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SEPA Biennial Review of 40 CFR Part 503 **As Required Under the Clean Water Act** Section 405(d)(2)(C)

Reporting Period 2011 Biennial Review

Biennial Review of 40 CFR Part 503 As Required Under the Clean Water Act Section 405(d)(2)(C)

Reporting Period Biennial Review 2011

U.S. Environmental Protection Agency Office of Water Office of Science and Technology Health and Ecological Criteria Division Ecological and Health Processes Branch Washington, D.C.

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NOTICE

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EXECUTIVE SUMMARY

In 1993, the U.S. Environmental Protection Agency (EPA) promulgated regulations in 40 CFR Part 503 as amended, setting numeric standards for certain metals in sewage sludge prior to being placed on land, on a surface disposal site, or in an incinerator, requiring vector attraction reduction (e.g., reducing birds, rodents and insects) for pathogens, and establishing operational standards for emissions from sewage sludge incinerators. Section 405(d)(2)(C) of the Clean Water Act (CWA) states that EPA shall review (but not necessarily generate a report) the sewage sludge regulations not less often than every two years for the purpose of identifying additional toxic pollutants and promulgating regulations for such pollutants consistent with the requirements of section 405(d).

In fulfilling this commitment for the 2011 Biennial Review Cycle, EPA collected and reviewed publicly available information. The Agency searched databases with articles published in English and in peer-reviewed and refereed journals for information on occurrence, fate and transport in the environment, human health and ecological effects, as well as other relevant information for pollutants that may occur in U.S. sewage sludge. If such data are available for pollutants that may occur in sewage sludge, the Agency will assess the potential risk associated with exposure to such pollutants when sewage sludge is applied to land as a fertilizer or soil amendment, placed in a surface disposal site, or incinerated.

The 2011 data search identified 23 new pollutants from the potential universe of pollutants, for which some data were available that fit the following criteria: (1) identified in the Targeted National Sewage Sludge Survey (TNSSS; U.S. EPA, 2009) or the open literature as having concentration data for sewage sludge, (2) not currently on EPA's list of potential candidates under evaluation for addition to the Part 503 standards, or (3) not previously regulated or evaluated (e.g., as potentially causing harm to humans or the environment) for sewage sludge. The available exposure or toxicity data are not sufficient at this time for many of the pollutants using current biosolids modeling tools. EPA is in the process of evaluating ten of these chemicals found in the TNSSS and is investigating alternative methods or tools for filling human health and ecological toxicity data gaps, estimating missing source concentration data, and performing screening-level deterministic assessments to estimate human health and ecological risk for biosolids land application scenarios; however, those assessments are still underway. EPA will continue this work subject to availability of resources and overall program priorities. At this time EPA has not identified additional toxic pollutants for regulation under Section 405(d)(2)(C) of the CWA.

CONTENTS

Page

1.	Introduction	1
	1.1 History of the Standards for Use or Disposal of Sewage Sludge	1
	1.2 Biennial Reviews	2
2.	2011 Biennial Review	4
	2.1 Human Health Assessment	4
	2.2 Ecological Assessment	4
	2.3 Results of the 2011 Biennial Review	5
3.	Additional Information	8
4.	References	8

APPENDICES

Appendix A: Technical Memorandum: Report on the Pollutants' Database and Suitability

TABLES

Table 1. Metals Regulated in 40 CFR 503	. 2
Table 2. Pollutants Identified During the 2011 Biennial Review with Human Health	
Benchmarks	. 6
Table 3. List of Pollutants for which Human Health Benchmark Data Are Lacking	. 7

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1. Introduction

This document summarizes the U.S. Environmental Protection Agency's (EPA) activities related to the 2011 biennial review of pollutants pursuant to the Clean Water Act (CWA) Section 405(d)(2)(C). That section requires EPA to review existing sewage sludge regulations at least every two years to identify additional pollutants for possible regulation. The biennial review covered by this report summary, the 2011 Biennial Review, obtained sewage sludge-related literature through July 2011. Published data after this date will be considered in the 2013 Biennial Review. This document summarizes the analysis of that literature (a Technical Memorandum contractor summary is included in Attachment A). EPA often uses the term "biosolids" interchangeably with "sewage sludge," which is defined in the regulations and used in the statute.

1.1 History of the Standards for Use or Disposal of Sewage Sludge

In Section 405 of the CWA, Congress set forth a comprehensive program designed to reduce potential health and environmental risks associated with using or disposing of sewage sludge. Under Section 405(d), EPA establishes numeric limits and management practices that protect public health and the environment from the reasonably anticipated adverse effects of chemical and microbial pollutants in sewage sludge. Under Section 405(d), it is unlawful for any person to dispose of sewage sludge from a publicly owned treatment works (POTWs) or any other treatment works treating domestic sewage for any use for which regulations have been established pursuant to Section 405(d), except in accordance with those regulations.

On February 19, 1993, EPA identified several pollutants which, on the basis of available information on their toxicity, persistence, concentration, mobility, or potential for exposure, were present in sewage sludge in concentrations which may adversely affect public health or the environment. At that time, the Agency promulgated regulations, 40 CFR Part 503 *Standards for the Use or Disposal of Sewage Sludge*, specifying acceptable management practices, numeric standards for 11 metals (see **Table 1**), and operational standards for microbial organisms (58 *FR* 9248).

1

Metal	Land Application	Incineration ^b	Surface Disposal
Arsenic	✓	✓	✓
Beryllium		✓	
Cadmium	✓	✓	✓
Chromium III	√a	✓	✓
Copper	✓		
Lead	✓	\checkmark	~
Mercury	✓	\checkmark	
Molybdenum	√a		
Nickel	✓	\checkmark	~
Selenium	√a		
Zinc	\checkmark		

Table 1. Metals Regulated in 40 CFR 503

^a Minor amendments published in 1994 and 1995 improved clarity and responded to the results of judicial review resulting in changes in land application limits for chromium (deleted all limits; 60 FR 54764), molybdenum (deleted limits in Tables 2, 3, and 4 of Section 503.13; 59 FR 9095), and selenium (revising the selenium value for Table 3 of Section 503.13; 60 FR 54764)

^b Beryllium and Mercury emissions are regulated as limits to air emissions either by monitoring the exhaust air from the incinerator or the ambient air around the incinerator. In either case, the concentration in the air must meet the National Emission Standards for Hazardous Air Pollutants (NESHAPs, 40 CFR Part 61). Individual facility limits are based on unit performance calculations for Arsenic, Cadmium, Chromium, Lead, and Nickel. Operational standards include monitoring total hydrocarbons (THC) or carbon monoxide (CO) not to exceed 100 ppm by volume to represent all organic compounds in the exhaust gas that are covered by the Part 503 Rule. See Subpart E, Section 503.43 for other incineration requirements.

The 1993 rule established requirements for the final use or disposal of sewage sludge when it is: (1) applied to land as a fertilizer or soil amendment; (2) placed in a surface disposal site, including sewage sludge-only landfills; or (3) incinerated. These requirements apply to publicly and privately owned treatment works that generate or treat domestic sewage sludge and to anyone who manages sewage sludge. The rule also requires monitoring, record keeping, and reporting of specific information regarding sewage sludge management.

1.2 Biennial Reviews

Section 405(d)(2)(C) of the CWA requires the Agency to review from time to time, but not less often than every 2 years (i.e., biennial reviews), the regulations for the purpose of identifying additional toxic pollutants and promulgating regulations for such pollutants (the Agency uses the term pollutant as defined in the CWA). The purpose of reviewing information on pollutants, or potential pollutants, is to assess the availability and sufficiency of the data to conduct exposure and hazard assessments. Such exposure and hazard assessments, where sufficient data exist, allow the Agency to determine the potential for harm to public health or the environment

following use or disposal of sewage sludge. To inform the exposure and hazard assessments of pollutants in sewage sludge, EPA typically uses models that require the following data:

- Toxicity to human and ecological receptors (e.g., toxicity defined in terms of reference dose, reference concentrations, cancer slope factor, lethal dose, lethal concentration, or chronic endpoints related to fecundity).
- Concentration data in sewage sludge. Both the ability to detect a given pollutant in sewage sludge and the determination of the concentration at which that pollutant is present are highly dependent on the existence of analytical methods for that pollutant in the sewage sludge matrix.
- Fate and transport data for pollutants that may be present in sewage sludge. These data are necessary for assessing exposure. Examples of chemical and physical properties that are considered depending on the nature of a given pollutant in sewage sludge include:
 - Molecular weight
 - Solubility
 - Vapor pressure
 - Henry's law constant
 - Soil-water partitioning coefficient
 - Soil adsorption coefficient (Kd and Koc)
 - Degradation rates in various media
 - Log octanol-water partition coefficient (Log Kow)
 - Diffusivity in air and water
 - Bioavailability
 - Air-to-plant transfer factor
 - Root uptake factor for above ground vegetation
 - Root concentration factor
 - Bioconcentration factors for animal products (e.g., meat and milk)

The Agency evaluates the sufficiency of such data for pollutants having acceptable analytical methods, source concentration values, human health and ecological toxicity data, and data on environmental fate to support potential rulemaking under 40 CFR Part 503 (i.e., the development of numeric standards).

2. 2011 Biennial Review

EPA has conducted Biennial Reviews in 2003, 2005, 2007, and 2009. This document presents the 2011 Biennial Review. In conducting the 2011 Biennial Review, EPA collected and reviewed publicly available information on pollutants to evaluate potential harm to human health or the environment following use or disposal of sewage sludge.

2.1 Human Health Assessment

To determine if data are available to evaluate human health risks, EPA conducted a literature search for information published from August 2009 through July 2011. EPA searched databases and the published literature for information such as occurrence, fate and transport in the environment, and human health for pollutants in U.S. sewage sludge. Searches included the following: PubMed, TOXLINE, and the Environmental Sciences and Pollution Management Database. Search terms and key words are identified in Attachment A.

For biosolids human health risk evaluations, EPA previously relied solely on available Integrated Risk Information System (IRIS) and the Office of Pesticide Programs (OPP) established oral human health toxicity data for environmental contaminants and pesticides used by the Agency for risk assessment and risk management activities. Because human health benchmarks (HHBs) for pharmaceuticals, personal care products, and many other trace organic chemicals are seldom available through IRIS or OPP, EPA utilized data available from other sources to evaluate health risks of these contaminants.

The other sources (see Attachment A) may have included Superfund Health Risk Technical Support Center (STSC), Agency for Toxic Substances and Disease Registry (ATSDR), California Environmental Protection Agency (CalEPA), National Academy of Science, Australian Government, Health Canada, Food and Drug Administration (FDA), Joint FAO/WHO Expert Committee on Food Additives, and scientific journals for sources of human health toxicity information to evaluate health risks of these and other contaminants.

2.2 Ecological Assessment

EPA conducted a literature search for information published from August 2009 through July 2011 and searched databases and the published literature to capture available information necessary for ecological risk evaluations (e.g., occurrence, fate and transport in the environment,

4

and ecological effects) for pollutants in U.S. sewage sludge. The Agency used articles published in English in peer-reviewed journals, databases such as ECOTOX, Aquatic Sciences and Fisheries Abstracts, Biological Sciences Database, and the Environmental Sciences and Pollution Management Database, as well data for eco-toxicity benchmarks (e.g., the recent EPA Ecological Soil Screening Levels for certain metals for use in assessments under Superfund).

2.3 Results of the 2011 Biennial Review

The Agency's search of these databases and the open literature for articles published since the 2009 Biennial Review (EPA-822-R-10-002) identified information for 23 pollutants (listed in Tables 2 and 3) relevant to human health or ecological assessments. Some pollutants (e.g., triclosan and azithromycin) have been reported in previous biennial reviews. EPA may revisit previously evaluated pollutants, especially if literature searches of bibliographic databases reveal newer data. The Agency evaluated the availability and acceptability of data addressing toxicity to human and ecological receptors, pollutant concentrations in sewage sludge based on analytical methods, physical and chemical properties, and fate and transport in the environment in order to be able to conduct exposure and hazard assessments.

Two criteria were established for selecting a pollutant for an exposure and hazard evaluation if relevant exposure data were available: 1) the pollutant has human health or ecological toxicity values (e.g., studies that are adequate for evaluating hazards following acute or chronic exposure) and (2) the pollutant concentrations in U.S. sewage sludge are adequate (i.e., data are considered adequate when sufficient details are provided regarding sampling, handling, and analysis) based on suitable analytical methodology for detecting and quantifying pollutant concentrations (i.e., analytical methodology are acceptable when the processes and techniques have been independently replicated and / or validated, and written standard operating procedures exist).

The Agency divided the list of 23 identified pollutants into two major groups (i.e., those with human health benchmarks and those without human health benchmarks). **Table 2** lists the ten chemicals identified in the 2011 Biennial Review that have human health toxicity values (e.g., toxicity defined in terms of reference dose, reference concentrations, or cancer slope factor). EPA is in the process of evaluating five of the identified pollutants listed in Table 2 (i.e., carbamazepine, ciprofloxacin, diltiazem, gemfibrozil, and norfloxacin), as they have human

5

health toxicity values and have source concentration data from the Targeted National Sewage Sludge Survey (TNSSS; U.S. EPA, 2009). Critical information gaps exist for the remaining chemicals listed in Table 2 and include source concentration data, ecological effects endpoints, physical and chemical properties (e.g., diffusivity in water, aerobic biodegradation in soil and water, and anaerobic biodegradation in sediment), as well as bioconcentration and biotransfer factors.

Constituent Name	CASRN	Benchmark Source	Analyte Group
Bensulide	741-58-2	U.S. EPA RED, 2015a	Pesticide/herbicide
Bisphenol A	80-05-7	U.S. EPA IRIS, 2014	Plastics/resins
Carbamazepine ^a	298-46-4	Snyder et al., 2008	Antibiotics/disinfectants/ antimicrobials
Ciprofloxacin ^a	85721-33-1	Cunningham et al., 2009	Antibiotics/disinfectants/ antimicrobials
Diclofenac	15307-86-5	Bruce et al., 2010	Antibiotics/disinfectants/ antimicrobials
Dimethazone/Clomazone	81777-89-1	U.S. EPA TRED, 2015b	Pesticide/herbicide
Diltiazem ^a	42399-41-7	Schwab et al., 2005	Antibiotics/disinfectants/ antimicrobials
Fipronil	120068-37-3	U.S. EPA TRED, 2015c	Antibiotics/disinfectants/ antimicrobials
Gemfibrozil ^a	25812-30-0	Bruce et al., 2010	Antibiotics/disinfectants/ antimicrobials
Norfloxacin ^a	70458-96-7	Schwab et al., 2005	Antibiotics/disinfectants/ antimicrobials

Table 2. Pollutants Identified During the 2011 Biennial Review with Human Health Benchmarks

^a Currently undergoing assessment as part of the Targeted National Sewage Sludge Survey pollutants (USEPA, 2009). The other chemicals are missing critical pieces of data (e.g., source concentration data) and will be evaluated as data or tools become available.

Table 3 lists the 13 pollutants in sewage sludge for which the search did not identify definitive (i.e., draft values or benchmarks from dissertations for example are not considered definitive) human toxicity values from any of the sources that were searched. These pollutants were identified when the literature search, based on the keywords detailed in **Appendix A**, found other available data (i.e., data other than toxicity values, such as chemical properties). EPA also identified select microbial pollutants during its 2011 Biennial Review literature search. Although some data are available for select microbial pollutants (e.g., Enterococci and E. coli: U.S. EPA, 2012) the Agency lacks sufficient concentration data for most microbial pollutants, and current biosolids modelling and tools for risk assessment cannot be applied to microbial pollutants at this time. In addition, rather than conducting risk assessments for microbial agents, EPA relies on wastewater treatment technologies (i.e., 40CFR503, Subpart E) for reducing and controlling pathogens and indicator organisms in land applied biosolids.

Constituent Name	CASRN	Class	
Chemical Pollutants			
Azithromycin	83905-01-5	Antibiotics/disinfectants/ antimicrobials	
Caffeine	58-08-2	Pharmaceutical	
Carbamazepine-10,11-epoxide	298-46-4	Anticonvulsant/mood stabilizer	
Clindamycin	18323-44-9	Antibiotics/disinfectants/ antimicrobials	
Clotrimazole	23593-75-1	Antibiotics/disinfectants/ antimicrobials	
Dichlorocarbanilide	Not available	Antibiotics/disinfectants/ antimicrobials	
Estrone	53-16-7	Hormone	
Galaxolide (HHCB)	1222-05-5	Fragrance	
Nonylphenol ^a	84852-15-3	Surfactant	
Norfluoxetine	54910-89-3	Antibiotics/disinfectants/ antimicrobials	
Tenofovir	147127-20-6	Antibiotics/disinfectants/ antimicrobials	
Tonalide (AHTN)	21145-77-7	Fragrance	
Triclocarban	101-20-2	Antibiotics/disinfectants/ antimicrobials	

Table 3. List of Pollutants for which Human Health Benchmark Data Are Lacking

^a NOAEL and LOAEL data on systemic, reproductive, developmental, and maternal toxicity from repeated dose rat studies are available for nonylphenol; however, no RfD has yet been developed from these data. See <u>http://www.epa.gov/chemrtk/hpvis/</u><u>hazchar/Category_Alkylphenols_Sept2009.pdf</u>.

