List of Resources and Documents for Joint Meeting of the Environmental Sustainability, Core Area Liquid Waste Committee, Solid Waste Advisory Committee, and Saanich Peninisula Wastewater Commission
May 25th, 2011

1. **Toronto Star 5 part series about the land application of biosolids in Canada (2008**). This is a very broad and useful examination of the history and many issues associated with the land application of biosolids, with a focus on Canada. http://tinyurl.com/3rcxgyb.

   Highlights
   p. 3 "Several major Food comoanies are not taking any chances. Del Monte, Campbell Soup and Gerber won't use food that has been fertilized with biosolids…Del Monte developed its no bio-solid policy in the early 80's, concerned that trace amounts of heavy metals and chemicals might find their way into the food chain. The other firms have had long-standing policies."
   p. 4 "Sweden, Switzerland, France and Holland are among countries that have either banned or introduced tougher standards on the use of biosolids as fertilizer. Instead, they are burning more of it in waste-to-energy plants."
   p. 7 "The Ecological Farmers Association of Ontario recently passed a resolution calling for a moratorium on spreading sludge and for more research on its long-term effects. The organization is concerned that harmful chemicals and pathogens in biosolids are contaminating the food chain."

2. **Review of State of Knowledge of Biosolids Science and Research: Contaminant Inventory**
   Report commissioned by the Canadian Council of Ministers of the Environment from 2009 by Hydromantis, Inc. in partnership with the University of Waterloo and Trent University.
   http://tinyurl.com/3kguwo9

   Highlights
   p. IV-VII is a list of chemicals identified in sludge, both pre and post processing.
   p. VII "From the review it appears that certain micro-constituents can be reduced by anaerobic digestion, while others are recalcitrant, or perhaps even increased in concentration, by anaerobic biotransformation processes."
   p. 1 "At present, there is a notable lack of sound science regarding the significance of a certain class of micro-constituents termed emerging contaminants (ECs), which include an array of pharmaceuticals, personal care products and industrial contaminants (such as plasticizers, surfactants and brominated flame retardants). While there is some documentation of ECs in biosolids, no focused study has been completed yet on an inventory of ECs in Canadian biosolids. Consequently, the CCME has issued a Request for Proposals

to document the occurrence of ECs in biosolids and to conduct a targeted sampling program which will provide a basis for the CCME to evaluate and manage the risks associated with ECs in biosolids with respect to managed land application, land reclamation, production of commercial soil amendments and energy production."

3. **Case for Caution Revisited: Health and Environmental Impacts of Application of Sewage Sludges to Agricultural Land**

   This paper is a recent literature review (March 2009) of the state of knowledge in regards to potential harms associated with the land application of biosolids written by Ellen Z. Harrison PhD, retired Director, and Murray McBride PhD, Director, Cornell Waste Management Institute, Dept. of Crop and Soil Sciences, Rice Hall, Cornell University, Ithaca, NY. It includes summaries of recent peer-reviewed research findings.
   http://cwmi.css.cornell.edu/case.pdf

   Highlights

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4. **A Critical Review of the U.S. EPA's Risk Assessment For the Land Application of Sewage Sludge**

   The following is a very recent research paper by J.M.J Mathney (MA, MPH)[1]

[1] **Current Employment:** Senior research associate at First Light Biosciences
Past: Research Assistant at The Forsyth Institute**;** Intern, Office of Research and Standards at MA Department of Environmental Protection**;** Research Assistant at SelectX
Education: Boston University MPH, Environmental Health **2**007 – 2010 **Relevant Coursework:** Introduction to Toxicology, Intermediate Toxicology, Epidemiologic Methods, Environmental Epidemiology, Risk Assessment Methods, Wastewater and Health, Biostatistics. **Columbia University in the City of New York,** MA, Biotechnology**,** 2003 – 2004**, Thesis:** Current research in prion diseases: Understanding the molecular biology of PrP proteins

explaining why the current EPA regulations governing the land application of biosolids on which Canada's own regulations are based (as indicated in the Stantec Report) are inadequate to protect public health and the environment. http://www.sludgefacts.org/Ref115.pdf

Highlights:
P. 43 Abstract: "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."

