of the biosolid products. The values in the parentheses are the total number of compounds per class that were determined (Table S1).

composition of biosolids produced in the United States. Of the 95 contaminants detected and quantified in the national reconnaissance of OWCs in surface water by Kolpin et al. (*11*), 30 also were detected in these biosolid samples. In addition to OWCs detected in common with Kolpin et al. (*11*), 25 additional OWCs were detected and quantified, including several pharmaceuticals, fragrances, detergent metabolites, PAH, and alkylated PAH (alkyl-PAH). Eight of the nonpolar OWCs (anthracene, benzo[*a*]pyrene, fluoranthene, naphthalene, phenanthrene, phenol, pyrene, and para-cresol) detected in the biosolids of this study also were detected in biosolids produced in Vancouver, BC (*39*). However, the maximum and average concentrations of the eight OWCs consistently were an order of magnitude or higher in the Vancouver biosolids than in the biosolid products measured in this study and may reflect differences in the level of wastewater treatment, biosolid source inputs, and biosolid production processes (*39*).

Concentration of individual OWCs detected in this study varied greatly between biosolids, generally ranging from 10 to 2000 g/kg biosolid dry weight. However, there are numerous examples of compounds that were detected at substantially higher concentrations. In many cases the steroids coprostanol, cholesterol, sitosterol, and stigmastanol and detergent metabolites that were detected exceeded the typical range of concentrations of other OWCs in the biosolids as well as accounted for a large portion of the total OWC content of the biosolids (Table S1 and Figure 1). The compound concentrations observed for these classes ranged between 167 μg/kg and 2×10^5, for 4-*tert*-octylphenol and total para-nonylphenol. Although individual OWCs were detected at microgram-per-kilogram dry-weight concentrations, the total sum of all OWCs measured in this study and detected in a given biosolid approached or exceeded 100 mg/kg dry weight.

At present (2006), it is not known if ambient environmental concentrations of OWCs have acute effects on terrestrial or aquatic organisms, when applied to soil or discharged to surface water. However, it has been demonstrated that some OWCs show endocrine-modulating activity and that exposure to these compounds may result in chronic effects on aquatic organisms at ambient environmental concentrations (*40*, *41*). The eight OWCs monitored in this study that are known or suspected endocrine-modulating compounds are denoted by a superscript "d" in Table S1. These compounds include nonylphenol and octylphenol detergent metabolites that were detected in greater concentrations (low to high milligrams per kilogram) than most other OWCs measured in the biosolids and bisphenol A, diethylhexyl phthalate, benzo(a)pyrene, and benzophenone. The mean and median concentrations of the sum total nonylphenol and total octylphenol detergent metabolites in all biosolid samples were 24 907 and 4100 μg/kg, respectively. The common practice of repeated land application of biosolid products upon individual sites, therefore, could result in substantial localized concentrations of known and potential endocrine-modulating and pharmacologically active contaminants, such as nonylphenol ethoxylates, PAHs, and carbamazepine.

Low levels of antibiotics originating from WWTPs have been directly linked to an increased presence of antibiotic resistance among bacteria in aquatic environments (*23*−*25*, *42*−*44*). We measured the antibiotics erythromycin−H_2O, sulfamethoxazole, and trimethoprim in microgram per kilogram concentrations in the biosolids. Six of the nine biosolids tested had one or more antibiotic present (Table S1). In other studies, a majority of bacteria isolates from some WWTPs demonstrate insensitivity to one or more antibiotics, including species of *Aeromonas, Klebsiellae,* and *Enterobacteriaceae* (*45*−*48*). Once established, resistance to a given antibiotic can be maintained, even in the absence of continued exposure to low concentrations of the antibiotic in the environment (*21*, *49*). An environmental presence of the disinfectant triclosan, commonly observed in these samples at low milligram-per-kilogram concentrations, also has been implicated in leading to antibacterial resistance similar to antibiotic resistance in bacteria (*50*, *51*).