EPA is in the process of evaluating five of the pollutants listed in Table 3 (estrone, estriol, caffeine, azithromycin, and triclocarban), as they were also found in the TNSSS (U.S. EPA, 2009) and thus have source concentration data. However, the absence of human health toxicity values of high data quality for these five pollutants, as well as the other pollutants listed in Table 3, is still a major factor and limits EPA's ability to characterize risks to human health. EPA is investigating alternative methods or tools for filling the human health toxicity data gaps by developing provisional toxicity values, for example utilizing uncertainty factors or other reference values. We are also considering Quantitative Structural Activity Relationship (QSAR) approaches for estimating human health endpoints, which includes a basic assumption that similar molecules have similar activities. The QSAR approach could provide EPA with provisional toxicity values that are most similar to the RfDs used in EPA risk assessments. Available QSAR models are likely to require modification and/or extensive physical-chemical property data that may be lacking for information-poor chemical pollutants.

Critical information gaps for the remaining eight chemicals listed in Table 3 include additional needed information beyond human health toxicity values that include source concentration data, ecological effects endpoints, physical and chemical properties (e.g., diffusivity in water, aerobic

biodegradation in soil and water, and anaerobic biodegradation in sediment), or bioconcentration and biotransfer factors.

In summary, significant data gaps (environmental properties, human health and ecotoxicity values, and acceptable concentration data in sewage sludge) exist for the majority of the pollutants that limit the use of EPA's current biosolids modelling and tools for risk assessment at this time. Thus, EPA has not identified any additional toxic pollutants during its 2011 Biennial Review for potential regulation.

The Agency will continue to assess the availability of sufficient information for these and other pollutants identified during the biennial review activities pursuant to section 405(d)(2)(C) of the CWA. EPA is also in the process of evaluating Targeted National Sewage Sludge Survey chemicals (TNSSS) (U.S. EPA, 2009). As its first priority, EPA is in the process of evaluating ten (10) of 145 TNSSS pollutants) pollutants (i.e., barium, beryllium, manganese, molybdenum, silver, 4-chloroaniline, fluoranthene, pyrene, nitrate, and nitrite). Those assessments are underway at this time. EPA has begun on a longer term basis the evaluation of the balance of the 135 TNSSS pollutants.

EPA also investigating alternative methods or tools for filling human health toxicity data gaps, estimating missing source concentration data, and performing screening-level deterministic assessments to estimate human health and ecological hazards for biosolids land application scenarios. EPA will continue this work subject to availability of resources and overall program priorities. At this time EPA has not identified additional toxic pollutants for regulation under Section 405(d)(2)(C) of the CWA.

3. Additional Information

For more information about EPA's Biosolids Program, contact Rick Stevens in the Health and Ecological Criteria Division, 1200 Pennsylvania Avenue, N.W., Washington, DC 20460 (telephone: 202-566-1135 or e-mail: stevens.rick@epa.gov).

4. References

Bruce, GM, Pleus RC, Snyder, SA. 2010. Toxicological relevance of pharmaceuticals in drinking water. *Environ Sci & Technol* 44(14):5619–5626.

- Cunningham VL, Binks SP, Olson MJ. 2009. Human health risk assessment from the presence of human pharmaceuticals in the aquatic environment. *Regul Toxicol Pharmacol.* 53(1):39-45.
- Schwab BW, Hayes EP, Fiori JM, et al. 2005. Human pharmaceuticals in US surface waters: a human health risk assessment. *Regul Toxicol Pharmacol.* 42(3):296-312.
- Snyder SA. 2008. Occurrence, treatment, and toxicological relevance of EDCs and pharmaceuticals in water. *Ozone: Science and Engineering* 30:65–69.
- U.S. EPA. 2009. Targeted National Sewage Sludge Survey Sampling and Analysis Technical Report. Office of Water, Washington, DC. EPA-822-R-08-016. January 2009. http://water.epa.gov/scitech/wastetech/biosolids/upload/2009_01_15_biosolids_tnssstech.pdf
- U.S. EPA. 2012. 2012 Recreational Water Quality Criteria, Office of Water, Last updated on Tuesday, January 20, 2015 <u>http://water.epa.gov/scitech/swguidance/standards/criteria/health/recreation/index.cfm</u>
- U.S. EPA. 2014. Integrated Risk Information System (IRIS). Bisphenol A. (CASRN 80-05-7). Office of Pesticide Programs. Last updated on Friday, October 31, 2014. <u>http://www.epa.gov/iris/subst/0356.htm</u>
- U.S. EPA. 2015a. Office of Pesticide Programs: Bensulide. Reregistration Eligibility Decision (RED). Office of Pesticide Programs. Last updated on Wednesday, February 18, 2015. http://iaspub.epa.gov/apex/pesticides/f?p=CHEMICALSEARCH:3:0::NO:1,3,31,7,12,25: P3_XCHEMICAL_ID:1407
- U.S. EPA. 2015b. Office of Pesticide Programs: Dimethazone/Clomazone. Tolerance Reassessment Eligibility Decision (TRED). Office of Pesticide Programs. Updated on Thursday, February 19, 2015. <u>http://iaspub.epa.gov/apex/pesticides/f?p=CHEMICALSEARCH:3:0::NO:1,3,31,7,12,25:</u> <u>P3_XCHEMICAL_ID:1851</u>
- U.S. EPA. 2015c. Office of Pesticide Programs: Fipronil. Tolerance Reassessment Eligibility Decision (TRED). Last updated on Wednesday, February 18, 2015. <u>http://iaspub.epa.gov/apex/pesticides/f?p=CHEMICALSEARCH:3:0::NO:1,3,31,7,12,25:</u> <u>P3_XCHEMICAL_ID:2377</u>

Appendix A Technical Memorandum Report on Pollutants' Database and Suitability

A.1 Introduction

The following technical memorandum identifies chemical and microbial pollutants in U.S. sewage sludge between 2009 and 2011, and provides EPA with information on the suitability for modeling and potential rulemaking for these pollutants. Where the literature search did not result in potential human health benchmarks (HHBs), alternative approaches to estimating endpoints, where feasible, are identified.

A.2 Data Search

The search for new data utilized the strategy developed under previous biennial review efforts; results from bibliographic databases were limited to articles published in English in refereed journals. The bibliographic databases searched included PubMed, Science Citation Index Expanded, Toxline, Aquatic Sciences and Fisheries Abstracts, Biological Sciences Database, Environmental Sciences and Pollution Management; and Social Science journal website. Publications from August 2009 through July 2011 were sought. The data search included a combination of the following key words:

Topic/Keyword: sewage sludge, biosolids, treated sewage, sludge treatment, sewage treatment, land application, farm, agriculture, soil, occurrence, concentration, properties, fate, transport, health effects, ecological effects AND

Topic/Keyword: pollutants, toxicants, pharmaceuticals, antibiotics, steroids, hormones, pathogens, micro* (microbial, etc.), *Salmonella*

Using this search strategy, 208 articles were initially identified as potential sources of information on chemical and microbial pollutants in biosolids. Studies addressing previously modeled pollutants that appeared to provide new data on their behavior in the environment or toxicity were also included. Topics of excluded studies included toxicity through a medium other than biosolids (e.g., wastewater effluent); non-U.S. data; analytical methods; or an abstract was not available and the title alone did not give evidence for inclusion. A large number of international studies were identified that examined the occurrence and/or fate and transport of

pollutants from agriculturally applied biosolids in soils. However, these studies, with the exception of the Canadian studies, were excluded from consideration because treatment technologies and regulatory requirements are not necessarily representative of the U.S.

For this version of the biennial inventory update, EPA included Canadian studies for the first time, as there is a governmental research group (Agriculture and Agri-Food Canada) conducting numerous studies of interest on the fate and transport of pharmaceuticals and personal care products in agricultural soils¹); these studies were included due to expected similarities in Canadian and U.S. North American soil types. The literature review identified chemical-specific ecotoxicity data for only three pollutants (triclosan, triclocarban, and bisphenol A).

From the initial 208 articles, 64 were identified as providing relevant information on chemical and microbial pollutants in biosolids. The pollutants identified in these articles were divided into two major groups:

- Pollutants that have not previously been modeled but have human health benchmarks available, and
- Pollutants that have been identified in recent studies on biosolids and for which human health benchmarks were not identified.

A.3 Human Health Benchmark Data Source Hierarchy

For biosolids human health risk evaluations, EPA previously relied solely on EPA's Integrated Risk Information System (IRIS) and EPA's Office of Pesticide Programs (OPP) established oral human health toxicity data for environmental contaminants and pesticides used by the Agency for risk assessment and risk management activities. Because human health benchmarks (HHBs) for pharmaceuticals, personal care products, and many other trace organics are seldom available through IRIS or OPP, EPA utilized data available from other sources to evaluate health risks of these contaminants. We gave priority to those sources of information that provide HHBs based on similar methodologies as those used by IRIS, those sources that are more current, the basis for the values is transparent, and the data sources have undergone peer and public review. Tier 1 and 2 sources (EPA and non-EPA, respectively) are considered to represent the highest quality HHBs available, and these HHBs are frequently used to support EPA risk analyses. Tier 3 includes other high-quality sources that are either outdated (HEAST) or use different methodology or data

¹ Note that all articles in the Reference Section include abstracts when available.

than IRIS. For example, FDA and journal articles may use minimum therapeutic dose data instead of toxicity data. Some present benchmarks that are not RfDs but are analogous (e.g., acceptable daily intakes). And some do not provide an RfD or analogous benchmark, but do contain data from which an RfD could potentially be derived.

The three tiers and the data sources that fall into them are as follows:

Tier 1: Highest Quality EPA Sources

- 1. Integrated Risk Information System (IRIS) <u>http://www.epa.gov/iris</u>
- 2. OPP Reregistration Eligibility Decisions (REDs), Interim Reregistration Eligibility Decisions (IREDs), or Tolerance Reassessment Eligibility Decisions (TREDs) – <u>http://www.epa.gov/pesticides/reregistration/status.htm</u>
- 3. Superfund Health Risk Technical Support Center (STSC) Provisional Peer Reviewed Toxicity Values (PPRTVs) – <u>http://hhpprtv.ornl.gov/</u>

Tier 2: Highest Quality Non-EPA Sources

- 4. Agency for Toxic Substances and Disease Registry (ATSDR) Minimum Risk Levels (MRLs) <u>http://www.atsdr.cdc.gov/toxpro2.htm</u>
- 5. California Environmental Protection Agency (CalEPA) Reference Exposure Levels (RELs) and Cancer Potency Factors – <u>http://www.oehha.org/air/hot_spots/tsd052909.html</u> and <u>http://www.oehha.org/air/chronic_rels/index.html</u>
- 6. National Academy of Science (NAS) Tolerable Upper Intake Levels http://www.nal.usda.gov/fnic/DRI/DRI_Tables/UL_vitamins_elements.pdf
- 7. Joint FAO/WHO Expert Committee on Food Additives <u>http://apps.who.int/food-additives-contaminants-jecfa-database/search.aspx</u>

Tier 3: Other High Quality Sources

8. **Health Effects Assessment Summary Tables (HEAST)** – http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=2877

HEAST was developed by EPA's Office of Superfund Remediation and Technology Innovation to provide a comprehensive listing of provisional risk assessment information relative to oral and inhalation routes of exposure for chemicals for use at Superfund and RCRA sites. However, HEAST has not been updated since 1997 and may be considered outdated. It was used as a source for only the boron reference concentration (RfC). EPA is currently evaluating the risks of boron in biosolids as part of a risk assessment of 135 TNSSS pollutants. 9. U.S. Food and Drug Administration (FDA) Tolerances for Residues of New Animal Drugs in Food – see 21CFR556.

Note these are for animal tissue (e.g., beef, fish, and milk) and would not address hazards resulting from consumption of fruits, vegetables, water, or soil.

- 10. FDA Center For Veterinary Medicine http://www.fda.gov/AnimalVeterinary/default.htm
- 11. FDA Center for Drug Evaluation and Research <u>http://www.fda.gov/Drugs/default.htm</u>

The above three FDA sources use different methodology or data than the Tier 1 and 2 sources. EPA is in the process of developing benchmarks from these data for some pharmaceuticals.

- 12. **Scientific journals.** These may use different methodology or data than the Tier 1 and 2 sources.
- 13. European Union European Medicines Agency <u>http://www.emea.europa.eu/</u> This source also uses different methodology or data than the Tier 1 and 2 sources.

For chemicals that do not have human health toxicity value available, EPA will investigate filling the data gaps by using available estimation tools. One such tool involves developing provisional reference doses (RfDs) utilizing a methodology developed by Cunningham et al. (2009) and Schwab et al. (2005). The methodology is similar to that used to develop an RfD but instead of using a no observed adverse effect level (NOAEL), lowest observed adverse effect level (LOAEL), or benchmark dose as a point of departure (POD), an alternate or provisional POD (e.g., minimum or lowest daily therapeutic dose) is used. The provisional RfD process includes identifying a POD and converting it to a mg/kg-day basis, if necessary, and then applying appropriate uncertainty factors (UFs). A UF may range from 1 to 10 each, to account for uncertainty for extrapolation from low effect dose to no effect dose; uncertainty for extrapolation from shorter to longer duration of exposure; uncertainty for extrapolation between species; uncertainties related to intra individual variability; and data quality uncertainties (see Schwab et al., 2005 for recommendations on UF values). Identifying appropriate POD data would require additional review of the scientific literature; FDA online databases and other resources; manufacturers' pharmaceutical product information and safety data sheets; and/or standard drug information resources (e.g., Goodman & Gilman).

Another tool involves use of quantitative structural activity relationships (QSAR). An underlying premise to using QSAR is that similar molecules or chemical structures have similar activities (e.g., have similar toxicity end points and behavior in the environment). QSAR approaches to estimating human health endpoints could provide EPA with provisional RfDs that are most similar to the RfDs used in EPA risk assessments in a highly cost-effective manner. Available QSAR models are quite limited and likely to require modification and/or extensive physical-chemical property which may be lacking for information-poor chemical pollutants.

A.4 Identification of Additional Pollutants in U.S. Sewage Sludge

A.4.1 Pollutants with Human Health Benchmarks

Table A-1 lists the 9 chemicals with human health benchmarks that were identified from the sources discussed above and that fit the following criteria: (1) identified in the Targeted National Sewage Sludge Survey (TNSSS 2009), (2) not currently on EPA's list of potential candidates for addition to the Part 503 standards, and (3) not previously regulated or evaluated for sewage sludge. The chemicals are also identified by analyte groups defined by similarity in structure and typical uses, as appropriate. The following analyte groups are included in the TNSSS:

- Metals
- Polycyclic aromatic hydrocarbons (PAHs)
- Semivolatile organics and inorganic anions
- Polybrominated diphenyl ethers (PBDEs), including tetra, hexa, penta, and deca congeners
- Antibiotics and their degradation products, disinfectants, and other antimicrobials
- Steroids, hormones, and other drugs; and pesticides.

A.4.2 Pollutants Without Human Health Benchmarks

Table A-2 lists additional pollutants of concern (e.g., pharmaceuticals, hormones) in sewage

 sludge for which our search failed to identify human health benchmarks in EPA-approved

 sources.

Constituent Name	CASRN	Benchmark Source	Analyte Group
Bensulide	741-58-2	U.S. EPA RED, 2015a	Pesticide/herbicide
Bisphenol A	80-05-7	U.S. EPA IRIS, 2014	Plastics/resins
Carbamazepine ^a	298-46-4	Snyder et al., 2008	Antibiotics/disinfectants/ antimicrobials
Ciprofloxacin ^a	85721-33-1	Cunningham et al., 2009	Antibiotics/disinfectants/ antimicrobials
Diclofenac	15307-86-5	Bruce et al., 2010	Antibiotics/disinfectants/ antimicrobials
Diltiazem ^a	42399-41-7	Schwab et al., 2005	Antibiotics/disinfectants/ antimicrobials
Dimethazone/Clo mazone	81777-89-1	U.S. EPA TRED, 2015b	Pesticide/herbicide
Fipronil	120068-37-3	U.S. EPA TRED, 2015c	Antibiotics/disinfectants/ antimicrobials
Gemfibrozil ^a	25812-30-0	Bruce et al., 2010	Antibiotics/disinfectants/ antimicrobials
Norfloxacin ^a	70458-96-7	Schwab et al., 2005	Antibiotics/disinfectants/ antimicrobials

^a Currently undergoing assessment as part of the Targeted National Sewage Sludge Survey pollutants (USEPA, 2009). The other chemicals lack critical data (e.g., source concentration) and will be evaluated as data or tools become available.