5. **Questions, Answers and Facts About Biosolids in Nova Scotia**
FAQs sheet about biosolids produced by the Nova Scotia Environmental Network's Biosolids and Waste Water Caucus (www.nsen.ca).
http://www.nsen.ca/documents/NSEN%20Biosolids%20FAQ'S.pdf

Highlights
Is land application of bio-solids safe?
No. The following adverse effects have been noted on lands following treated biosolids application: Loss of soil fertility (excessive loading of soils with heavy metals - copper, zinc, molybdenum - destroy good soil microorganisms) Contamination of ground water sources with pollutants (copper, lead, zinc, and pesticides via facilitated transport onto organic molecules).
Regrowth of pathogens in biosolids after mixing with soil (potential for food-borne illnesses – salmonella, coliform bacteria, viruses, prions).
Contamination of soils with persistant and bio-accumulative toxins, such as heavy metals (lead, cadmium - breast cancer), persistent and volatile organic pollutants (such as flame retardants - carcinogenic), hormones/steroids (affect human and wildlife reproductive function), any thousands of other chemicals Uptake and storage of contaminants by some plants/forages.
Livestock illness and death (nutrient imbalances, direct ingestion of biosolid contaminants/pathogens in forages) Risk of liability to end users (farmers) regarding complaints of human or animal illness, environmental contamination , and potential loss of property value.
Is it safe to eat foods grown or raised on soils where Biosolids are used? No. It has been well documented that livestock ingesting biosolid treated fields can store

contaminants in body fat and milk glands (thallium and flame retardants are both lipophillic (fat loving) and carcinogenic. Some plants, such as green leafy vegetables take up and store heavy metals (lead, molybdenum, etc). Incidences of cancer, respiratory diseases and food –borne illnesses are on the rise and we must consider that land application of toxic sludge could be a major contributor.

6. Additionally, I have attached letters by the Sierra Club of BC, the Island Organic Producers Association, and the Dogwood Initiative all supporting the CALW ban on the land application of biosolids, and opposing the Saanich Peninsula Waste Commission's pilot-project for large scale application of biosolids to farmland.

Toronto Star Sludge Series: Soiled Land

July 12 - 15, 2008

Carola Vyhnak
Urban Affairs Reporter

Part I: Is sewage fertilizer safe?

July 12, 2008

Is sewage fertilizer safe?

Worries grow over 'stew' of chemicals spread on farmland

Feces, urine, vomit, blood. Synthetic hormones, heart pills, antibiotics, illicit drugs, Viagra. Bacteria, viruses, E. coli, parasites. Household cleaners, shampoo, solvents, pesticides and traces of arsenic, mercury, cadmium, lead, dioxins and flame retardants.

Each day, this chemical cocktail is piped from our homes, businesses and industries to sewage plants across the province. The water is filtered and reclaimed.

The solid waste that remains is turned into biosolids, more commonly called sludge. For more than 30 years, Ontario's sludge has been trucked out to farmland for use as fertilizer.

Then in 1996, the province, which monitors sludge dispersal, increased promotion of the nutrient-rich goo to farmers as a beneficial alternative to chemical fertilizers. Officials insist sludge is tested and safe and that there are no documented cases of adverse health effects when requirements are followed.

But some rural residents who live near properties where sludge has been used have argued for years that what ends up on fields isn't benign fertilizer, but a "toxic stew" that's harming them and the environment.

"It takes the air out of your lungs and burns your eyes. It's nasty, nasty stuff," said Crystal Chordis, a resident of Corbetton north of Orangeville.

Country-dwellers exposed to sludge complain of a litany of ailments including respiratory problems, diarrhea, headaches, nausea, rashes, fatigue and pneumonia.

Ontario's acting chief medical officer of health, Dr. David Williams, says a clear link to adverse health effects hasn't been established. He is satisfied that the practice of using biosolids on farmers' fields is safe and says the process of monitoring possible health issues is "active and ongoing."

Just what is making people ill is difficult to pin down but two things are apparent.

Firstly, what is making its way into our sewage system has changed with new drugs and chemicals raising questions as to whether the testing and tests are keeping pace.

Secondly, local officials who investigate health complaints are not required to report their findings to the province.

And while experts on both sides of the issue are mostly at odds, they agree on the first point: There is still a lot to learn about sludge.