Previous research has identified the presence of one or more classes of OWCs in wastewater effluent, biosolids, and susceptible surface water (*10*, *11*, *39*, *52*, *53*). The compound classes potentially present in biosolids is as diverse as the industrial, pharmaceutical, and personal-care product chemicals that are likely to be disposed through sewage systems, and numerous additional OWC classes warrant investigation; however, to our knowledge, this is the first study to describe and measure such a wide array of OWC classes in representative samples of biosolids destined for land application in the United States. Two OWCs commonly detected in this and other studies are the synthetic musk fragrances tonalide and galaxolide, detected in all nine biosolids. With the exception of biosolid I, which has a much greater concentration than the other biosolids tested, the range and mean concentrations of tonalide and galaxolide in tested biosolids was 78−5800 μg/kg (mean = 3441 μg/kg) and 13−3700 μg/kg (mean = 1461 μg/kg), respectively. This is lower than the concentration observed in the sludge samples reported by Difrancesco et al. (*54*) of 8.1×10^3−17×10^3 μg/kg and 17.7×10^3−51×10^3 μg/kg, for tonalide and galaxolide, respectively. Similarly, nonylphenol ethoxylates were routinely detected in these biosolids, in concentrations ranging from 1.0×10^3 to more than 40×10^3 μg/kg, lower by approximately an order of magnitude or more than concentrations reported in a recent review of organic contaminants in biosolids of 981×10^3 to 1380×10^3 μg/kg (*14*). These concentration differences may reflect the biosolids sources, types, and processing, the latter of which can include dilution with plant material and other OWC-free organic matter.

These results demonstrate that compounds representative of a range of sources—medicinal, industrial, and household—and chemical properties (Table 2) are present in substantial concentrations in biosolids, suggesting that these compounds are incompletely removed during wastewater treatment and sequestered in biosolids that are subsequently land applied. Preferential partitioning of OWCs into the organic carbon rich solid waste stream is recognized as an important removal mechanism from the aqueous phase for many OWCs included in this study. This mechanism is most important for hydrophobic (large log K_{ow}) compounds, such as musk fragrances and detergent metabolites (*14*, *54*−*56*). In contrast, many pharmaceuticals are removed from wastewater via

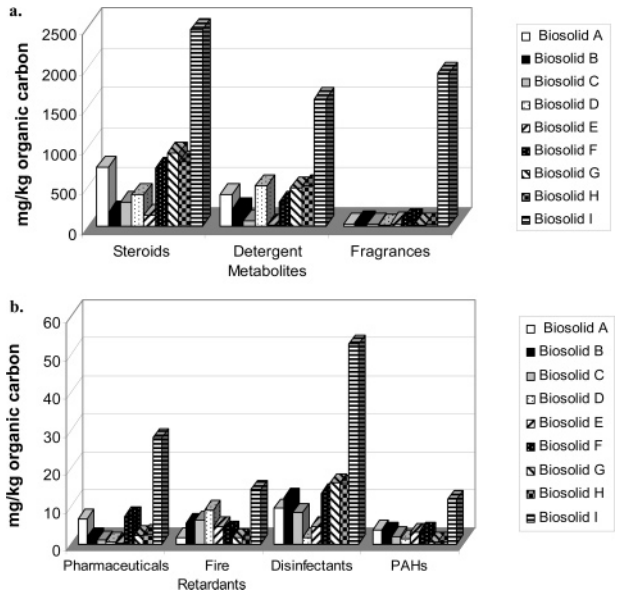

FIGURE 2. Carbon normalized concentrations of organic wastewater contaminants for various classes of organic compounds in each of the biosolid products. The compound class (a) with the greatest carbon normalized concentrations and (b) with moderate and lower carbon normalized concentrations.

biodegradation rather than absorption to solids, but this appears to be a compound-specific observation (*14*, *55*, *56*). Nineteen different pharmaceuticals were detected in these biosolid samples, representing a wide range of physical chemical properties, including compounds with low log K_{ow} and high water solubility values, such as acetaminophen and caffeine, which were detected in eight of nine biosolids (Table S1). The biosolids in this study originate from differing locations and populations, represent at least four distinct preparation techniques, and show a striking compositional uniformity (Tables 1 and 2; Table S1). This suggests that when considering wastewater-derived contaminants, biosolids and wastewater effluent need to be considered as two distinct and equally important sources of OWCs to the environment. Because of the wide variety of uses for biosolids (*8*) and the variety of OWCs found in these biosolids, it is likely that biosolids can constitute a ubiquitous nonpoint source of OWCs to the environment when land applied.