Constituent Name	CASRN	Class	
Chemical Pollutants			
Azithromycin	83905-01-5	Antibiotics/disinfectants/ antimicrobials	
Caffeine	58-08-2	Pharmaceutical	
Carbamazepine-10,11-epoxide	298-46-4	Anticonvulsant/mood stabilizer	
Clindamycin	18323-44-9	Antibiotics/disinfectants/ antimicrobials	
Clotrimazole	23593-75-1	Antibiotics/disinfectants/ antimicrobials	
Dichlorocarbanilide	Not available	Antibiotics/disinfectants/ antimicrobials	
Estrone	53-16-7	Hormone	
Galaxolide (HHCB)	1222-05-5	Fragrance	
Nonylphenol ^a	84852-15-3	Surfactant	
Norfluoxetine	54910-89-3	Antibiotics/disinfectants/ antimicrobials	
Tenofovir	147127-20-6	Antibiotics/disinfectants/ antimicrobials	
Tonalide (AHTN)	21145-77-7	Fragrance	
Triclocarban	101-20-2	Antibiotics/disinfectants/ antimicrobials	

Table A-2. List of Pollutants for which Human Health Benchmarks Were Not Identified

(continues)

Constituent Name	CASRN	Class	
Microbial Pollutants ^b			
Adenovirus	Not applicable	Virus	
Bacteroides fragilis phage	Not applicable	Bacteriophage (viral indicator)	
Clostridia	Not applicable	Bacteria	
Clostridium perfringens	Not applicable	Bacteria	
Enterococci	Not applicable	Bacterial indicator	
Enterovirus	Not applicable	Virus	
Escherichia coli (E. coli)	Not applicable	Bacteria	
Giardia	Not applicable	Parasite	
HAV (Human Adenoviruses)	Not applicable	Virus	
Human polyomaviruses	Not applicable	Virus	
Listeria monocytogenes	Not applicable	Bacteria	
Listeria spp.	Not applicable	Bacteria	
Murine norovirus	Not applicable	Virus	
Salmonella senftenberg	Not applicable	Bacteria	
Somatic coliphage	Not applicable	Bacteriophage	

^a NOAEL and LOAEL data on systemic, reproductive, developmental, and maternal toxicity from repeated dose rat studies are available for nonylphenol; however, no RfD has yet been developed from these data. See http://www.epa.gov/chemrtk/hpvis/hazchar/Category_Alkylphenols_Sept2009.pdf.

^b Although data are available for some of the microbial pollutants listed (e.g., Enterococci and E. coli: U.S. EPA, 2012), EPA relies on treatment technologies, rather than risk assessment, for reducing and controlling pathogens and indicator organisms in land applied biosolids.

A.5 References

- Bruce GM, Pleus RC, Snyder SA. 2010. Toxicological relevance of pharmaceuticals in drinking water. *Environ Sci Technol.* 44(14):5619-26.
- Cunningham VL, Binks SP, Olson MJ. 2009. Human health risk assessment from the presence of human pharmaceuticals in the aquatic environment. *Regul Toxicol Pharmacol.* 53(1):39-45.
- Schwab BW, Hayes EP, Fiori JM, et al. 2005. Human pharmaceuticals in US surface waters: a human health risk assessment. *Regul Toxicol Pharmacol.* 42(3):296-312.
- Snyder SA. 2008. Occurrence, treatment, and toxicological relevance of EDCs and pharmaceuticals in water. *Ozone: Science and Engineering* 30:65-69.
- U.S. EPA (Environmental Protection Agency). 2009. Targeted National Sewage Sludge Survey Statistical Analysis Report. Office of Water, Washington, DC. EPA-822-R-08-018. Available online at http://water.epa.gov/scitech/wastetech/biosolids/tnsss-overview.cfm.
- U.S. EPA. 2012. 2012 Recreational Water Quality Criteria, Office of Water, Last updated on Tuesday, January 20, 2015 http://water.epa.gov/scitech/swguidance/standards/criteria/health/recreation/index.cfm
- U.S. EPA. 2014. Integrated Risk Information System (IRIS). Bisphenol A. (CASRN 80-05-7). Last updated on Friday, October 31, 2014. http://www.epa.gov/iris/subst/0356.htm

- U.S. EPA. 2015a. Office of Pesticide Programs: Bensulide. Reregistration Eligibility Decision (RED). Last updated on Wednesday, February 18, 2015. http://iaspub.epa.gov/apex/pesticides/f?p=CHEMICALSEARCH:3:0::NO:1,3,31,7,12,25: P3_XCHEMICAL_ID:1407
- U.S. EPA. 2015b. Office of Pesticide Programs: Dimethazone/Clomazone. Tolerance Reassessment Eligibility Decision (TRED). Office of Pesticide Programs. Updated on Thursday, February 19, 2015. <u>http://iaspub.epa.gov/apex/pesticides/f?p=CHEMICALSEARCH:3:0::NO:1,3,31,7,12,25:</u> <u>P3_XCHEMICAL_ID:1851</u>
- U.S. EPA. 2015c. Office of Pesticide Programs: Fipronil. Tolerance Reassessment Eligibility Decision (TRED). Last updated on Wednesday, February 18, 2015. http://iaspub.epa.gov/apex/pesticides/f?p=CHEMICALSEARCH:3:0::NO:1,3,31,7,12,25: P3_XCHEMICAL_ID:2377

Reference Abstracts

Mathney, J. M. (2011). "A critical review of the U.S. EPA's risk assessment for the land application of sewage sludge." <u>New Solut</u> **21**(1): 43-56.

Sewage sludge is a complex mixture of inorganic and organic materials and pathogens generated by the treatment of domestic sewage. Section 40 of the Code of Federal Regulations Part 503 regulates the land application of sewage sludge based on pathogen content and sets standards for nine inorganic chemicals. It is believed that the Part 503 standards are protective of human health and the environment and that sewage sludge applied to land poses little risk. A critical inspection of the pertinent literature, however, reveals that the standards were based on outdated methods, outdated data, inaccurate data, and flawed assumptions, leading to underestimation of risk. The standards are not sufficiently protective, and even if changes were made, sewage sludge is so complex that it is very unlikely it could be monitored to ensure the protection of human health and the environment. For these reasons, the practice of land application of sewage sludge must be discontinued.

Covaci, A., S. Harrad, et al. (2011). "Novel brominated flame retardants: a review of their analysis, environmental fate and behaviour." <u>Environ Int</u> **37**(2): 532-556.

This review summarizes current knowledge about production volumes, physico-chemical properties, analysis, environmental occurrence, fate and behaviour and human exposure to the "novel" brominated flame retardants (NBFRs). We define the term NBFRs as relating to BFRs which are new to the market or newly/recently observed in the environment. Restrictions and bans on the use of some polybrominated diphenyl ether (PBDE) formulations, in many jurisdictions, have created a market for the use of NBFRs. To date, most data on NBFRs have arisen as additional information generated by research designed principally to study more "traditional" BFRs, such as PBDEs. This has led to a wide variety of analytical approaches for sample extraction, extract purification and instrumental analysis of NBFRs. An overview of environmental occurrence in abiotic matrices, aquatic biota, terrestrial biota and birds is presented. Evidence concerning the metabolism and absorption of different NBFRs is reviewed. Human exposure to NBFRs via different exposure pathways is discussed, and research gaps related to analysis, environmental sources, fate, and behaviour and human exposure are identified.

Clarke, B. O. and S. R. Smith (2011). "Review of 'emerging' organic contaminants in biosolids and assessment of international research priorities for the agricultural use of biosolids." <u>Environ</u> Int **37**(1): 226-247.

A broad spectrum of organic chemicals is essential to modern society. Once discharged from industrial, domestic and urban sources into the urban wastewater collection system they may transfer to the residual solids during wastewater treatment and assessment of their significance and implications for beneficial recycling of the treated sewage sludge biosolids is required. Research on organic contaminants (OCs) in biosolids has been undertaken for over thirty years and the increasing body of evidence demonstrates that the majority of compounds studied do not place human health at risk when biosolids are recycled to farmland. However, there are 143,000 chemicals registered in the European Union for

industrial use and all could be potentially found in biosolids. Therefore, a literature review of 'emerging' OCs in biosolids has been conducted for a selection of chemicals of potential concern for land application based upon human toxicity, evidence of adverse effects on the environment and endocrine disruption. To identify monitoring and research priorities the selected chemicals were ranked using an assessment matrix approach. Compounds were evaluated based upon environmental persistence, human toxicity, evidence of bioaccumulation in humans and the environment, evidence of ecotoxicity and the number and quality of studies focused on the contaminant internationally. The identified chemicals of concern were ranked in decreasing order of priority: perfluorinated chemicals (PFOS, PFOA); polychlorinated alkanes (PCAs), polychlorinated naphthalenes (PCNs); organotins (OTs), polybrominated diphenyl ethers (PBDEs), triclosan (TCS), triclocarban (TCC); benzothiazoles; antibiotics and pharmaceuticals; synthetic musks; bisphenol A, quaternary ammonium compounds (QACs), steroids; phthalate acid esters (PAEs) and polydimethylsiloxanes (PDMSs). A number of issues were identified and recommendations for the prioritization of further research and monitoring of 'emerging' OCs for the agricultural use of biosolids are provided. In particular, a number of 'emerging' OCs (PFOS, PFOA and PCAs) were identified for priority attention that are environmentally persistent and potentially toxic with unique chemical properties, or are present in large concentrations in sludge, that make it theoretically possible for them to enter human and ecological foodchains from biosolids-amended soil.

Heidler, J. and R. U. Halden (2009). "Fate of organohalogens in US wastewater treatment plants and estimated chemical releases to soils nationwide from biosolids recycling." <u>J Environ Monit</u> **11**(12): 2207-2215.

This study examined the occurrence in wastewater of 11 aromatic biocides, pesticides and degradates, and their fate during passage through US treatment plants, as well as the chemical mass contained in sewage sludge (biosolids) destined for land application. Analyte concentrations in wastewater influent, effluent and sludge from 25 facilities in 18 US states were determined by liquid chromatography electrospray (tandem) mass spectrometry. Dichlorocarbanilide, fipronil, triclocarban, and triclosan were found consistently in all sample types. Dichlorophene, hexachlorophene, and tetrachlorocarbanilide were detected infrequently only, and concentrations of the phenyl urea pesticides diflubenzuron, hexaflumuron, and linuron were below the limit of detection in all matrixes. Median concentrations (+/-95% confidence interval) of quantifiable compounds in influent ranged from 4.2 +/- 0.8 microg L(-1) for triclocarban to 0.03 +/- 0.01 microg L(-1) for fipronil. Median concentrations in effluent were highest for triclocarban and triclosan (0.23 ± 0.08) and 0.07 + 0.04 microg L(-1), respectively). Median aqueous-phase removal efficiencies (+/-95% CI) of activated sludge treatment plants decreased in the order of: triclosan (96 +/-2% > triclocarban (87 +/- 7%) > dichlorocarbanilide (55 +/- 20%) > fipronil (18 +/- 22%). Median concentrations of organohalogens were typically higher in anaerobically than in aerobically digested sludges, and peaked at 27 600 +/- 9600 and 15 800 +/- 8200 microg kg(-1) for triclocarban and triclosan, respectively. Mass balances obtained for three primary pesticides in six activated sludge treatment plants employing anaerobic digestion suggested a decreasing overall persistence from fipronil (97 + 70%) to triclocarban (87 + 29%) to triclosan (28 +/- 30%). Nationwide release of the investigated organohalogens to agricultural land via municipal sludge recycling and into surface waters is estimated to total 258 000 +/-

110 00 kg year(-1) (mean +/- 95% confidence interval), with most of this mass derived from antimicrobial consumer products of daily use. This study addresses some of the data gaps identified by the National Research Council in its 2002 study on standards and practices of biosolids application on land.

Lindstrom, A. B., M. J. Strynar, et al. (2011). "Application of WWTP Biosolids and Resulting Perfluorinated Compound Contamination of Surface and Well Water in Decatur, Alabama, USA." <u>Environ Sci Technol</u> **2011 Apr 22. [Epub ahead of print]**.

Perfluorinated chemicals (PFCs) such as perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) have been produced and used in a wide range of industrial and consumer products for many decades. Their resistance to degradation has led to their widespread distribution in the environment, but little is known about how humans become exposed. Recent studies have demonstrated that the application of PFC contaminated biosolids can have important effects on local environments, ultimately leading to demonstrable human exposures. This manuscript describes a situation in Decatur, Alabama where PFC contaminated biosolids from a local municipal wastewater treatment facility that had received waste from local fluorochemical facilities were used as a soil amendment in local agricultural fields for as many as twelve years. Ten target PFCs were measured in surface and groundwater samples. Results show that surface and well water in the vicinity of these fields had elevated PFC concentrations, with 22% of the samples exceeding the U.S. Environmental Protection Agency's Provisional Health Advisory level for PFOA in drinking water of 400 ng/L. Water/soil concentration ratios as high as 0.34 for perfluorohexanoic acid, 0.17 for perfluoroheptanoic acid, and 0.04 for PFOA verify decreasing mobility from soils with increasing chain length while indicating that relatively high transport from soils to surface and well water is possible.

Washington, J. W., H. Yoo, et al. (2010). "Concentrations, distribution, and persistence of perfluoroalkylates in sludge-applied soils near Decatur, Alabama, USA." <u>Environ Sci Technol</u> **44**(22): 8390-8396.

Sludges generated at a wastewater treatment plant (WWTP) in Decatur, Alabama have been applied to agricultural fields for more than a decade. Waste-stream sources to this WWTP during this period included industries that work with fluorotelomer compounds, and sludges from this facility have been found to be elevated in perfluoroalkylates (PFAs). With this knowledge, the U.S. Environmental Protection Agency collected soil samples from sludgeapplied fields as well as nearby "background" fields for PFA analysis. Samples from the sludge-applied fields had PFAs at much higher concentrations than in the background fields; generally the highest concentrations were perfluorodecanoic acid ($\leq 990 \text{ ng/g}$), perfluorododecanoic acid (</= 530 ng/g), perfluorooctanoic acid (</= 320 ng/g), and perfluorooctane sulfonate (</= 410 ng/g). Contrasts in PFA concentration between surface and deeper soil samples tended to be more pronounced in long-chain congeners than shorter chains, perhaps reflecting relatively lower environmental mobilities for longer chains. Several PFAs were correlated with secondary fluorotelomer alcohols (sec-FTOHs) suggesting that PFAs are being formed by degradation of sec-FTOHs. Calculated PFA disappearance half-lives for C6 through C11 alkylates ranged from about 1 to 3 years and increase with increasing chain-length, again perhaps reflecting lower mobility of the longerchained compounds.

Agyin-Birikorang, S., M. Miller, et al. (2010). "Retention-release characteristics of triclocarban and triclosan in biosolids, soils, and biosolids-amended soils." <u>Environ Toxicol Chem</u> **29**(9): 1925-1933.

Transport models that incorporate retention/release characteristics of organic compounds in soils and sediments typically assume that organic-carbon normalized partition coefficients (K(OC)) apply to all solid matrices and that the partitioning process is completely reversible. Partition coefficients (K(d)) (from which the K(OC) was calculated), and retention/release characteristics of triclocarban (TCC) and triclosan (TCS) in biosolids, soils, and biosolidsamended soils were determined. Four soils of different physicochemical properties amended with biosolids at 10 g/kg, together with unamended soils, and several biosolids were separately spiked with either [(14)C]TCC or [(14)C]TCS for the various determinations. The hysteresis coefficient values of the two compounds were consistently <1 in all three solid matrices, suggesting strong hysteresis. Multiple desorption steps (24 h each) over several days revealed incomplete desorption of the two compounds from all three solid matrices. The K(d) values determined in biosolids (log K(d) 3.34 ± 0.13 for TCC and 3.76 ± 0.39 for TCS) were greater than those determined in soils (log K(d) 1.71 ± 0.09 for TCC and 2.25 +/- 0.26 for TCS) and biosolids-amended soils (log K(d)1.90 +/- 0.16 for TCC and 2.31 +/-0.19 for TCS), however, the K(OC) values of all three solid matrices were similar (log K(OC) of 3.82 +/- 0.16 for TCC and 4.26 +/- 0.31 for TCS). Thus, it was concluded that a single or a narrow range of K(OC) values for TCC and TCS may be appropriate to describe retention of the compounds in soils and sediments. However, models that assume complete reversibility of the retention/release processes of the compounds in soils and sediments may not adequately describe the retention/release characteristics of the compounds in soils and sediments, especially when the chemicals are biosolids borne.