"A complete analytical characterization of sludge's pathogen, endotoxin and chemical contaminant composition has never been attempted," says researcher Dr. Rob Hale of the Virginia Institute of Marine Science. Forty per cent of the sludge produced by Ontario's municipal sewage plants – 120,000 dry tonnes each year spread on 15,000 hectares – is put on soil where crops are grown.
The bulk of it, which is given to farmers free, ranges in consistency from a thick liquid to a drier cakelike form.

A plant in Windsor turns a small amount of biosolids into dried pellets for which farmers pay about $19 a tonne. (What's not spread on farmland is burned or sent to landfills.)

Eighty per cent of Ontario's municipalities spread sludge on agricultural land. Last year, 13 per cent of Toronto's sludge was put on farmers' fields.

What to do with municipal sludge is nothing new and for years it was incinerated, sent to landfills or simply dumped into the nearest Great Lake.

Diverting some of it to fields got its start in the 1970s but went into high gear in 1996 after the Great Lakes Water Quality Agreement stiffened sewage treatment guidelines and in turn created more sludge.

Since then, the province has pushed so-called land application as a safe option for municipalities struggling to deal with fast-filling landfills and a U.S. border that is slowly closing to Ontario's waste.

But the provincial regulations governing testing and application were last updated in 1998 and now a whole new range of chemical compounds is turning up in our sewer systems. Many of these, such as pharmaceuticals and personal care products, simply aren't tested for because there are few labs that can do that kind of analysis, no accepted methodology, and no benchmarks to say what's safe.

Yet, the lack of epidemiological studies means it cannot be determined whether sludge is making people sick. That has prompted a call for more research from Toronto toxicologist Dr. Anne Mildon.

"It's cause and effect," says Mildon, who treats several patients who believe sludge made them ill. "I'm too good a scientist to say, `Yes, this is definitely it,' but it's very likely."

Several major food companies are not taking any chances. Del Monte, Campbell Soup and Gerber won't use food that has been fertilized with biosolids. Not enough is known about biosolids, they say.

Del Monte developed its no-biosolids policy in the early '80s, concerned that trace amounts of heavy metals and chemicals might find their way into the food chain. The other firms have also had long-standing policies.

Mildon, who led a provincial task force on radioactive waste in Port Hope during the 1990s, says the provincial government has been in a "state of denial" and has failed miserably to address public health concerns about sludge.

Her concerns are echoed here and around the world.

In parts of the United States, several deaths have been linked to sludge exposure. In Ontario, several citizens' group including those in Prince Edward County and near Orangeville have succeeded in halting or restricting sludging.

Sweden, Switzerland, France and Holland are among the countries that have either banned or introduced tougher standards on the use of biosolids as fertilizer. Instead, they are burning more of it in energy-from-waste plants.

Since 2002, Ellen Harrison, recently retired director of the Waste Management Institute, a research and training branch of Cornell University in Ithaca, N.Y., has argued for a ban on sludge application. She expresses frustration over the paucity of health studies.

One of the few is a recently published report by researchers from the University of Toledo in Ohio, which found a significant increase in problems such as abdominal bloating, jaundice and weight loss among residents exposed to treated fields.

The 2005 study surveyed 613 people over one month and researchers also noted an increased risk for respiratory, gastrointestinal and some chronic diseases such as multiple sclerosis. Four hundred and thirty-seven of the people surveyed lived within 1.6 kilometres of fields treated with biosolids, 176 lived further away.

In 2002, under pressure from concerned residents, the City of Ottawa commissioned a review on the health and safety of spreading biosolids.

Struck by the lack of medical information, the consultants concluded that a "surveillance system for monitoring health effects from biosolids does not appear to exist in any jurisdiction.

"While anecdotal cases are occasionally reported by the news media, few of these are investigated by trained teams of agronomists, engineers, toxicologists, microbiologists or public health professionals, let alone make their way into peer-reviewed research literature," the final report read.

After a two-year moratorium, sludge-spreading resumed in Ottawa.
Today, provincial officials do not know how many health complaints have been reported or how many investigations have been done in Ontario.

(Complicating the issue is people who experience illnesses they believe are related to sludge often are afraid to report anything because it would mean blowing the whistle on neighbours they value as friends and helping hands.)

Here's how the complaint system works in Ontario: Anyone with a health complaint they believe is related to biosolid-spreading should report it to their local health unit. The local medical officer of health investigates the complaint to determine whether a health hazard exists.