The organic carbon content of each biosolid was measured in an attempt to determine if organic carbon content was a likely contributing factor to the variations in OWC composition and concentration between biosolids. The concentrations of the 25 contaminants that were detected in all nine biosolids were normalized to the organic carbon content of each biosolid (Table 2 and Figure 2). Additionally, other physicochemical properties of the OWCs, such as hydrophobic partitioning (log K_{ow}), water solubility, and vapor pressure, were considered as potential factors affecting their presence in the biosolids tested. Although in many cases the concentrations of the OWCs were similar when normalized to organic carbon, there was almost always one or more outlying value for each OWC, so that the range of values differs by about a factor of 10. When carbon-normalized OWC concentrations of each biosolid were compared to the respective log K_{ow}, for each OWC, only four of the nine biosolids were significantly ($P = 0.05$) correlated, suggesting that organic carbon content is likely not the only factor controlling OWC concentration in biosolids. Other studies indicate that log K_{ow} and water solubility are likely inadequate to predict OWC presence (*13*).

While it is generally expected that compounds with low water solubilities and large log K_{ow} values will more likely be present in organic-rich biosolids compared to highly soluble, polar compounds, our results show that soluble polar compounds, such as the pharmaceuticals detected in this study, also are present at appreciable concentration in all nine biosolids. The 25 OWCs detected in all nine biosolids range in water solubility from 1.3×10^{-5} to 8.28×10^{4} mg/L and in log K_{ow} from 1.50 to 9.65 (Table 2). The presence of OWCs in biosolids with this wide range of physicochemical properties suggests that multiple mechanisms may be responsible for the incorporation of OWCs into biosolids; a similar observation has been made for antibiotics in manure (*57*). It also is probable that other factors not related to the chemical properties of OWCs, such as the quantity of OWCs entering the influent stream (which likely vary from location to location and day to day), the total volume of influent, biosolids/liquid ratios, and sludge retention time contribute to variations in the OWC composition and concentration in biosolids.

Differences in OWC concentrations between the biosolids in part may be attributed to the different preparation methods, such as addition or inclusion of plant material during composting. The addition of plant material effectively dilutes the biosolid samples, while possibly increasing the organic matter content of the biosolid product. Composting has been recognized as an effective means to limit or eliminate some organic contaminants (*58*−*61*), but when the biosolids that are composted are compared to the unamended sludges and granulated bisolid products, the comparable concentrations observed in this study suggest that composting is relatively ineffective at reducing OWC concentrations.

The mass of biosolids applied per unit area of soil surface will have a substantial effect on the potential dispersal of OWCs from biosolids during land application. Biosolid application rates are site specific and are determined based on soil properties, the biosolid nutrient and heavy metal concentrations, and the nutritional needs of the crop or plant cover that will use the biosolid nutrients. Following USEPA guidance on agricultural application rates for biosolids, 10 dry tons per acre or 116 000 gallons wet septage (~2% solid material) would be representative rates for many crop types (*62*). When this representative application rate and the average concentration of OWCs found in Table S1 are used, loadings of about 3.4 kg/acre (0.83 g/m^2) of total OWCs measured in this study result as well as about 0.2 g/acre (49 μg/m^2) of the pharmaceutical carbamazepine, 20 g/acre (4900 g/m^2) of the disinfectant triclosan, 36 g/acre (8900 μg/m^2) of the synthetic musk tonalide (AHTN), and 760 g/acre (16 000 μg/m^2) of para-nonylphenol detergent metabolites. These loading estimates are for a single application; biosolids often are applied multiple times to the same site. Kinney et al. (*34*) evaluated the potential for 19 pharmaceuticals to accumulate in soil where reclaimed water derived from wastewater effluent was used for irrigation and nutrient amendment. The estimated total loading of carbamazepine from reclaimed water during the entire irrigation season described in their paper was 185 μg/m^2, with soil carbamazepine concentrations increasing over the course of the irrigation season. Carbamazepine and other hydrophobic OWCs derived from biosolids and resistant to biodegradation or volatilization would be expected to exhibit similar accumulation in biosolids-amended soil, particularly with repeated application.