Al-Rajab, A. J., L. Sabourin, et al. (2010). "Fate of the antiretroviral drug tenofovir in agricultural soil." <u>Sci Total Environ</u> **408**(22): 5559-5564. [Canadian Article]

Tenofovir (9-(R)-(2-phosphonylmethoxypropyl)-adenine) is an antiretroviral drug widely used for the treatment of human immunodeficiency virus (HIV-1) and Hepatitis B virus (HBV) infections. Tenofovir is extensively and rapidly excreted unchanged in the urine. In the expectation that tenofovir could potentially reach agricultural lands through the application of municipal biosolids or wastewater, and in the absence of any environmental fate data, we evaluated its persistence in selected agricultural soils. Less than 10% of [adenine-8-(14)C]-tenofovir added to soils varying widely in texture (sand, loam, clay loam) was mineralized in a 2-month incubation under laboratory conditions. Tenofovir was less readily extractable from clay soils than from a loam or a sandy loam soil. Radioactive residues of tenofovir were removed from the soil extractable fraction with DT(50)s ranging from 24+/-2 to 67+22days (first order kinetic model) or 44+9 to 127+55days (zero order model). No extractable transformation products were detectable by HPLC. Tenofovir mineralization in the loam soil increased with temperature (range 4 degrees C to 30 degrees C), and did not occur in autoclaved soil, suggesting a microbial basis. Mineralization rates increased with soil moisture content, ranging from air-dried to saturated. In summary, tenofovir was relatively persistent in soils, there were no extractable transformation products detected, and the response of [adenine-8-(14)C]-tenofovir mineralization to soil temperature and heat sterilization indicated that the molecule was biodegraded by aerobic microorganisms. Sorption isotherms with dewatered biosolids suggested that tenofovir

residues could potentially partition into the particulate fraction during sewage treatment.

Wong, K., B. M. Onan, et al. (2010). "Quantification of enteric viruses, pathogen indicators, and Salmonella bacteria in class B anaerobically digested biosolids by culture and molecular methods." <u>Appl Environ Microbiol</u> **76**(19): 6441-6448.

The most common class B biosolids in the United States are generated by mesophilic anaerobic digestion (MAD), and MAD biosolids have been used for land application. However, the pathogen levels in MAD biosolids are still unclear, especially with respect to enteric viruses. In this study, we determined the occurrence and the quantitative levels of enteric viruses and indicators in 12 MAD biosolid samples and of Salmonella enterica in 6 MAD biosolid samples. Three dewatered biosolid samples were also included in this study for purposes of comparison. Human adenoviruses (HAdV) had the highest gene levels and were detected more frequently than other enteric viruses. The gene levels of noroviruses (NV) reported were comparable to those of enteroviruses (EV) and human polyomaviruses (HPyV). The occurrence percentages of HAdV, HAdV species F, EV, NV GI, NV GII, and HPyV in MAD samples were 83, 83, 42, 50, 75, and 58%, respectively. No hepatitis A virus was detected. Infectious HAdV was detected more frequently than infectious EV, and all infectious HAdV were detected when samples were propagated in A549 cells. Based on most-probable-number (MPN) analysis, A549 cells were more susceptible to biosolidassociated viruses than BGM cells. All indicator levels in MAD biosolids were approximately 10(4) MPN or PFU per gram (dry), and the dewatered biosolids had significantly higher indicator levels than the MAD biosolids. Only two MAD samples tested positive for Salmonella enterica, where the concentration was below 1.0 MPN/4 g. This study provides a broad comparison of the prevalence of different enteric viruses in MAD biosolids and reports the first detection of noroviruses in class B biosolids. The observed high quantitative and infectivity levels of adenoviruses in MAD biosolids indicate that adenovirus is a good indicator for the evaluation of sludge treatment efficiency.

Wei, J., Y. Jin, et al. (2010). "Survival of murine norovirus and hepatitis A virus in different types of manure and biosolids." <u>Foodborne Pathog Dis</u> **7**(8): 901-906.

Noroviruses and hepatitis A virus (HAV) are common causes of foodborne disease. They are usually shed in feces and have been found in sewage water, biosolids, and animal manures. With the wide application of manure and biosolids on agricultural lands, there is an increasing interest in investigating virus survival in manure and biosolids. In this study, Murine norovirus-1 (MNV) and HAV were inoculated into different types of animal manure and three types of differently treated biosolids at 20 degrees C and 4 degrees C for up to 60 days. Both HAV and MNV viral genomes degraded immediately in high pH biosolids type 2 and 3 at time zero. For other types of manure and biosolids, HAV RNA was significantly reduced in biosolids type 1 and in liquid dairy manure (DM) after 60 days stored at 20 degrees C, but was stable in all types of manure and biosolids type 1 at 4 degrees C. MNV RNA was unstable in pelletized poultry litter and biosolids type 1 at 20 degrees C, and less stable in liquid DM at both temperatures. For MNV infectivity, there was no significant difference among pelletized poultry litter, alum-treated poultry litter, raw poultry litter, and swine manure at either 20 degrees C or 4 degrees C after 60 days of storage. However, HAV stored in swine manure and raw poultry litter had significantly higher infectivity levels than HAV stored in alum-treated poultry litter at both 20 degrees C and 4 degrees C. Overall, both viruses were inactivated rapidly in alkaline pH biosolids and unstable in liquid DM, but alum added in poultry litter had different effects on the two viruses: alum inactivated some HAV at both temperatures but had no effect on MNV.

Antonious, G. F. (2010). "Mobility and half-life of bensulide in agricultural soil." <u>J Environ Sci</u><u>Health B</u> **45**(1): 1-10.

Environmentally and economically viable agriculture requires the use of cultivation practices that maximize agrochemical efficacy while minimizing their off-site movement. Bensulide [O, O-diisopropyl S-2-phenylsulfonylaminoethyl phosphorodithioate] is one of the few herbicides from the organophosphate group used for control of weeds that threaten numerous crops. A field study was conducted on a silty-loam soil of 10% slope at Kentucky State University Research Farm to monitor off-site movement and persistence of bensulide in soil. Eighteen plots of 22 x 3.7 m each were separated using metal borders and the soil in six plots was mixed with sewage sludge and yard waste compost (SS-YW) at 15 t acre(-1) on dry weight basis, six plots were mixed with sewage sludge (SS) at 15 t acre(-1), and six unamended plots (NM) were used for comparison purposes. Plots were planted with summer squash, Cucurbita pepo as the test plant. The objectives of this investigation were to: 1) determine the dissipation and half-life (T(1/2)) of bensulide in soil under three management practices; 2) monitor the concentration of bensulide residues in runoff and infiltration water following natural rainfall; and 3) determine the effect of soil amendments on the transport of NO(3), NH(4), and P into surface and subsurface water. Half-life (T(1/2)) values of bensulide in soil were 44.3, 37.6, and 27.1 d in SS-YW, SS, and NM treatments, respectively. Addition of SS-YW and SS to native soil increased water infiltration, lowering runoff water volume and bensulide residues in runoff following natural rainfall events.

Snyder, E. H., G. A. O'Connor, et al. (2010). "Fate of 14C-triclocarban in biosolids-amended soils." <u>Sci Total Environ</u> **408**(13): 2726-2732.

Triclocarban (TCC) is an antibacterial compound commonly detected in biosolids at partsper-million concentrations. Approximately half of the biosolids produced in the United States are land-applied, resulting in a systematic release of TCC into the soil environment. The extent of biosolids-borne TCC environmental transport and potential human/ecological exposures will be greatly affected by its bioavailability and the rate of degradation in amended soils. To investigate these factors, radiolabeled TCC ((14)C-TCC) was incorporated into anaerobically digested biosolids, amended to two soils, and incubated under aerobic conditions. The evolution of (14)CO2 (biodegradation) and changes in chemical extractability (bioavailability) was measured over time. Water extractable TCC over the study period was low and significantly decreased over the first 3 weeks of the study (from 14% to 4% in a fine sand soil and from 3 to <1% in a silty clay loam soil). Mineralization (i.e. ultimate degradation), as measured by evolution of (14)CO(2), was <4% over 7.5 months. Methanol extracts of the amended soils were analyzed by radiolabel thin-layer chromatography (RAD-TLC), but no intermediate degradation products were detected. Approximately 20% and 50% of the radioactivity in the amended fine sand and silty clay loam soils, respectively, was converted to bound residue as measured by solids combustion. These results indicate that biosolids-borne TCC becomes less bioavailable over time and biodegrades at a very slow rate.

Wu, C., A. L. Spongberg, et al. (2010). "Dissipation and leaching potential of selected pharmaceutically active compounds in soils amended with biosolids." <u>Arch Environ Contam</u> <u>Toxicol</u> **59**(3): 343-351.

Biosolids land application is an important pathway introducing pharmaceuticals into the environment. In this work, laboratory column and dissipation experiments were performed using soils of varying properties in order to study the fate and transport of pharmaceutical residues introduced by the land application of biosolids. For experimentation, five pharmaceutical compounds (carbamazepine, diphenhydramine, fluoxetine, diltiazem, and clindamycin) and two metabolites (carbamazepine-10,11-epoxide and norfluoxetine) commonly found in biosolids were selected. Leaching experiments indicate that the selected pharmaceuticals have low mobility in tested soils. However, small portions of the applied pharmaceuticals were recovered in the leachates, likely attributed to sorption to dissolved organic matter. Dissipation experiments show that carbamazepine, diphenhydramine, and fluoxetine were persistent in soils, whereas the dissipation of diltiazem and clindamycin was affected by redox conditions and soil properties.

Antonious, G. F. (2011). "Mitigation of dimethazone residues in soil and runoff water from agricultural field." J Environ Sci Health B **46**(1): 76-83.

Dimethazone, also known as clomazone [2-[(2-chlorophenyl) methyl]- 4,4-dimethyl-3isoxaolidinone] is a pre-emergent nonionic herbicide commonly used in agriculture. A field study was conducted on a silty-loam soil of 10 % slope to monitor off-site movement and persistence of dimethazone in soil under three management practices. Eighteen plots of 22 x 3.7 m each were separated using stainless steel metal borders and the soil in six plots was mixed with municipal sewage sludge (MSS) and yard waste (YW) compost (MSS+YW) at 15 t acre(1) on dry weight basis, six plots were mixed with MSS at 15 t acre(1), and six unamended plots (NM) were used for comparison purposes. The objectives of this investigation were to: (i) monitor the dissipation and half-life (T/) of dimethazone in soil under three management practices; (ii) determine the concentration of dimethazone residues in runoff and infiltration water following natural rainfall events; and (iii) assess the impact of soil amendments on the transport of NO, NH, and P into surface and subsurface water. Gas chromatography/mass spectrometry (GC/MS) analyses of soil extracts indicated the presence of ion fragments at m/z 125 and 204 that can be used for identification of dimethazone residues. Initial deposits of dimethazone varied from 1.3 mug g(1) dry native soil to 3.2 and 11.8 mug g(1) dry soil in MSS and MSS+YW amended soil, respectively. Decline of dimethazone residues in the top 15 cm native soil and soil incorporated with amendments revealed half-life (T/) values of 18.8, 25.1, and 43.0 days in MSS+YW, MSS, and NM treatments, respectively. Addition of MSS+YW mix and MSS alone to native soil increased water infiltration, lowering surface runoff water volume and dimethazone residues in runoff following natural rainfall events.

Al-Rajab, A. J., L. Sabourin, et al. (2010). "The non-steroidal anti-inflammatory drug diclofenac is readily biodegradable in agricultural soils." <u>Sci Total Environ</u> **409**(1): 78-82. [Canadian Article]

Diclofenac, 2-[2-[(2,6-dichlorophenyl)amino]phenyl]acetic acid, is an important nonsteroidal anti-inflammatory drug widely used for human and animals to reduce inflammation and pain. Diclofenac could potentially reach agricultural lands through the application of municipal biosolids or wastewater, and in the absence of any environmental fate data, we evaluated its persistence in agricultural soils incubated in the laboratory. (14)C-Diclofenac was rapidly mineralized without a lag when added to soils varying widely in texture (sandy loam, loam, clay loam). Over a range of temperature and moisture conditions extractable (14)C-diclofenac residues decreased with half-lives <5days. No extractable transformation products were detectable by HPLC. Diclofenac mineralization in the loam soil was abolished by heat sterilization. Addition of biosolids to sterile or non-sterile soil did not accelerate the dissipation of diclofenac. These findings indicate that diclofenac is readily biodegradable in agricultural soils.

Andrade, N. A., L. L. McConnell, et al. (2010). "Persistence of polybrominated diphenyl ethers in agricultural soils after biosolids applications." <u>J Agric Food Chem</u> **58**(5): 3077-3084.

This study examines polybrominated diphenyl ethers (PBDE) levels, trends in biosolids from a wastewater treatment plant, and evaluates potential factors governing PBDE concentrations and the fate in agricultural soils fertilized by biosolids. The mean concentration of the most abundant PBDE congeners in biosolids (summation operatorBDE-47, BDE-99, and BDE-209) generated by one wastewater treatment plant was 1250 +/- 134 microg/kg d.w. with no significant change in concentration over 32 months (n = 15). In surface soil samples from the Mid-Atlantic region, average PBDE concentrations in soil from fields receiving no biosolids (5.01 +/- 3.01 microg/kg d.w.) were 3 times lower than fields receiving one application (15.2 +/- 10.2 microg/kg d.w.) and 10 times lower than fields that had received multiple applications (53.0 +/- 41.7 microg/kg d.w.). The cumulative biosolids application rate and soil organic carbon were correlated with concentrations and persistence of PBDEs in soil. A model to predict PBDE concentrations in soil after single or multiple biosolids applications provides estimates which fall within a factor of 2 of observed values.

Ismail, Z. Z., U. Tezel, et al. (2010). "Sorption of quaternary ammonium compounds to municipal sludge." <u>Water Res</u> **44**(7): 2303-2313.

The sorptive behavior of four quaternary ammonium compounds (QACs) - hexadecyl trimethyl ammonium chloride (C(16)TMA), dodecyl trimethyl ammonium chloride (C(12)TMA), hexadecyl benzyl dimethyl ammonium chloride (C(16)BDMA), and dodecyl benzyl dimethyl ammonium chloride (C(12)BDMA) - to municipal primary, waste activated, mesophilic digested, and thermophilic digested sludges was assessed at 22 degrees C. Batch adsorption of all four separately tested QACs to primary sludge reached equilibrium within 4h. At a nominal, initial QAC concentration of 300mg/L and a sludge volatile solids concentration of 1g/L, the extent of adsorption was 13, 88, 67, and 89% for the C(12)TMA, C(16)TMA, C(12)BDMA, and C(16)BDMA, respectively, and correlated positively to the QAC hydrophobicity and negatively to their critical micelle concentration. Equilibrium partitioning data were described by the Freundlich isotherm model. The adsorption capacity of the four sludges was very similar. In binary QAC mixtures, QACs with relatively high adsorption affinity and at relatively high aqueous concentrations decreased the adsorption of QACs with a low adsorption affinity. At pH 7, about 40% of the sludge-C(12)TMA desorbed, whereas less than 5% of the sludge-C(16)BDMA desorbed in 10 days. The effect of pH was negligible on the desorption extent of C(12)TMA at a pH range 4-10 over 10 days, whereas increasing the solution pH to 10 resulted in more than 50% desorption of C(16)BDMA. Given the fact that approximately 50% of the municipal biosolids are landapplied in the US, the data of this study would help in the assessment of the fate of QACs and their potential effect on human and environmental health.

Lozano, N., C. P. Rice, et al. (2010). "Fate of triclosan in agricultural soils after biosolid applications." <u>Chemosphere</u> **78**(6): 760-766.

Triclosan (5-chloro-2-[2,4-dichloro-phenoxy]-phenol (TCS) is an antimicrobial compound that is added to a wide variety of household and personal care products. The consumer use of these products releases TCS into urban wastewater and this compound ends up in the environment when agricultural land is fertilized with wastewater biosolids. This study examines the occurrence of TCS in biosolids and its fate in biosolid-treated soils. TCS levels in biosolids generated from one repeatedly-sampled wastewater treatment plant averaged 15.6 + or - 0.6 mg kg(-1) dry wt. (mean + or - standard error) with a slight increase from 2005 to 2007. Surface soil samples were collected from several farms in northern Virginia, US that had received no biosolids, one biosolid application or multiple biosolid applications since 1992. Farm soils that received one application presented TCS concentrations between 4.1 and 4.5 ng g(-1) dry wt. when time since application was over 16 months and between 23.6 and 66.6 ng g(-1) dry wt. for farms where sampling time after application was less than a year. Our results suggest that TCS content of biosolids are rapidly dissipated (estimated half-life of 107.4 d) when applied to agricultural fields. Statistical differences were found (p<0.05) for residual build-up of TCS between multiple-application farms (at least 480 d after application) and controls suggesting that there was a slight build-up of TCS, although the concentrations for these farms were low (<10 ng g(-1) dry wt.).