He or she notifies and consults with the environment ministry, which assesses if the sludge was applied according to provincially set regulations and standards. The medical officer of health also consults with the agriculture ministry. If provincial guidelines were violated or a health hazard exists the environment ministry can order the problem fixed and may lay charges.
After investigating, the health officer sends a written report to the complainant but there is no requirement to send the report to anyone at the provincial level.

Communication is at the discretion of the local health units, said David Jensen, a spokesperson for the health ministry. He added that Williams, the acting chief medical health officer, is required by law to keep himself informed "on matters related to occupational and environmental health."

Williams said he expects local medical officers of health to keep him in the loop but "there is no requirement by law to tell me everything they're doing."

Cornell's Harrison finds it "appalling" that Ontario does not catalogue complaints or do a thorough and immediate investigation. When a sludge-related health issue is suspected, she says, an investigation should be launched at the source, as it would be in an outbreak of food-borne disease.

Harrison had some advice for politicians: "If it is possible to err on the side of caution, do it: Put in place a system for complaints investigation, and (don't) continue with a `head in the sand' approach that everything is all right."

Where your waste goes

How yesterday's meal ends up on a farmer's field

The human body excretes about 1.5 litres of urine and 150 grams of feces every day. It and everything else dumped in the sewer system ends up in a sewage treatment plant where it's turned into clean water and solid waste known as biosolids.

Five hours after a toilet flushes in north Whitby, the contents arrive at the Corbett Creek Water Pollution Control Plant near the shoreline of Lake Ontario. Corbett Creek, one of 11 waste water treatment facilities in Durham Region, handles 65 million litres a day.

The first thing you notice in the arriving muck is that Whitbyites eat a lot of corn. They also practise safe sex.

But all evidence has disappeared by the time the final product emerges at the other end looking like thick black paint.

It's a 15-day process that starts with screening the raw waste water to remove large objects such as sticks and cans. From there it goes to a tank where sand, gravel and other heavy material settles to the bottom.

In the next stage, the waste water is kept in a tank for several hours so the solid particles can sink.

The water moves on to an aeration tank which plant manager Thom Sloley likens to a giant feed lot. Instead of cattle, microbes or "bugs" feed on the material.

"They eat it, get big and fat and happy, and then we sink them out – that's the biosolids."

The liquids are sent on for chlorination before being piped out to the lake while the sludge is moved to a digester, which is like a big cooking vat.

The process to this point takes eight to 10 hours.

The sludge then spends the next two weeks in the digester where it's heated to 37C to break down into organic molecules and kill off pathogenic bacteria. The methane that's produced is burned off and the sludge goes to storage tanks, incinerators or farmland for spreading.

Sewage material is sampled coming into and leaving the plant and also on a daily basis to ensure it is safe, explains Sloley.

"We want those biosolids to be clean. If they're not, you have a problem."

Treatment plants are heavily regulated by the environment ministry because "we're in the business of protecting public and animal health, and the environment," says Sloley, adding Durham Region's biosolids program is the best in the province.

About one-third of the region's sludge is used as fertilizer on agricultural land. It's tested for E. coli, heavy metals, potassium, nitrogen and phosphorous. About 99 per cent of the pathogens are destroyed and those that survive die off in the field, he says.

"We do everything we can to make sure it's safe based on the information we have so far. Do we catch everything that could happen? That's not possible. But (sludge) has to meet the guidelines or it can't go on the fields."

Part II: Farmers split over safety

July 13, 2008

Farmers split over safety

Free biosolids tempting at a time when prices of commercial fertilizer are skyrocketing

The price is right. With savings of more than $100 an acre for fertilizer, the offer of free stuff is tempting for farmers struggling to make a living in the face of rising costs and diminishing returns.

Harry Buurma wishes he could get enough sludge to cover his entire 3,000-acre farm in Watford, partway between London and Sarnia. As it is, he supplements commercial fertilizer with biosolids to feed his cash crops. Sludging cornfields, for example, saves him $75 on fertilizing costs of $150 per acre, he says.

"Fertilizer prices have doubled and tripled so biosolids are a better deal than ever," says Buurma, who uses both free liquid sludge and dried sludge pellets that cost him $19 a tonne.

The pellets have less odour and a reduced pathogen content, and there are fewer restrictions on when and how they can be applied, he says.