This study was not designed to investigate the environmental fate of OWCs originating from land application of biosolids. However, the high frequency of OWC detection in the biosolids tested and the high concentrations of individual OWCs present suggest that biosolids can be an important OWC source to terrestrial environments, and projections about their environmental fate are warranted. Previous research has demonstrated that some compounds, including some synthetic fragrances, pharmaceuticals, and brominated fire retardants, can be persistent once introduced into soil environments (*34*, *54*, *63*). Matscheko et al. (*63*) observed

brominated fire retardants at almost 8000 times higher than background concentrations in soil samples 20 years after the last application of biosolid. Difrancesco et al. (*54*) measured the synthetic musk tonalide (AHTN) and musk ketone to persist for 1 year, whereas the other 20 synthetic fragrances they studied rapidly dissipated in biosolid amended soil, suggesting that at least two fragrances, one of which was routinely detected in these biosolids, are persistent in spite of their volatility (Table 2). Kinney et al. (*34*) measured several pharmaceuticals in soil irrigated with reclaimed water; 15 of the 19 pharmaceuticals, including carbamazepine, were present in soil 6 months after the cessation of the previous irrigation season. Hesselsøc et al. (*64*) and Xia and Jeong (*65*) observed rapid degradation of nonylphenols is soil and biosolids, and Rabølle and Spliid (*66*) monitored rapid and complete leaching of the antibiotic olaquindox through soil. Traina et al. (*67*) used a multiyear field dissipation study and laboratory studies to evaluate the persistence or degradation of polydimethylsiloxane (PDMS) in biosolids-amended plots and in laboratory studies; they showed that soil moisture was the controlling factor in the persistence or degradation of PDMS and that there was no direct effect of biosolids on PDMS persistence or degradation. These previous studies suggest that the brominated flame retardants, fragrances, pharmaceuticals, and surfactant degradates determined in biosolids from this study would be expected to persist or degrade similarly after land application.

Multiple possible mechanisms could lead to persistence or degradation of any OWC detected in biosolids-amended soil. Hydrophobic partitioning into soil organic matter may play a role in the retention of the more nonpolar OWCs, whereas ion exchange, ion bridging with clays, or hydrogen-bonding likely play a role in hydrophilic retention of compounds, such as pharmaceuticals in soil (*14*, *68*). Depending on the specific OWC, mechanisms of degradation removal from soil can include leaching, biotic and abiotic transformations, volatilization, and photolysis (*54*, *56*, *64*, *65*). Physicochemical properties might serve as broad indicators of OWC behavior once introduced into the environment. However, the landscapes in which biosolids are routinely applied in the United States range from arid to humid and from temperate to subtropical, and the ultimate environmental behavior of any biosolid-derived OWC will be likely be highly dependent on the climate and setting of biosolids application, as has been demonstrated by Traina and others (*67*).

Additional research is needed to determine if in fact these results are representative of most biosolids, in the United States or internationally. The U.S. Environmental Protection Agency estimated in 1998 that 6.9 million dry mg of biosolids was produced in the United States, and this is expected to increase to 8.2 million dry mg by 2010 (*8*), of which 50% or more is destined for land application (*1*). In addition to measuring OWC content in a greater number of biosolids, future research needs to address the transport and behavior of OWCs derived from land application of biosolids under realistic field conditions and in laboratory settings. Extended field studies, such as those by Traina et al. (*67*), provide useful models for future field research in the fate of OWCs. Such research might focus on the processes by which OWCs can be mobilized from biosolid products under the typical condition of their application to soil in agriculture, landscaping, and land-surface remediation. Well-designed experiments that identify the salient processes and chemical properties that result in desorption of OWCs from biosolids and into soil and water will provide the information necessary to determine quantitatively the potential for surface water and groundwater contamination from the beneficial use of biosolids and thus the potential for exposure to soil and aquatic organisms.

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Supporting Information Available

Average concentrations of all 55 organic wastewater contaminants detected in one or more of the nine biosolids studied, matrix spike recoveries for each compound in each biosolid, and the method detection level for each compound (Table S1). This material is available free of charge via the Internet at http://pubs.acs.org.

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