Sabourin, L., A. Beck, et al. (2009). "Runoff of pharmaceuticals and personal care products following application of dewatered municipal biosolids to an agricultural field." <u>Sci Total</u> <u>Environ</u> **407**(16): 4596-4604. [Canadian Article]

Municipal biosolids are a useful source of nutrients for crop production, and commonly used in agriculture. In this field study, we applied dewatered municipal biosolids at a commercial rate using broadcast application followed by incorporation. Precipitation was simulated at 1, 3, 7, 21 and 34 days following the application on 2 m(2) microplots to evaluate surface runoff of various pharmaceuticals and personal care products (PPCPs), namely atenolol, carbamazepine, cotinine, caffeine, gemfibrozil, naproxen, ibuprofen, acetaminophen, sulfamethoxazole, triclosan and triclocarban. There was little temporal coherence in the detection of PPCPs in runoff, various compounds being detected maximally on days 1, 3, 7 or 36. Maximum concentrations in runoff ranged from below detection limit (gemfibrozil) to 109.7 ng L(-1) (triclosan). Expressing the total mass exported as a percentage of that applied, some analytes revealed little transport potential (<1% exported; triclocarban, triclosan, sulfamethoxazole, ibuprofen, naproxen and gemfibrozil) whereas others were readily exported (>1% exported; acetaminophen, carbamazepine, caffeine, cotinine, atenolol). Those compounds with little transport potential had log K(ow) values of 3.18 or greater, whereas those that were readily mobilized had K(ow) values of 2.45 or less. Maximal concentrations of all analytes were below toxic concentrations using a variety of endpoints available in the literature. In summary, this study has quantified the transport potential in surface runoff of PPCPs from land receiving biosolids, identified that log K(ow) may be a determinant of runoff transport potential of these analytes, and found maximal concentrations of all chemicals tested to be below toxic concentrations using a variety of endpoints.

Wu, C., A. L. Spongberg, et al. (2009). "Adsorption and degradation of triclosan and triclocarban in soils and biosolids-amended soils." J Agric Food Chem **57**(11): 4900-4905.

Triclosan and triclocarban are antibacterial agents that are widely used in numerous personal care products. Limited information is available on their environmental behavior in soils and soils land applied with wastewaters and biosolids. In this study, laboratory experiments were performed to investigate their adsorption and degradation in soils. Both antibacterial agents adsorbed strongly to the sandy loam and silty clay soils with and without addition of biosolids, with distribution coefficients (K(d)) ranging from 178 to 264 L kg(-1) for triclosan and from 763 to 1187 L kg(-1) for triclocarban. Sorption of triclosan decreased with increase in soil pH from 4 to 8, whereas triclocarban sorption showed no effect within the tested pH range. Competitive sorption was observed when triclosan and triclocarban coexisted, but the cosolute effect was concentration dependent. Biosolids amendment increased the sorption of triclosan and triclocarban, likely due to the addition of soil organic matter, but displayed no significant effect on degradation.

Barron, L., J. Havel, et al. (2009). "Predicting sorption of pharmaceuticals and personal care products onto soil and digested sludge using artificial neural networks." <u>Analyst</u> **134**(4): 663-670.

A comprehensive analytical investigation of the sorption behaviour of a large selection of over-the-counter, prescribed pharmaceuticals and illicit drugs to agricultural soils and freezedried digested sludges is presented. Batch sorption experiments were carried out to identify which compounds could potentially concentrate in soils as a result of biosolid enrichment. Analysis of aqueous samples was carried out directly using liquid chromatography-tandem mass spectrometry (LC-MS/MS). For solids analysis, combined pressurised liquid extraction and solid phase extraction methods were used prior to LC-MS/MS. Solid-water distribution coefficients (K(d)) were calculated based on slopes of sorption isotherms over a defined concentration range. Molecular descriptors such as log P, pK(a), molar refractivity, aromatic ratio, hydrophilic factor and topological surface area were collected for all solutes and, along with generated K(d) data, were incorporated as a training set within a developed artificial neural network to predict K(d) for all solutes within both sample types. Therefore, this work represents a novel approach using combined and cross-validated analytical and computational techniques to confidently study sorption modes within the environment. The logarithm plots of predicted versus experimentally determined K(d) are presented which showed excellent correlation (R(2) > 0.88), highlighting that artificial neural networks could be used as a predictive tool for this application. To evaluate the developed model, it was used to predict K(d) for meclofenamic acid, mefenamic acid, ibuprofen and furosemide and subsequently compared to experimentally determined values in soil. Ratios of experimental/predicted K(d) values were found to be 1.00, 1.00, 1.75 and 1.65, respectively.

Brown, S., D. Devin-Clarke, et al. (2009). "Fate of 4-nonylphenol in a biosolids amended soil." <u>Chemosphere</u> **75**(4): 549-554.

The fate of the endocrine disrupting compound 4-nonylphenol (NP) in an agricultural soil amended with biosolids was assessed in a greenhouse study. A biosolids with a total NP concentration of 900 mg kg(-1) was incorporated into the 4 cm surface layer of soil columns at an agronomic rate equivalent to 1.7 kg m(2). Half of the columns were planted with

Triticum aestivum L., red hardy winter wheat seeds, whereas the remaining columns were unplanted to evaluate the influence of plant growth on the fate of NP. The degradation of total NP and eight NP isomers was monitored over 45 d. The half-life of NP in this soil system ranged from 16 to 23 d depending on treatment. After 45 d from the start of the trail, 15% of the initial biosolids-NP remained in the planted columns, whereas approximately 30% remained in the unplanted columns, indicating enhanced degradation in the presence of plants. The eight NP isomers exhibited different degradation rates, but minimal amounts of all isomers persisted after 45 d. Movement of NP below the zone of incorporation was slight (<2% of total NP present at any sampling interval) and no NP was detected in column leachates or in wheat leaves.

Stietiya, M. H. and J. J. Wang (2011). "Effect of organic matter oxidation on the fractionation of copper, zinc, lead, and arsenic in sewage sludge and amended soils." <u>J Environ Qual</u> **40**(4): 1162-1171.

Long-term land application of sewage sludge (SS) has caused concern over the potential release of trace metals into the environment following the degradation of organic matter (OM). This study was performed to assess the impact of OM degradation on the relative distribution of Cu, Zn, Pb, and As in SS and SS-amended soils. Three SSs of different ages and two soils treated with SS were subjected to incubation and direct chemical oxidation using diluted HO, followed by a sequential extraction. The majority of Cu, Pb, and As were bound to OM, whereas the majority of Zn was bound with Fe/Mn oxides for all three SSs. Incubation of SS for 6 mo did not result in a substantial decrease in OM content or a change in the relative distribution of Cu, Zn, Pb, and As. Direct OM oxidation to 30 and 70% by diluted HO resulted in a significant decrease in organically bound Cu but increased its exchangeable, carbonate-bound, and Fe/Mn-bound fractions. Oxidation of OM slightly decreased organically bound Zn but significantly increased exchangeable Zn in all SSs. Oxide- and carbonate-bound Zn also decreased following OM oxidation. Exchangeable fractions of As and Pb were minute before and after OM degradation, indicating that release into the environment would be unlikely. The relative distribution of Cu, Zn, Pb, and As in SS-treated soils was similar to that of SS, suggesting a dominant role of SS properties in controlling metal distribution following OM oxidation. Overall, OM oxidation increased the mobility and bioavailability of Zn and Cu, whereas it had less impact on Pb and As.

Viau, E., K. Bibby, et al. (2011). "Toward a consensus view on the infectious risks associated with land application of sewage sludge." <u>Environ Sci Technol</u> **45**(13): 5459-5469.

The science linking processed sewage sludge (biosolids) land application with human health has improved in the last ten years. The goal of this review is to develop a consensus view on the human health impacts associated with land-applying biosolids. Pre-existing risk studies are integrated with recent advances in biosolids pathogen exposure science and risk analysis. Other than accidental direct ingestion, the highest public risks of infection from land application are associated with airborne exposure. Multiple, independent risk assessments for enteroviruses similarly estimate the yearly probabilities of infection near 10(-4). However, the inclusion of other emerging pathogens, specifically norovirus, increases this yearly infectious risk by over 2 orders of magnitude. Quantitative microbial risk assessment for biosolids exposure more effectively operates as a tool for analyzing how exposure can be reduced rather than being used to assess "safety". Such analysis demonstrates that the tradition of monitoring pathogen quality by Salmonella spp. and enterovirus content underestimates the infectious risk to the public, and that a rigorous biosolids pathogen treatment process, rather than extending community separation distances, is the most efficient method for reducing pathogen exposure and infectious risk.

Sepulvado, J. G., A. C. Blaine, et al. (2011). "Occurrence and Fate of Perfluorochemicals in Soil Following the Land Application of Municipal Biosolids." <u>Environ Sci Technol</u> 2011 Mar 29. [Epub ahead of print].

The recent implementation of soil and drinking water screening guidance values for two perfluorochemicals (PFCs), perfluorooctanoate (PFOA) and perfluorooctane sulfonate (PFOS) by the U.S. Environmental Protection Agency (EPA), reflects the growing concerns regarding the presence of these persistent and bioaccumulative chemicals in the natural environment. Previous work has established the potential risk to the environment from the land application of industrially contaminated biosolids, but studies focusing on environmental risk from land application of typical municipal biosolids are lacking. Thus, the present study investigated the occurrence and fate of PFCs from land-applied municipal biosolids by evaluating the levels, mass balance, desorption, and transport of PFCs in soils receiving application of municipal biosolids at various loading rates. This study is the first to report levels of PFCs in agricultural soils amended with typical municipal biosolids. PFOS was the dominant PFC in both biosolids (80-219 ng/g) and biosolids-amended soil (2-483 ng/g). Concentrations of all PFCs in soil increased linearly with increasing biosolids loading rate. These data were used to develop a model for predicting PFC soil concentrations in soils amended with typical municipal biosolids using cumulative biosolids loading rates. Mass balance calculations comparing PFCs applied vs those recovered in the surface soil interval indicated the potential transformation of PFC precursors. Laboratory desorption experiments indicated that the leaching potential of PFCs decreases with increasing chain length and that previously derived organic-carbon normalized partition coefficients may not be accurate predictors of the desorption of long-chain PFCs from biosolids-amended soils. Trace levels of PFCs were also detected in soil cores from biosolids-amended soils to depths of 120 cm, suggesting potential movement of these compounds within the soil profile over time and confirming the higher transport potential for short-chain PFCs in soils amended with municipal biosolids.

Yoo, H., J. W. Washington, et al. (2010). "Concentrations, distribution, and persistence of fluorotelomer alcohols in sludge-applied soils near Decatur, Alabama, USA." <u>Environ Sci</u><u>Technol</u> **44**(22): 8397-8402.

Soil samples were collected for fluorotelomer alcohol (FTOH) analyses from six fields to which sludge had been applied and one "background" field that had not received sludge. Ten analytes in soil extracts were quantified using GC/MS. Sludge-applied fields had surface soil FTOH concentrations exceeding levels found in the background field. For 8:2nFTOH, which can degrade to perfluorooctanoic acid, impacted surface-soils ranged from 5 to 73 ng/g dry weight, clearly exceeding the background field in which 8:2nFTOH was not detected. The highest [FTOH] generally was 10:2nFTOH, which had concentrations of <5.6 to 166 ng/g. For the first time, we document the persistence of straight-chained primary FTOHs (n-FTOHs) and branch-chained secondary FTOHs (sec-FTOHs), which are transformation products of n-FTOHs, in field soils for at least five years after sludge application. Ratios of

sec-FTOHs to n-FTOHs were highest for 7:2sFTOH/8:2nFTOH (approximately 50%) and decreased with increasing chain length to a minimum for the longest-chained analytes, 13:2sFTOH/14:2nFTOH (approximately 10%). Disappearance half-lives for FTOHs, calculated with these data, ranged from 0.85 to 1.8 years. These analytical results show that the practice of sludge application to land is a pathway for the introduction of FTOHs and, accordingly, their transformation products, perfluorocarboxylic acids, into the environment.

Walters, E., K. McClellan, et al. (2010). "Occurrence and loss over three years of 72 pharmaceuticals and personal care products from biosolids-soil mixtures in outdoor mesocosms." <u>Water Res</u> **44**(20): 6011-6020.

Municipal biosolids are in widespread use as additives to agricultural soils in the United States. Although it is well known that digested sewage sludge is laden with organic wastewater contaminants, the fate and behavior of micropollutants in biosolids-amended agricultural soils remain unclear. An outdoor mesocosm study was conducted in Baltimore, Maryland, to explore the fate of 72 pharmaceuticals and personal care products (PPCPs) over the course of three years in that were placed in plastic containers made from polyvinylchloride and kept exposed to ambient outdoor conditions. Of the 72 PPCPs tested for using EPA Method 1694, 15 were initially detected in the soil/biosolids mixtures at concentrations ranging from low parts-per-billion to parts-per-million levels. The antimicrobials triclocarban and triclosan showed the highest initial concentrations at 2715 and 1265 mug kg(-1), respectively. Compounds showing no discernable loss over three years of monitoring included diphenhydramine, fluoxetine, thiabendazole and triclocarban. The following half-life estimates were obtained for compounds showing first-order loss rates: azithromycin (408-990 d) carbamazepine (462-533 d), ciprofloxacin (1155-3466 d), doxycycline (533-578 d), 4-epitetracycline (630 d), gemfibrozil (224-231 d), norfloxacin (990-1386 d), tetracycline (578 d), and triclosan (182-193 d). Consistent with other outdoor degradation studies, chemical half-lives determined empirically exceeded those reported from laboratory studies or predicted from fate models. Study results suggest that PPCPs shown in the laboratory to be readily biotransformable can persist in soils for extended periods of time when applied in biosolids. This study provides the first experimental data on the persistence in biosolids-amended soils for ciprofloxacin, diphenhydramine, doxycycline, 4-epitetracycline, gemfibrozil, miconazole, norfloxacin, ofloxacin, and thiabendazole.

Viau, E., F. Levi-Schaffer, et al. (2010). "Respiratory toxicity and inflammatory response in human bronchial epithelial cells exposed to biosolids, animal manure, and agricultural soil particulate matter." <u>Environ Sci Technol</u> **44**(8): 3142-3148.

This study investigated cytotoxicity and inflammation caused by human bronchial epithelial cells exposed to respirable aerosols produced during the land application of stabilized sewage sludges (biosolids). BEAS-2B cells were exposed to respirable aerosols (PM(10)) derived from soils, biosolids stabilized by mesophilic anaerobic digestion (MAD), temperature-phased anaerobic digestion (TPAD), and composting (COM) as well as animal manures stabilized by mesophilic anaerobic digestion (AMAD) and composting (ACOM). Anaerobically digested particles (MAD, TPAD, AMAD) induced the highest cytotoxicity with LD(50) levels of 70 microg/cm(2), 310 microg/cm(2) for, and 375 microg/cm(2) for MAD, AMAD, and TPAD, respectively. Conversely, there was no observed cytotoxicity for soils, composted biosolids, or composted manures at the in vitro doses tested. Inflammatory

responses, measured by interleukin (IL)-6 and IL-8 release, were 2- to 15-fold greater in biosolids and manures than for equivalent doses in soils. Biosolids treatment rankings for human bronchial epithelial cell toxicity and inflammation were similar to the rankings found in recent biosolids pathogen content studies-from lowest pathogen content or toxicity to highest, rankings were as follows: COM < TPAD < MAD. Coupling in vitro responses with modeled tracheobronchial lung surface doses that may occur during a biosolids land application event suggests that an inflammatory aerosol exposure in the TB region could only occur under worst case scenarios (exercising human with reduced lung capacity at <65 m setbacks), but examination of lower in vitro doses as well as consideration of the head and lower lung respiratory tract regions are needed to more definitively describe the links between biosolids aerosols and the potential for respiratory inflammation.

Banasik, M., M. Hardy, et al. (2009). "Provisional human health risk assessment of PBDEs in sewage sludge used for agricultural purposes." <u>Chemosphere</u> **77**(5): 699-701; author reply 702-693.

Abstract not available.

Snyder, E. H., G. A. O'Connor, et al. (2010). "Fate of 14Ca triclocarban in biosolids-amended soils." <u>Science of the Total Environment</u> **408**(13): 2726-2732.

Triclocarban (TCC) is an antibacterial compound commonly detected in biosolids at partsper-million concentrations. Approximately half of the biosolids produced in the United States are land-applied, resulting in a systematic release of TCC into the soil environment. The extent of biosolids-borne TCC environmental transport and potential human/ecological exposures will be greatly affected by its bioavailability and the rate of degradation in amended soils. To investigate these factors, radiolabeled TCC (14Ca"TCC) was incorporated into anaerobically digested biosolids, amended to two soils, and incubated under aerobic conditions. The evolution of 14CO2 (biodegradation) and changes in chemical extractability (bioavailability) was measured over time. Water extractable TCC over the study period was low and significantly decreased over the first 3weeks of the study (from 14% to 4% in a fine sand soil and from 3 to <1% in a silty clay loam soil). Mineralization (i.e. ultimate degradation), as measured by evolution of 14CO2, was <4% over 7.5months. Methanol extracts of the amended soils were analyzed by radiolabel thin-layer chromatography (RAD-TLC), but no intermediate degradation products were detected. Approximately 20% and 50% of the radioactivity in the amended fine sand and silty clay loam soils, respectively, was converted to bound residue as measured by solids combustion. These results indicate that biosolids-borne TCC becomes less bioavailable over time and biodegrades at a very slow rate.