The beauty of biosolids, apart from the price, is their nutrients and organic matter, says Buurma, who believes they are as safe as pig or cow manure. "The soil and plants process this stuff and render everything harmless. We're not eating it directly."

He scoffs at statements that contaminants from industrial waste find their way into biosolids.

"That's a total misconception. Industrial waste doesn't go into city sewers. Twenty or 30 years ago, yes, but the level of heavy metals – except for copper – has been reduced 90 per cent. We have a really tight system now."

The "real crime," he says, is that not all biosolids are spread on farmland.

"Taking the stuff to landfill is wrong, really wrong," especially when the ingredients in fertilizer will run out in 50 years, Buurma says.

But while biosolids are fine for his crops, he is not so sure about vegetables. He thinks it could be bad for business. "If I had a vegetable stand, I wouldn't be selling it because of the image."

People recoil at the thought of eating food grown in their own feces, regardless of how it might have been treated. That perception helps fuel the division within the agricultural community on the merits of sludge. While some farmers like Buurma can't get enough of the stuff, others are reluctant because of public opposition and concerns about safety and liability.

The Ecological Farmers Association of Ontario recently passed a resolution calling for a moratorium on spreading sludge and for more research to determine its long-term effects. The organization is concerned that harmful chemicals and pathogens in biosolids are contaminating the food chain.

While a number of farmers believe sludge has made their livestock ill, the evidence is largely anecdotal. But some argue the uncertainty is enough to raise questions about its use.

Fred Price used biosolids on his farm near Hanover more than 10 years ago. After feeding his 100 head of beef cattle hay grown on sludge-treated land, only half the usual number of cows became pregnant, he says.

"You couldn't find anything wrong with them but once we quit spreading sludge and once they went onto pasture in the spring, they were breeding again."

Price suspects the cause was hormones found in birth control drugs finding their way into the land through the sludge. He says three other farmers experienced similar problems.

His farm is now sludge-free. "I sure don't want it any more. I'm always looking for cheaper ways to get fertilizer but it ended up costing a lot."

Donald Good is an Ottawa lawyer who has spent years warning farmers about the inherent risk of using material "contaminated with human diseases."

"Never adopt a practice that undermines the confidence of consumers in the safety of food you produce," he advises farmers. "The application of sewage sludge to farmland does just that."

Ill When wells are contaminated

They started to become ill when a farmer spread sludge on his fields, then their wells became contaminated

There's a note of quiet desperation in Wendy Deavitt's voice.

"I'm sitting here being continually poisoned. I am scared to death."

It started in the summer of 2006 when a farmer near her home in Trent Hills, 90 minutes northeast of Toronto, spread sewage sludge on his fields. She and her family immediately developed diarrhea, headaches, coughs and hoarseness. Since then she has been plagued with fatigue, nausea, cramps, malfunctioning kidneys and bowels and elevated levels of lead, barium and potassium.

"I've never been this unhealthy in my life. My bowels are shutting down at 47 years old."

Deavitt and her husband, William, are one of four couples in the area who are being treated by a Toronto toxicologist for everything from chronic diarrhea to pneumonia.

Dianne and Wayne Cooke have spent $6,000 on weekly infusions of vitamins and minerals to help rid their bodies of toxins. Their immune systems have been compromised since the wind first brought contaminants to their hilltop home in the fall of 2005.

Linda Donaldson and her husband, Roger, sold the farm that had been in her family for 150 years and moved to Campbellford to get away from sludge – only to find it being spread there, too.

"It has just totally devastated us," says Linda, a retired nurse practitioner. When they became ill after the sludging, she drove to every house in a large circle around the site and discovered 22 people were sick with the same symptoms.

Since then, families' wells have become contaminated and people have been told not to drink the water. They blame it all on biosolids.

"I remember standing on the road watching it spray 25, 30 feet into the air," says Deavitt. "My eyes were burning from the smell. It makes you want to vomit."

Her six horses, donkey, cats and dog all became ill with diarrhea, swollen lymph nodes and other ailments. She has sent some to live elsewhere for the sake of their health.

In the fall of 2006, the couples organized a letter- and email-writing campaign to local health authorities and government officials about their medical problems. Last December, the medical officer of health for the area, Dr. Lynn Noseworthy, sent them letters saying she had investigated but failed to find a "causal relationship" between their ill health and biosolids.

Noseworthy said her investigation of the cases included reviews of literature, correspondence and medical information as well as consultations with the environment ministry and health ministry. While she concluded sludge didn't cause the illnesses, she didn't determine what did.

The group, however, isn't satisfied. The attitude at Queen's Park is "we're making all this up," says Deavitt, who worked in the medical field for 14 years.

"There's so much proof here – what are they waiting for, a body count?"

Oakville family files suit over treated sewage lagoon near their home

Don't try to tell Laurie Eagles sludge is safe.

Twelve years after prolonged exposure to a pit of human waste, she and her family of four are still suffering from bouts of pneumonia, bowel disease and respiratory disorders.

They were living in rural Oakville in 1996 when a lagoon for Toronto's treated sewage opened less than a kilometre from their home.

The lagoon, which was the size of two football fields, was supposed to be a temporary storage facility at the W.A. Bill Johnson Biosolids Management Centre but "it got bigger and bigger and fuller and fuller," recalls Eagles.

"It was an open pit. The stink would bring you to your knees – it burned your nose, it was horrendous."

"I didn't know what was in the stuff but the more I learned, the more scared I got. At one point, I was calling (authorities) five and six times a day, the smell was so bad."

In the summer of 2000, their well tested positive for E. coli, Eagles says, and her husband, Allan, spent 10 days in hospital with Crohn's disease.

When the pit was finally closed in 2002 after years of fighting with the regional and provincial governments, their symptoms suddenly stopped, they say.

"I am absolutely convinced it was the cause of our health problems. It was as if we were living beside a field that was being sludged every day with no let-up."

The family has filed a $2 million lawsuit against Halton Region and American Water, the parent company of Azurix North America (Canada) Inc., which transported the sludge to the site.

The lawsuit has yet to be heard in court.

The Halton health department investigated complaints by the Eagles and other families who lived near the lagoon but according to medical officer of health Dr. Bob Nosal, no conclusions could be drawn because of the small number of people involved and their varied responses. (Of 14 surveyed, four linked their health problems to the facility.)

However, subsequent reports by the health department warned of potential risks to soil, water, food and human health if sludge is not stored and handled properly.

Nosal told the Star he could not discuss the Eagles' case because of the lawsuit.

Part III: When sludge disposal rules are broken

July 14, 2008

When sludge disposal rules are broken

Trucking tonnes of smelly black goo out to the country and spreading it on farmland is safe, says the environment ministry – as long as rules are obeyed and guidelines followed.

Still, things can go horribly wrong, as residents of Cedarville, Ont. found out on a hot, muggy August day when a load of sewage sludge was dumped on fields in their tiny hamlet in Southgate Township, about 40 kilometres northwest of Orangeville.

"The stench was like nothing you'd experienced in this life," recalls resident Glenn Norman. "We were literally swarmed with flies as we stood outside discussing it." Their eyes were red and swollen for hours after and several residents were forced out of the area for the day by the odour.

"How dare they say it's safe? It's not," Norman fumes eight years later.

Terratec Environmental Ltd., the hauler, later pleaded guilty to odour violations and was fined $12,000.

Eileen Smith of the Ministry of the Environment insists biosolids guidelines and established procedures do serve to safeguard human health and the environment.

Yes, rules are sometimes broken "but it's not a big problem," says Smith, policy and special projects manager for the environment ministry's waste management policy branch.

Similar to the Cedarville incident, a small rural neighbourhood in Oakville felt it when things went awry at a sludge storage facility, including several spills, a torn tank liner and an open pit of Toronto's muck that was overfilled.

In reports following investigations into odour, health and contamination complaints, Halton Region's medical officer of health called for more vigilant monitoring and warned of the need to properly store and handle biosolids because of potential risks to human health, soil, water and food.

Sludge is the solid waste left over at the end of the sewage treatment process, when clean water is removed from the soup of human, commercial, hospital and industrial waste that comes down the pipe.

Each year, close to half of Ontario's nutrient-rich sludge is taken by farmers and spread over 15,000 hectares of land. Municipalities have for years struggled with what to do with sludge and so it has been incinerated, sent to landfills or simply dumped into the nearest lake.