Holt, L. M., A. E. Laursen, et al. (2010). "Effects of land application of municipal biosolids on nitrogen-fixing bacteria in agricultural soil." <u>Biology and Fertility of Soils</u> **46**(4): 407-413. [Canadian Article]

Effects of municipal biosolids on microbial N sub(2)-fixation in agricultural soil were assessed in a 3-month laboratory study which included analysis of metals, pharmaceuticals, and personal care products. Reference agricultural soil was amended with organic manure or municipal biosolids from a southern Ontario wastewater plant, with a biosolids-only

treatment included to evaluate metabolic activity in this inoculum. Microbial N sub(2)fixation in reference and manure-amended soils were similar (p=0.144) over 3months and lower than in biosolids-amended soil (p=0.001); however, differences among soil treatments decreased over time, with no significant difference at test termination. In general, one-time application of biosolids caused short-term stimulation of N sub(2)-fixing activity with a return to reference conditions within 3months. Although no residual effects were detected, biosolids introduced elevated levels of metals, pharmaceuticals, personal care products (particularly analgesics), and viable bacteria; long-term effects caused by repeat applications (as commonly practiced) requires further investigation.

Gillis, J. and G. Price (2009). Evaluation of Anthropogenic Organic Contaminant Concentrations and Soil Chemical Properties over Time in a Field Soil Amended with an Alkaline- Stabilized Biosolid. 2009 Annual Meetings of the American Society of Agronomy, Crop Science Society of America and Soil Science Society of America (ASA-CSSA-SSSA 2009).

Abstract not available.

Hundal, L., K. Xia, et al. (2009). <u>Long-Term Assessments of Microconstituents Fate in</u> <u>Biosolids-Amended Soils</u>. 2009 Conference on Residuals and Biosolids.

Abstract not available.

Brooks, J., C. Gerba, et al. (2009). <u>Comparative Microbial Risks of Land Applied Biosolids and Animal Manure</u>. 2009 Conference on Residuals and Biosolids.

Technical Abstract: The transmission of pathogens by land application of untreated human and animal wastes has been known for more than 100 years. In the United States there are more than 450,000 (EPA) animal feeding operations producing more than 100 million tons of animal manure per year. In addition, grazing animals also deposit large quantities of manure on range land. This study attempts to look at the relative risks of pathogens in biosolids vs. animal manure applied to land using a quantitative microbial risk assessment approach. This process involves four basic steps: pathogen identification, exposure assessment, doseresponse and risk characterization. Several examples which were evaluated included risk to workers, produce crops, and children playing in a field. While some of these exposures may be considered worst case for biosolids because of site and application restrictions, they can commonly occur in areas where manure is applied. The risks are largely determined by the degree of treatment that the manure receives before land application, but risks compared to biosolids can be comparable, depending upon assumptions used in the risk model. Generally, annual risks of infection were greater than the accepted 1:10000 chance of infection for bacteria (applied manures) and viruses (biosolids) when land applied and only allowed an 1month decay time period. Using more appropriate conditions of soil decay (> 6 months), land application of either residual resulted in risks below the accepted risk. This analysis can provide insight on the relative risks of animal waste and biosolid land application that will allow for a better grasp of the risks to the public and industry.

Viau, E. J. (2009). Human pathogenic and lung inflammatory aerosol exposures associated with the land application of biosolids.

Land application of biosolids (treated sewage sludge) to agricultural fields generates a significant emission of respirable aerosols that can move off-site and expose downwind

communities to biosolids constituents. Accurate estimation of infectious and respiratory health risks associated with this practice requires better knowledge of pathogen content and lung inflammatory potential for biosolids of class A and class B quality. Biosolids from twenty-nine U.S. utilities were surveyed for resistant bacterial and viral pathogens, associated indicators, and lung inflammatory agents (e.g., endotoxins)--four stabilization processes were targeted including class B mesophilic anaerobic digestion, and class A temperature-phased anaerobic digestion, composting, and heat pelletization. Both culturable indicator and qPCRbased concentrations of human adenovirus spp., Legionella pneumophila, Staphylococcus aureus, and Clostridium difficile were significantly lower and detected less frequently in class A biosolids, while lung inflammatory agents were comparable between the two classes. Enterococci qPCR was more indicative of pathogen inactivation behavior than fecal coliforms through biosolid treatment and could circumvent specificity issues associated with using multi-species indicators when monitoring in complex biosolid microbiological communities. Pathogen densities in bulk biosolids were translated to an aerosol inhalation dose for a land application event using previously calibrated aerosol reconstruction, Gaussian aerosol transport, and intermittent exposure time models. Pathogen aerosol dose was affected more by biosolid treatments than separation distance from the biosolid field--using class A composting over class B MAD resulted in 2.6 log exposure reductions, while 165-500 m separation distances reduced exposure by 0.5-1.3 logs for a worst-case aerosol scenario. To further elucidate human health effects from a biosolid aerosol exposure, in vitro experiments exposed bronchial epithelial cells to respirable biosolids, animal manure, and soils. Lung cell cytotoxicity and induction of inflammatory cytokines were significantly increased in biosolid and animal manure exposures compared to agricultural soils--lung cytotoxicity results followed pathogen content trends, with MAD biosolids showing the highest cytotoxicity. Research outcomes provide a scientific basis for updating U.S. biosolid regulations--biosolid aerosol exposures could be decreased by 4-logs through mandating class A composting or similar treatment and requiring a minimum of 150 m separation distance between biosolidapplied fields and downwind communities.

Sidhu, J. P. and S. G. Toze (2009). "Human pathogens and their indicators in biosolids: a literature review." <u>Environ Int</u> **35**(1): 187-201.

A growing beneficial reuse of biosolids in agriculture has led to concerns about potential contamination of water resources and the food chain. In order to comprehend the potential risks of transmission of diseases to the human population, an advanced quantitative risk assessment is essential. This requires good quantitative data which is currently limited due to the methodological limitations. Consequently, further development and standardization of methodologies for the detection, enumeration and viability assessment of pathogens in biosolids is required. There is a paucity of information on the numbers and survival of enteric virus and protozoan pathogens of concern in biosolids. There is a growing urgency for the identification of more reliable alternative indicators, both index and model microorganisms, which could be used for potential public health risk assessment. In this review, we have summarized reported literature on the numbers and fate of enteric pathogens and indicators in biosolids. The advantages and limitations of the use of conventional and alternative index and model microorganisms for the prediction of pathogen presence in biosolids are also discussed.

Zerzghi, H., C. P. Gerba, et al. (2010). "Long-term effects of land application of class B biosolids on the soil microbial populations, pathogens, and activity." J Environ Qual 39(1): 402-408.

This study evaluated the influence of 20 annual land applications of Class B biosolids on the soil microbial community. The potential benefits and hazards of land application were evaluated by analysis of surface soil samples collected following the 20th land application of biosolids. The study was initiated in 1986 at the University of Arizona Marana Agricultural Center, 21 miles north of Tucson, AZ. The final application of biosolids was in March 2005, followed by growth of cotton (Gossypium hirsutum L.) from April through November 2005. Surface soil samples (0-30 cm) were collected monthly from March 2005, 2 wk after the final biosolids application, through December 2005, and analyzed for soil microbial numbers. December samples were analyzed for additional soil microbial properties. Data show that land application of Class B biosolids had no significant long-term effect on indigenous soil microbial numbers including bacteria, actinomycetes, and fungi compared to unamended control plots. Importantly, no bacterial or viral pathogens were detected in soil samples collected from biosolid amended plots in December (10 mo after the last land application) demonstrating that pathogens introduced via Class B biosolids only survived in soil transiently. However, plots that received biosolids had significantly higher microbial activity or potential for microbial transformations, including nitrification, sulfur oxidation, and dehydrogenase activity, than control plots and plots receiving inorganic fertilizers. Overall, the 20 annual land applications showed no long-term adverse effects, and therefore, this study documents that land application of biosolids at this particular site was sustainable throughout the 20-yr period, with respect to soil microbial properties.

White, J. W., F. J. Coale, et al. (2010). "Phosphorus runoff from waste water treatment biosolids and poultry litter applied to agricultural soils." <u>J Environ Qual</u> **39**(1): 314-323.

Differences in the properties of organic phosphorus (P) sources, particularly those that undergo treatment to reduce soluble P, can affect soil P solubility and P transport in surface runoff. This 2-yr field study investigated soil P solubility and runoff P losses from two agricultural soils in the Mid-Atlantic region after land application of biosolids derived from different waste water treatment processes and poultry litter. Phosphorus speciation in the biosolids and poultry litter differed due to treatment processes and significantly altered soil P solubility and dissolved reactive P (DRP) and bioavailable P (FeO-P) concentrations in surface runoff. Runoff total P (TP) concentrations were closely related to sediment transport. Initial runoff DRP and FeO-P concentrations varied among the different biosolids and poultry litter applied. Over time, as sediment transport declined and DRP concentrations became an increasingly important component of runoff FeO-P and TP, total runoff P was more strongly influenced by the type of biosolids applied. Throughout the study, application of lime-stabilized biosolids and poultry litter increased concentrations of soil-soluble P, readily desorbable P, and soil P saturation, resulting in increased DRP and FeO-P concentrations in runoff. Land application of biosolids generated from waste water treatment processes that used amendments to reduce P solubility (e.g., FeCl(3)) did not increase soil P saturation and reduced the potential for DRP and FeO-P transport in surface runoff. These results illustrate the importance of waste water treatment plant process and determination of specific P source coefficients to account for differential P availability among organic P sources.

Snyder, E. H., G. A. O'Connor, et al. (2011). "Toxicity and bioaccumulation of biosolids-borne triclocarban (TCC) in terrestrial organisms." <u>Chemosphere</u> **82**(3): 460-467.

Triclocarban (TCC) toxicity and bioaccumulation data are primarily limited to direct human and animal dermal exposures, animal ingestion exposures to neat and feed-spiked TCC, and/or aquatic organism exposures. Three non-human, terrestrial organism groups anticipated to be the most highly exposed to land-applied, biosolids-borne TCC are soil microbes, earthworms, and plants. The three ecological receptors are expected to be at particular risk due to unique modes of exposure (e.g. constant, direct contact with soil; uptake of amended soil and pore water), inherently greater sensitivity to environmental contaminants (e.g. increased body burdens, permeable membranes), and susceptibility to minute changes in the soil environment. The toxicities of biosolids-borne TCC to Eisenia fetida earthworms and soil microbial communities were characterized using adaptations of the USEPA Office of Prevention, Pesticides, and Toxic Substances (OPPTS) Guidelines 850.6200 (Earthworm Subchronic Toxicity Test) and 850.5100 (Soil Microbial Community Toxicity Test), respectively. The resultant calculated TCC LC50 value for E. fetida was 40 mg TCC kg amended fine sand(-1). Biosolids-borne TCC in an amended fine sand had no significant effect on soil microbial community respiration, ammonification, or nitrification. Bioaccumulation of biosolids-borne TCC by E. fetida and Paspulum notatum was measured to characterize potential biosolids-borne TCC movement through the food chain. Dry-weight TCC bioaccumulation factor (BAF) values in E. fetida and P. notatum ranged from 5.2-18 and 0.00041-0.007 (gsoil gtissue(-1)), respectively.

Kwon, J. W., K. L. Armbrust, et al. (2010). "Transformation of triclosan and triclocarban in soils and biosolids-applied soils." <u>J Environ Qual</u> **39**(4): 1139-1144.

Triclosan (TCS) and triclocarban (TCC), widely used as antibacterial agents, have been frequently detected in biosolids. Biosolids land application may introduce pharmaceuticals and personal care products (PPCPs) such as TCS and TCC into the environment. Microcosm studies were conducted to investigate TCS and TCC transformation in Marietta fine loam and McLaurin coarse loam. Both compounds were spiked into the soils with and without biosolids amendment under non-sterilized and sterilized conditions and incubated aerobically at 30 degrees C for up to 100 d. In both soils, transformation of TCS followed second-order reaction kinetics, with estimated reaction rate constants of $(5.27 + 0.920) \times 10(-1)$ and (9.13)+/-1.58) x 10(-2) (mg kg(-1))(-1) d(-1) for Marietta fine loam and McLaurin coarse loam, respectively. Transformation of TCC in both soils was slower than that for TCS. After 100 d, 53 +/- 1% and 71 +/- 2% of the initially added TCC and only 2.8 +/- 0.35% and 6.2 +/-0.80% of initially added TCS remained in Marietta fine loam and McLaurin coarse loam, respectively. The transformation of both compounds were faster in the Marietta fine loam (pH 7.8; 1.8% organic matter) than in the McLaurin coarse loam (pH 4.7; 0.65% organic matter). Our result suggests that biotic processes are more of a controlling factor affecting TCS transformation, whereas abiotic processes may affect TCC transformation more significantly. Addition of biosolids to the two soils slowed the transformation of both compounds, indicating interactions between both compounds and biosolids may adversely affect their transformation in soils, an important factor that must be included in models predicting environmental fate of biosolids-associated PPCPs.

Fuchsman, P., J. Lyndall, et al. (2010). "Terrestrial ecological risk evaluation for triclosan in land-applied biosolids." <u>Integr Environ Assess Manag</u> **6**(3): 405-418.

Triclosan is an antimicrobial compound found in many consumer products including soaps and personal care products. Most triclosan is disposed of down household drains, whereupon it is conveyed to wastewater treatment plants. Although a high percentage of triclosan biodegrades during wastewater treatment, most of the remainder is adsorbed to sludge, which may ultimately be applied to land as biosolids. We evaluated terrestrial ecological risks related to triclosan in land-applied biosolids for soil microbes, plants, soil invertebrates, mammals, and birds. Exposures are estimated using a probabilistic fugacity-based model. Triclosan concentrations in biosolids and reported biosolids application rates are compiled to support estimation of triclosan concentrations in soil. Concentrations in biota tissue are estimated using an equilibrium partitioning model for plants and worms and a steady-state model for small mammals; the resulting tissue concentrations are used to model mammalian and avian dietary exposures. Toxicity benchmarks are identified from a review of published and proprietary studies. The results indicate that adverse effects related to soil fertility (i.e., disruption of nitrogen cycling) would be expected only under "worst-case" exposures, under certain soil conditions and would likely be transient. The available data indicate that adverse effects on plants, invertebrates, birds, and mammals due to triclosan in land-applied biosolids are unlikely.

Staples, C., U. Friederich, et al. (2010). "Estimating potential risks to terrestrial invertebrates and plants exposed to bisphenol A in soil amended with activated sludge biosolids." <u>Environ Toxicol</u> <u>Chem</u> **29**(2): 467-475.

Bisphenol A (BPA) is a high production volume substance primarily used to produce polycarbonate plastic and epoxy resins. During manufacture and use, BPA may enter wastewater treatment plants. During treatment, BPA may become adsorbed to activated sludge biosolids, which may expose soil organisms to BPA if added to soil as an amendment. To evaluate potential risks to organisms that make up the base of the terrestrial food web (i.e., invertebrates and plants) in accordance with international regulatory practice, toxicity tests were conducted with potworms (Enchytraeids) and springtails (Collembolans) in artificial soil, and six plant types using natural soil. No-observed-effect concentrations (NOEC) for potworms and springtails were equal to or greater than 100 and equal to or greater than 500 mg/kg (dry wt), respectively. The lowest organic matter-normalized NOEC among all tests (dry shoot weight of tomatoes) was 37 mg/kg-dry weight. Dividing by an assessment factor of 10, a predicted-no-effect concentration in soil (PNEC(soil)) of 3.7 mg/kg-dry weight was calculated. Following international regulatory guidance, BPA concentrations in soil hypothetically amended with biosolids were calculated using published BPA concentrations in biosolids. The upper 95th percentile BPA biosolids concentration in North America is 14.2 mg/kg-dry weight, and in Europe is 95 mg/kg-dry weight. Based on recommended biosolids application rates, predicted BPA concentrations in soil (PEC(soil)) would be 0.021 mg/kg-dry weight for North America and 0.14 mg/kg-dry weight for Europe. Hazard quotients (ratio of PEC(soil) and PNEC(soil)) for BPA were all equal to or less than 0.04. This indicates that risks to representative invertebrates and plants at the base of the terrestrial food web are low if exposed to BPA in soil amended with activated sludge biosolids.

Bai, Y., W. Chen, et al. (2010). "Uptake of metals by food plants grown on soils 10 years after biosolids application." J Environ Sci Health B **45**(6): 531-539.

Potentially hazardous trace elements such as Cd, Cu, Cr, Ni and Zn are expected to accumulate in biosolids-amended soil and remain in the soil for a long period of time. In this research, uptake of metals by food plants including cabbage, carrot, lettuce and tomato grown on soils 10 years after biosolids application was studied. All the five metals were significantly accumulated in the biosolids-amended soils. The accumulation of metal in soil did not result in significant increase in concentrations of Cu, Cr and Ni in the edible plant tissues. However, the Cd and Zn concentrations of the edible tissues of plants harvested from the biosolids receiving soils were significantly enhanced in comparison with those of the unaffected soils. The plant uptake under Greenfield sandy loam soil was generally higher than those under the Domino clayey loam soil. The metal concentration of edible plant tissue exhibited increases were plant species dependent. The indigenous soil metals were absorbed by the plants in much higher rates than those of the biosolids-receiving soils. It appeared that the plant uptake of the indigenous soil-borne metal and the added biosolids-borne metals are independent of one another and mathematically are additive.

Antonious, G. F., J. C. Snyder, et al. (2010). "Heavy metals in summer squash fruits grown in soil amended with municipal sewage sludge." <u>J Environ Sci Health B</u> **45**(2): 167-173.

The increasing awareness of the value of vegetables and fruits in the human diet requires monitoring of heavy metals in food crops. The effects of amending soil with compost made from municipal sewage sludge (MSS) and MSS mixed with yard waste (MSS-YW) on Cd, Cr, Mo, Cu, Zn, Pb, and Ni concentrations in soil and the potential bioaccumulation of heavy metals in squash fruits at harvest were investigated. A field study was conducted in a siltyloam soil at Kentucky State University Research Farm. Eighteen plots of 22 x 3.7 m each were separated using metal borders and the soil in six plots was mixed with MSS at 15 t acre(-1), six plots were mixed with MSS-YW at 15 t acre(-1) (on dry weight basis), and six unamended plots (no-mulch) were used for comparison purposes. Plots were planted with summer squash and heavy metals were analyzed in soil and mature fruits at harvest. Analysis of heavy metals in squash fruits was conducted using inductively coupled plasma spectrometry. Zinc and Cu concentrations in soil mixed with MSS were extremely high compared to other metals. In squash fruits, concentrations of Zn were generally greater than Cu. Total squash marketable yield was greatest in MSS-YW and MSS treatments compared to no-mulch conventional soil. Concentrations of Cd and Pb in soil amended with MSS averaged 0.1 and 1.4 mg kg(-1), respectively. These levels were much lower than the limits in the U.S. guidelines for using MSS in land farming. Data revealed that maximum concentrations of Cd and Pb in squash fruits were 0.03 and 0.01 microg g(-1) dry fruit, respectively. Nickel concentration in squash fruits fluctuated among harvest dates reaching a maximum of 2.5 microg g(-1) dry fruit. However, these concentrations were far below their permissible limits in edible fruits.

Sabourin, L., A. J. Al-Rajab, et al. (2011). "Fate of the antifungal drug clotrimazole in agricultural soil." <u>Environ Toxicol Chem</u> **30**(3): 582-587. [Canadian Article]

Clotrimazole is a broad-spectrum antimycotic drug used for the treatment of dermatological and gynecological infections; it is incompletely broken down during sewage treatment and

could potentially reach agricultural land through the application of municipal biosolids or wastewater. In the absence of any environmental fate data, we evaluated the persistence and dissipation pathways of (3)H-clotrimazole during laboratory incubations of agricultural soils. Clotrimazole was removed from a loam (time to dissipate 50% = 68 d), a sandy loam (time to dissipate 50% = 36 d), and a clay loam (time to dissipate 50% = 55 d), with formation of nonextractable residues being the major sink for (3) H. Their parent compound had no significant mineralization, as evidenced by the lack of formation of (3) H(2) O. Up to 15% of the applied radioactivity was recovered in the form of [(3)H]-(2-chlorophenyl)diphenyl methanol. The rate of clotrimazole dissipation in the loam soil did not vary with moisture content, but it was slower at a lower temperature (number of days to dissipate 50% = 275.6 d at 4 degrees C). Addition of municipal biosolids to the loam soil did not vary the clotrimazole dissipation rate. In summary, the present study has established that clotrimazole is dissipated in soil, at rates that varied with soil texture and temperature. Clotrimazole dissipation was accompanied by the formation of nonextractable residues and detectable extractable residues of the transformation product (2-chlorophenyl)diphenyl methanol.

Mashtare, M. L., B. Khan, et al. (2011). "Evaluating stereoselective sorption by soils of 17alphaestradiol and 17beta-estradiol." <u>Chemosphere</u> **82**(6): 847-852.

The application of manure and biosolids onto agricultural land has increased the risk of estrogenic exposure to aquatic systems. Both alphaE2 and betaE2 have been routinely detected in surface and ground waters with higher concentrations reported near concentrated animal feeding operations and agricultural fields. Although movement through the soil to a water body is highly dependent on hormone-soil interactions, to date, only the interaction of betaE2 with soils has been characterized despite alphaE2 often being the more common form excreted by livestock such as beef cattle and dairy. In predicting the transport of estradiol, sorption characteristics for the stereoisomers have been assumed to be the same. To evaluate this assumption, sorption of alphaE2 and betaE2 was measured on seven surface soils representing a range in soil properties. Soils were autoclave-sterilized to minimize loss due to biotransformation, and both solution and soil phase concentrations were measured. Overall, E2 sorption is best correlated to soil organic carbon (OC) with an average log OC-normalized distribution coefficient (logKoc, L kgoc(-1)) of 2.97+/-0.13 for alphaE2 and 3.14+/-0.16 for betaE2 with betaE2 consistently exhibiting higher sorption than alphaE2 with the highest beta/alpha sorption ratio of 1.9. Assuming that the two isomers sorb the same is not a conservative decision making approach. The lower sorption affinity of alphaE2 increases the likelihood that it will be leached from agricultural fields.

Gottschall, N., E. Topp, et al. (2010). "Polybrominated diphenyl ethers, perfluorinated alkylated substances, and metals in tile drainage and groundwater following applications of municipal biosolids to agricultural fields." <u>Sci Total Environ</u> **408**(4): 873-883. [Canadian Article]

Polybrominated diphenyl ethers (PBDEs), perfluorinated alkylated substances (PFAS), and metals were monitored in tile drainage and groundwater following liquid (LMB) and dewatered municipal biosolid (DMB) applications to silty-clay loam agricultural field plots. LMB was applied (93,500 L ha(-1)) in late fall 2005 via surface spreading on un-tilled soil (SS(LMB)), and a one-pass aerator-based pre-tillage prior to surface spreading (AerWay SSD) (A). The DMB was applied (8 Mg d wha(-1)) in early summer 2006 on the same plots by injecting DMB beneath the soil surface (DI), and surface spreading on un-tilled soil

(SS(DMB)). Key PBDE congeners (BDE-47, -99, -100, -153, -154, -183, -209) comprising 97% of total PBDE in LMB, had maximum tile effluent concentrations ranging from 6 to 320 ng L(-1) during application-induced tile flow. SS(LMB) application-induced tile mass loads for these PBDE congeners were significantly higher than those for control (C) plots (no LMB) (p<0.05), but not A plots (p>0.05). PBDE mass loss via tile (0-2h post-application) as a percent of mass applied was approximately 0.04-0.1% and approximately 0.8-1.7% for A and SS(LMB), respectively. Total PBDE loading to soil via LMB and DMB application was 0.0018 and 0.02 kg total PBDE ha(-1)yr(-1), respectively. Total PBDE concentration in soil (0-0.2m) after both applications was 115 ng g(-1)dw, (sampled 599 days and 340 days post LMB and DMB applications respectively). Of all the PFAS compounds, only PFOS (max concentration=17 ng L(-1)) and PFOA (12 ng L(-1)) were found above detectable limits in tile drainage from the application plots. Mass loads of metals in tile for the LMB applicationinduced tile hydrograph event, and post-application concentrations of metals in groundwater, showed significant (p<0.05) land application treatment effects (SS(LMB)>A>C for tile and SS(LMB) and A>C for groundwater for most results). Following DMB application, no significant differences in metal mass loads in tile were found between SS(DMB) and DI treatments (PBDE/PFAS were not measured). But for many metals (Cu, Se, Cd, Mo, Hg and Pb) both SS(DMB) and DI loads were significantly higher than those from C, but only during <100 days post DMB application. Clearly, pre-tilling the soil (e.g., A) prior to surface application of LMB will reduce application-based PBDE and metal contamination to tile drainage and shallow groundwater. Directly injecting DMB in soil does not significantly increase metal loading to tile drains relative to SS(DMB), thus, DI should be considered a DMB land application option.

Antonious, G. F. (2009). "Enzyme activities and heavy metals concentration in soil amended with sewage sludge." J Environ Sci Health A Tox Hazard Subst Environ Eng **44**(10): 1019-1024.

Municipal sewage sludge (MSS) and yard waste compost (YWC) provide amendments useful for improving soil structure and nutrient status. However, soil amendments contain heavy metals that may potentially affect soil microbes and the enzymes they produce. A field study was conducted using three soil management practices (MSS, YWC, and native soil). Broccoli (Brassica oleracea L.) seedlings were planted, and the activities of the enzymes hydrolyzing urea (urease), sucrose (invertase), and p-nitrophenyl phosphate (acid and alkaline phosphatase) were determined in spring and fall agricultural soil. The greater soil urease and invertase activities in spring soil amended with MSS provided evidence of increased soil microbial population. On the contrary, the application of YWC in spring did not alter soil urease or invertase activities to any appreciable extent. Overall acid and alkaline phosphatase were stimulated in soil amended with YWC. Nickel, Zn, and Cu increased in soil amended with MSS while, concentration of Pb increased after addition of YWC to native soil. Nickel and Pb were taken up by broccoli plants grown in MSS amended soil, but their concentration in broccoli heads were below the Codex Commission Allowable Limits.

Al-Rajab, A. J., L. Sabourin, et al. (2009). "Impact of biosolids on the persistence and dissipation pathways of triclosan and triclocarban in an agricultural soil." <u>Sci Total Environ</u> **407**(23): 5978-5985. [Canadian Article]

The broad spectrum antimicrobial agents triclosan (TCS) and triclocarban (TCC) are widely used in many personal care products. Knowledge concerning the fate of these two

compounds in different environmental matrices is scarce. In this study, the fate of TCS and TCC in soil following direct addition, or when residues were applied via either liquid municipal biosolids (LMB) or dewatered municipal biosolids (DMB) was investigated in laboratory dissipation experiments and under outdoor conditions using radioisotope methods. In laboratory incubations, (14)C-TCC or (14)C-TCS was added to microcosms containing a loam soil and the rate of (14)CO(2) accumulation and loss of solvent-extractable (14)C were determined during incubation at 30 degrees C. Compared to when TCC or TCS was added directly to soil, both chemicals were mineralized more rapidly when applied in LMB, and both were mineralized more slowly when applied in DMB. The application matrix had no effect on the rate of removal of extractable residues. In field experiments, parent compounds were incorporated directly in soil, incorporated via LMB, or a single aggregate of amended DMB was applied to the soil surface. During the experiment soil temperatures ranged from 20 degrees C to 10 degrees C. Dissipation was much slower in the field than in the laboratory experiments. Removal of non-extractable residues was faster in the presence of LMB than the other treatments. Recovery of extractable and non-extractable residues suggested that there was little atmospheric loss of (14)C. Triclocarban readily formed non-extractable residues with DMB whereas TCS did not. Overall, this study has identified that both the pathways and the kinetics of TCS and TCC dissipation in soil are different when the chemicals are carried in biosolids compared to when these chemicals are added directly to the soil.

Edwards, M., E. Topp, et al. (2009). "Pharmaceutical and personal care products in tile drainage following surface spreading and injection of dewatered municipal biosolids to an agricultural field." <u>Sci Total Environ</u> **407**(14): 4220-4230. [Canadian Article]

Land application of municipal biosolids can be a source of environmental contamination by pharmaceutical and personal care products (PPCPs). This study examined PPCP concentrations/temporally discrete mass loads in agricultural tile drainage systems where two applications of biosolids had previously taken place. The field plots received liquid municipal biosolids (LMB) in the fall of 2005 at an application rate of approximately 93,500 L ha (-1), and a second land application was conducted using dewatered municipal biosolids (DMB) applied at a rate of approximately 8Mg dw ha (-1) in the summer of 2006 [corrected]. The DMB land application treatments consisted of direct injection (DI) of the DMB beneath the soil surface at a nominal depth of approximately 0.11 m, and surface spreading (SS) plus subsequent tillage incorporation of DMB in the topsoil (approximately 0.10 m depth). The PPCPs examined included eight pharmaceuticals (acetaminophen, fluoxetine, ibuprofen, gemfibrozil, naproxen, carbamazepine, atenolol, sulfamethoxazole), the nicotine metabolite cotinine, and two antibacterial personal care products triclosan and triclocarban. Residues of naproxen, cotinine, atenolol and triclosan originating from the fall 2005 LMB application were detected in tile water nearly nine months after application (triclocarban was not measured in 2005). There were no significant differences (p>0.05) in PPCP mass loads among the two DMB land application treatments (i.e., SS vs. DI); although, average PPCP mass loads late in the study season (>100 days after application) were consistently higher for the DI treatment relative to the SS treatment. While the concentration of triclosan (approximately 14,000 ng g(-1) dw) in DMB was about twice that of triclocarban (approximately 8000 ng g(-1) dw), the average tile water concentrations for triclosan were much higher (43+/-5 ng L(-1)) than they were for triclocarban (0.73+/-0.14 ng L(-1)).

Triclosan concentrations (maximum observed in 2006 approximately 235 ng L(-1)) in tile water resulting from land applications may warrant attention from a toxicological perspective.

Cha, J. and A. M. Cupples (2009). "Detection of the antimicrobials triclocarban and triclosan in agricultural soils following land application of municipal biosolids." <u>Water Res</u> **43**(9): 2522-2530.

The occurrence of the antimicrobials triclocarban (TCC) and triclosan (TCS) was investigated in agricultural soils following land application of biosolids using liquid chromatography-tandem mass spectrometry (LC-MS-MS) with negative ion multimode ionization. The method detection limits were 0.58 ng TCC/g soil, 3.08 ng TCC/g biosolids, 0.05 ng TCS/g soil and 0.11 ng TCS/g biosolids and the average recovery from all of the sample matrices was >95%. Antimicrobial concentrations in biosolids from three Michigan wastewater treatment plants (WWTPs) ranged from 4890 to 9280 ng/g, and from 90 to 7060 ng/g, for TCC and TCS respectively. Antimicrobial analysis of soil samples, collected over two years, from ten agricultural sites previously amended with biosolids, indicated TCC was present at higher concentrations (1.24-7.01 ng/g and 1.20-65.10 ng/g in 2007 and 2008) compared to TCS (0.16-1.02 ng/g and from the method detection limit, <0.05-0.28 ng/g in 2007 and 2008). Soil antimicrobial concentrations could not be correlated to any soil characteristic, or to the time of last biosolids application, which occurred in either 2003, 2004 or 2007. To our knowledge, our data represent the first report of TCC, and the first comparison of TCC and TCS concentrations, in biosolids-amended agricultural soils. Such information is important because approximately 50% of US biosolids are land applied, therefore, any downstream effects of either antimicrobial are likely to be widespread.

Banaitis, M. R., I. J. Fernandez, et al. (2009). "Biogeochemical response of a northeastern forest ecosystem to biosolids amendments." J Environ Qual **38**(2): 792-803.

In the northeastern United States interest in the use of biosolids on forest lands is growing due to the prevalence of extensive forests and market incentives for waste disposal, yet much of the regulatory framework for biosolids land application is based on agronomic practice. This study evaluated the response of soils in a young (approximately 20 yr old) deciduous forest to lime-stabilized biosolids amendments focusing on (i) the temporal and spatial evolution of the pH response, (ii) soil exchangeable cation response, (iii) the risk of trace metal accumulations, and (iv) a bioindicator of treatments (i.e., foliar chemistry). Eighteen plots were established in two study phases with lime-stabilized biosolids loading targets of 0 (control), 4.5, 6.7, 13.4, 20.2, 26.9, and 33.6 Mg (megagram) calcium carbonate equivalents (CCE) ha(-1), with the lowest target rate of addition representing the current regulated loading limit for forest biosolids applications in Maine. The pH of the O horizon increased immediately >2 pH units, and then declined with time, while B horizon pH increased gradually, taking over 1 yr to achieve approximately 1.0 pH unit increase at the highest loading target. O-horizon exchangeable Ca concentration increases dominated soil chemical change and resulted in decreases in exchangeable H and Al. Few significant increases in soil trace metal concentrations had occurred at any soil depth after 1 yr of treatment. Foliar response generally reflected changes in soil chemistry, with Ca concentration increases most significant. This research provides critical insights on forest soil response to application of lime-stabilized biosolids and suggests opportunities for higher loading targets in forests

should be examined.

Shober, A. L. and J. T. Sims (2009). "Evaluating phosphorus release from biosolids and manureamended soils under anoxic conditions." <u>J Environ Qual</u> **38**(1): 309-318.

The solubility of P in biosolids and manures has been shown to influence the potential for dissolved P losses in runoff and leachate when these materials are land applied. As a result, some Mid-Atlantic US states have developed P source coefficients (PSCs) to account for differences in P solubility between fertilizers, manures, and biosolids in P risk assessment tools. The reliability of these PSCs has not been evaluated under anoxic conditions, where environmental changes may affect the P solubility of biosolids or manures. The objective of this study was to assess the effects of anoxic conditions on the release of P from a range of Mid-Atlantic soils amended with manures and biosolids. The concentration of dissolved P released into solution (0.01 mol L(-1) NaCl) from the Pamunkey, Berks, and Manor soils was significantly lower under reducing conditions than under oxidized conditions (median DeltaP = -0.70, -0.49, and -0.07 mg L(-1), respectively; all significant at the 0.001 probability level). There was no significant P source effect on dissolved P released into solution after anoxic incubation of soils. Calculated solubility diagrams and increases in oxalate-extractable Fe and P sorption index under reducing conditions for all soils suggest the precipitation of (i) an Fe(II)-oxide that increased the P sorption capacity of the soils or (ii) an Fe(II)-phosphate that decreased the solubility of P. We propose that current PSCs do not need alteration to account for differences in P solubility of organic sources under reducing conditions under relatively static conditions (e.g., seasonable high water table, periodically submerged soils, stagnant drainage ditches).

Giudice, B. D. and T. M. Young (2011). "Mobilization of endocrine disrupting chemicals and estrogenic activity in simulated rainfall runoff from land-applied biosolids." <u>Environ Toxicol</u> <u>Chem</u> 2011 Jul 22. doi: 10.1002/etc.631. [Epub ahead of print].

Municipal biosolids are commonly applied to land as soil amendment or fertilizer as a form of beneficial reuse of what could otherwise be viewed as waste. Balanced against this benefit are potential risks to groundwater and surface water quality from constituents that may be mobilized during storm events. The objective of the present study was to characterize the mobilization of selected endocrine disrupting compounds (EDCs), heavy metals, and total estrogenic activity in rainfall runoff from land-applied biosolids. Rainfall simulations were conducted on soil plots amended with biosolids. Surface runoff and leachate was collected and analyzed for the EDCs bisphenol A, 17alpha-ethynylestradiol, triclocarban, triclosan, octylphenol, and nonylphenol; a suite of sixteen metals; and estrogenic activity via the ER-CALUX bioassay. Triclocarban (2.3-17.3 ng/L), triclosan (<51-309 ng/L), and octylphenol (<4.9-203 ng/L) were commonly detected. Chromium (2.0-22 microg/L), cobalt (2.5-10 microg/L), nickel (28-235 microg/L), copper (14-110 microg/L), arsenic (1.2-2.7 microg/L), and selenium (0.29-12 microg/L) were quantifiable over background levels. Triclosan, nickel, and copper were detected at levels that might pose some risk to aquatic life, though levels of metals in the biosolids were well below maximum allowable regulatory limits. ER-CALUX results were mostly explained by background bisphenol A contamination and octylphenol in runoff, though unknown contributors and/or matrix effects were also found.

Antonious, G. F., S. O. Dennis, et al. (2011). "Ascorbic acid, beta-carotene, sugars, phenols, and heavy metals in sweet potatoes grown in soil fertilized with municipal sewage sludge." J Environ Sci Health B **46**(2): 112-121.

Municipal sewage sludge (MSS) used for land farming typically contains heavy metals that might impact crop quality and human health. A completely randomized experimental design with three treatments (six replicates each) was used to monitor the impact of mixing native soil with MSS or yard waste (YW) mixed with MSS (YW +MSS) on: i) sweet potato yield and quality; ii) concentration of seven heavy metals (Cd, Cr, Mo, Cu, Zn, Pb, and Ni) in sweet potato plant parts (edible roots, leaves, stem, and feeder roots); and iii) concentrations of ascorbic acid, total phenols, free sugars, and beta-carotene in sweet potato edible roots at harvest. Soil samples were collected and analyzed for total and extractable metals using two extraction procedures, concentrated nitric acid (to extract total metals from soil) as well as CaCl solution (to extract soluble metals in soil that are available to plants), respectively. Elemental analyses were performed using inductively coupled plasma mass spectrometry (ICP-MS). Overall, plant available metals were greater in soils amended with MSS compared to control plots. Concentration of Pb was greater in YW than MSS amendments. Total concentrations of Pb, Ni, and Cr were greater in plants grown in MSS+YW treatments compared to control plants. MSS+YW treatments increased sweet potato yield, ascorbic acid, soluble sugars, and phenols in edible roots by 53, 28, 27, and 48%, respectively compared to plants grown in native soil. B-carotene concentration (157.5 mug g(1) fresh weight) was greater in the roots of plants grown in MSS compared to roots of plants grown in MSS+YW treatments (99.9 mug g(1) fresh weight). Concentration of heavy metals in MSS-amended soil and in sweet potato roots were below their respective permissible limits.

Higgins, C. P., Z. J. Paesani, et al. (2011). "Persistence of triclocarban and triclosan in soils after land application of biosolids and bioaccumulation in Eisenia foetida." <u>Environ Toxicol Chem</u> **30**(3): 556-563.

The presence of the antimicrobial chemicals triclocarban (TCC) and triclosan (TCS) in municipal biosolids has raised concerns about the potential impacts of these chemicals on soil ecosystems following land application of municipal biosolids. The relative persistence of TCC and TCS in agricultural fields receiving yearly applications of biosolids at six different loading rates over a three-year period was investigated. Soil and biosolids samples were collected, extracted, and analyzed for TCC and TCS using liquid chromatography-tandem mass spectrometry. In addition, the potential for bioaccumulation of TCC and TCS from the biosolids-amended soils was assessed over 28 d in the earthworm Eisenia foetida. Standard 28-d bioaccumulation tests were conducted for three biosolids loading rates from two sites, representing agronomic and twice the agronomic rates of biosolids application plots as well as control plots receiving no applications of biosolids. Additional bioaccumulation kinetic data were collected for the soils receiving the high biosolids loadings to ensure attainment of quasi steady-state conditions. The results indicate that TCC is relatively more persistent in biosolids-amended soil than TCS. In addition, TCC bioaccumulated in E. foetida, reaching body burdens of 25 +/- 4 and 133 +/- 17 ng/g(ww) in worms exposed for 28 d to the two soils amended with biosolids at agronomic rates. The 28-d organic carbon and lipid-normalized biota soil accumulation factors (BSAFs) were calculated for TCC and ranged from 0.22 +/-0.12 to 0.71 ± 0.13 . These findings suggest that TCC bioaccumulation is somewhat consistent with the traditional hydrophobic organic contaminant (HOC) partitioning

paradigm. However, these data also suggest substantially reduced bioavailability of TCC in biosolids-amended soils compared with HOC partitioning theory.

Langdon, K. A., M. S. Warne, et al. (2010). "Aquatic hazard assessment for pharmaceuticals, personal care products, and endocrine-disrupting compounds from biosolids-amended land." Integr Environ Assess Manag **6**(4): 663-676.

Reuse of biosolids on agricultural land is a common practice. Following the application of biosolids to land, contaminants in the biosolids have the potential to migrate offsite via surface runoff and/or leaching and pose a hazard to aquatic ecosystems. The aim of this screening-level assessment study was to determine the relative hazard posed to aquatic ecosystems by pharmaceuticals, personal care products, and endocrine-disrupting compounds (EDCs) that have been detected and quantified in biosolids. This involved estimating maximum possible runoff water concentrations of compounds, using an equilibrium partitioning approach and then comparing these with the lowest available aquatic toxicity data, using the hazard quotient (HQ) approach. A total of 45 pharmaceuticals, personal care products, and EDCs have been detected in biosolids. Ten of these compounds (tonalide, galaxolide, 17beta-estradiol, 17alpha-ethinylestradiol, ciprofloxacin, doxycycline, norfloxacin, ofloxacin, triclosan, and triclocarban) posed a high (HQ >1.0) hazard to aquatic ecosystems relative to the other compounds. This hazard assessment indicated that further research into potential offsite migration and deleterious effects on aquatic ecosystems is warranted for the 10 organic contaminants identified, and possibly for chemicals with similar physicochemical and toxicological properties, in biosolids-amended soils. Because many antibiotic compounds (e.g., ciprofloxacin, norfloxacin, and ofloxacin) have ionic properties, the methods used may have overestimated their predicted aqueous concentrations and hazard. Further research that includes site-specific variables, e.g., dilution factors in waterways, rain intensity, slope of land, degradation, and the use of management strategies such as buffer zones, is likely to decrease the hazard posed by these high hazard compounds.

Xia, K., L. S. Hundal, et al. (2010). "Triclocarban, triclosan, polybrominated diphenyl ethers, and 4-nonylphenol in biosolids and in soil receiving 33-year biosolids application." <u>Environ</u> <u>Toxicol Chem</u> **29**(3): 597-605.

Land application of biosolids is a common practice throughout the world. However, concerns continue to be raised about the safety of this practice, because biosolids may contain trace levels of organic contaminants. The present study evaluated the levels of triclocarban (TCC), triclosan (TCS), 4-nonylphenol (4-NP), and polybrominated diphenyl ethers (PBDEs) in biosolids from 16 wastewater treatment plants and in soils from field plots receiving annual applications of biosolids for 33 years. All of the four contaminants evaluated were detected in most of the biosolids at concentrations ranging from hundreds of microg/kg to over 1,000 mg/kg (dry wt basis). They were detected at microg/kg levels in the biosolids-amended soil, but their concentrations decreased sharply with increasing soil depth for 4-NP, PBDEs, and TCC, indicating limited soil leaching of those compounds. However, potential leaching of TCS in the biosolids-amended soils was observed. The levels of all four compounds in the surface soil increased with increasing biosolids application rate. Compared with the estimated 33-year cumulative input to the soil during the 33-year consecutive biosolids application, most of the PBDEs and a small percentage of 4-NP, TCC, and TCS remained in the top 120-cm soil layer. These observations suggest slow degradation of PBDEs but rapid

transformation of 4-NP, TCC, and TCS in the biosolids-amended soils.

Wu, C., A. L. Spongberg, et al. (2010). "Uptake of pharmaceutical and personal care products by soybean plants from soils applied with biosolids and irrigated with contaminated water." <u>Environ</u> <u>Sci Technol</u> **44**(16): 6157-6161.

Many pharmaceuticals and personal care products (PPCPs) are commonly found in biosolids and effluents from wastewater treatment plants. Land application of these biosolids and the reclamation of treated wastewater can transfer those PPCPs into the terrestrial and aquatic environments, giving rise to potential accumulation in plants. In this work, a greenhouse experiment was used to study the uptake of three pharmaceuticals (carbamazepine, diphenhydramine, and fluoxetine) and two personal care products (triclosan and triclocarban) by an agriculturally important species, soybean (Glycine max (L.) Merr.). Two treatments simulating biosolids application and wastewater irrigation were investigated. After growing for 60 and 110 days, plant tissues and soils were analyzed for target compounds. Carbamazepine, triclosan, and triclocarban were found to be concentrated in root tissues and translocated into above ground parts including beans, whereas accumulation and translocation for diphenhydramine and fluoxetine was limited. The uptake of selected compounds differed by treatment, with biosolids application resulting in higher plant concentrations, likely due to higher loading. However, compounds introduced by irrigation appeared to be more available for uptake and translocation. Degradation is the main mechanism for the dissipation of selected compounds in biosolids applied soils, and the presence of soybean plants had no significant effect on sorption. Data from two different harvests suggest that the uptake from soil to root and translocation from root to leaf may be rate limited for triclosan and triclocarban and metabolism may occur within the plant for carbamazepine.

Zerzghi, H., J. P. Brooks, et al. (2010). "Influence of long-term land application of Class B biosolids on soil bacterial diversity." <u>J Appl Microbiol</u> **109**(2): 698-706.

AIM: To evaluate the effect of long-term annual land applications of Class B biosolids on soil bacterial diversity at University of Arizona Marana Agricultural Field Center, Tucson, Arizona. METHODS AND RESULTS: Following the final of 20 consecutive years of application of Class B biosolids in March 2005, followed by cotton growth from April to November 2005 surface soil samples (0-30 cm) were collected from control (unamended) and biosolid-amended plots. Total bacterial community DNA was extracted, amplified using 16S rRNA primers, cloned, and sequenced. All 16S rRNA sequences were identified by 16S rRNA sequence analysis and comparison to known sequences in GenBank (NCBI BlastN and Ribosomal Database Project II, RDP). Results showed that the number of known genera (identifiable > 96%) increased in the high rate biosolid plots compared to control plots. Biosolids-amended soils had a broad phylogenetic diversity comprising more than four major phyla: Proteobacteria (32%), Acidobacteria (21%), Actinobacteria (16%), Firmicutes (7%), and Bacteroidetes (6%) which were typical to bacterial diversity found in the unamended arid southwestern soils. CONCLUSION: Bacterial diversity was either enhanced or was not negatively impacted following 20 years of land application of Class B biosolids. SIGNIFICANCE AND IMPACT OF THE STUDY: This study illustrates that long-term land application of biosolids to arid southwestern desert soils has no deleterious effect on soil microbial diversity.

Kukier, U., R. L. Chaney, et al. (2010). "Phytoavailability of cadmium in long-term biosolidsamended soils." J Environ Qual **39**(2): 519-530.

Agronomic use of biosolids has raised concern that plant availability of biosolids-Cd will increase with time after cessation of biosolids application. It has been demonstrated that chemical extractability of Cd is persistently decreased in biosolids-amended soils. This study was conducted to determine if Cd phytoavailability in long-term biosolids-amended soils was also persistently decreased. Paired control and biosolids-amended soils were collected from three experimental sites where large cumulative rates of biosolids were applied about 20 yr ago. The pH of all soils [in 0.01 mol L(-1) Ca(NO(3))(2)] was adjusted to 6.5 ± 0.2 . Increasing rates of Cd-nitrate (from 0 to 10.0 mg Cd kg(-1) soil) enriched in (111)Cd stable isotope were added to all soils, and Romaine lettuce (Lactuca sativa L. var. longifolia Lam.) was grown in pots to bioassay phytoavailable Cd. After harvest, Cd concentrations in shoots and labile pool of Cd (Cd(L)) in soils were determined. The relationship between added salt-Cd and Cd concentrations in lettuce shoots was linear for all soils tested. Ratios of (shoot Cd):(soil Cd) slopes were highest in the control soils. Biosolids amendment decreased (shoot Cd):(soil Cd) slopes to varied extent depending on biosolids source, properties, and application rate. The decrease in slope in comparison to the control was an indication of the lower phytoavailability of Cd in biosolids-amended soils. A significant negative correlation existed between Cd uptake slopes and soil organic matter, free and amorphous Fe and Al oxides, Bray-P, and soil and plant Zn. Biosolids-Cd was highly labile (%L 80-95) except for Fulton County soil (%L = 61).

Ippolito, J. A., K. A. Barbarick, et al. (2010). "Infrequent composted biosolids applications affect semi-arid grassland soils and vegetation." <u>J Environ Manage</u> **91**(5): 1123-1130.

Monitoring of repeated composted biosolids applications is necessary for improving beneficial reuse program management strategies, because materials will likely be reapplied to the same site at a future point in time. A field trial evaluated a single and a repeated composted biosolids application in terms of long-term (13-14 years) and short-term (2-3 years) effects, respectively, on soil chemistry and plant community in a Colorado semi-arid grassland. Six composted biosolids rates (0, 2.5, 5, 10, 21, 30 Mg ha(-1)) were surface applied in a split-plot design study with treatment (increasing compost rates) as the main factor and co-application time (1991, or 1991 and 2002) as the split factor applications. Short- and long-term treatment effects were evident in 2004 and 2005 for soil 0-8 cm depth pH, EC, NO(3)-N, NH(4)-N, total N, and AB-DTPA soil Cd, Cu, Mo, Zn, P, and Ba. Soil organic matter increases were still evident 13 and 14 years following composted biosolids application. The repeated composted biosolids application increased soil NO(3)-N and NH(4)-N and decreased AB-DTPA extractable Ba as compared to the single composted biosolids application in 2004; differences between short- and long-term applications were less evident in 2005. Increasing biosolids rates resulted in increased native perennial grass cover in 2005. Plant tissue Cu, Mo, Zn, and P concentrations increased, while Ba content decreased depending on specific plant species and year. Overall, the lack of many significant negative effects suggests that short- or long-term composted biosolids application at the rates studied did not adversely affect this semi-arid grassland ecosystem.

Pepper, I. L., D. M. Quanrud, et al. (2010). Fate of Chemical and Biological (prions) Emerging Contaminants in Biosolids through Wastewater Reclamation Process and After Land Application. 83rd Annual Conference and Exhibition of the Arizona Water and Pollution Control Association (AWPCA 2010).

Abstract not available.

Pepper, I. and C. Gerba (2009). <u>Sustainability and pathogen hazards of land appliation of Class B</u> <u>liquid biosolids in the United States</u>. 2009 Conference on Sustainable Management of Water and Wastewater Sludges (Sludge 2009).

Abstract not available.