Diverting some of it to fields began in the 1970s. Then in 1996, the Great Lakes Water Quality Agreement stiffened sewage treatment guidelines. This created more sludge and Ontario started recommending it for use as fertilizer for farm crops. Faced with fast-filling landfills and a U.S. border slowly closing to Ontario's waste, many municipalities accepted.

But today – 10 years after the regulations governing testing and application were updated – a whole new range of chemical compounds is turning up in our sewer systems. And local officials who investigate health complaints are not required to report their findings to the province.

Terratec, the province's biggest sludge hauler, and two affiliated companies have racked up more than 40 convictions for violating environmental laws between 2001 and 2007, according to documents provided by sludge watchdog Maureen Reilly and acquired from the province.

Fines levied against Terratec, Azurix North America (Canada) Inc., which no longer exists, and their parent company, wastewater giant American Water Services Canada Corp., totalled more than $300,000.

Several charges against Terratec are still before the courts. The firm, which has been in the biosolids business more than 25 years, handles about half of the land-applied sludge in the Golden Horseshoe area, servicing more than 300 farmers and spreading waste from urban centres on 4,000 to 6,000 hectares a year.

President Phil Sidhwa admits there have been a "small number of cases" where guidelines weren't followed, but maintains the majority of his company's convictions had to do with maintenance and operations. Those that were related to biosolids were mostly beyond their control such as odour issues, which he said originate at the treatment plant.

"We run a safe operation and use best management practices to protect the environment and people's health," he says.

But the ministry has taken action against Terratec, says spokesperson John Steele, citing fines, frequent inspections and increased setback distances.

The province does about 200 inspections a year – at less than half of the 400 to 500 sites that are treated with biosolids annually. The MOE also handles about 100 complaints a year, usually to do with odour or application conditions. Typically, five to 10 convictions result each year, says Steele.

In a move that Eileen Smith says will raise safety, odour and application standards, the government is introducing changes that will drop the requirement for a certificate of approval for sludge spreading and allow it to be handled by farmers as part of the Nutrient Management Act. And biosolids will be referred to as "non-agricultural source materials."

With the proposed changes, which Smith says won't take effect for more than a year, sludge will be the joint responsibility of the Ministry of Agriculture, Food and Rural Affairs and the environment ministry, which will continue to handle compliance and enforcement.
"The changes we're proposing are designed to further protect human health and the environment while enhancing productivity of the soil," she explains.

But sludge opponents see the move as a step backwards that will wrap the biosolids program in bureaucracy and secrecy.

Certificates are "transparent, publicly available documents" that tell people what's being spread, where and in what amounts, notes Reilly. Eliminating them means that information will be difficult to get from the agriculture ministry, she says.

Federal workplace safety agency recognizes sewage sludge disease

People who contend that sludge contains hazardous chemicals and has intact biological components appear to have an ally in the federal government.

More than 10 years ago, "sewage sludge disease" was recognized as a potentially fatal illness.

One of a group of diseases known as extrinsic allergic alveolitis, it is triggered by intense or prolonged exposure to animal and vegetable dust, according to the federally funded Canadian Centre for Occupational Health and Safety.

The centre's website says that sewage sludge disease is triggered by the body's natural defensive reactions and is associated with sewage waste that has been heat-treated.

An acute attack would include symptoms such as: fever, muscular aches and a general, unwell feeling. These symptoms are accompanied by tightness in the chest, a dry cough, and shortness of breath.

A lower level of exposure is marked by coughing, shortness of breath, sweating, sore throat, headache, and nausea.

The disease can become chronic with both types of exposure. And chronic cases can lead to permanent lung damage and even death.

Prevention includes dust control, protective clothing and good ventilation.

Part IV: Food firms shun sludge use

July 15, 2008

Food firms shun sludge use

The corn flakes you had for breakfast this morning could have been made from corn nourished by sewage sludge. The milk in the bowl could have come from a cow raised on sludge-fertilized feed.

Is this safe? The provincial government says yes, and claims to have science on its side. But several food companies, including Del Monte, Campbell Soup and Gerber baby foods, aren't so sure and have decided to err on the side of caution.

Del Monte developed a no-biosolids policy in the early 1980s, over concerns that trace amounts of heavy metals and chemicals might find their way into the food chain.

"Our main concern is upholding the high standard of healthy, nutritious foods that our consumers trust and expect," says Mary Sestric, a spokesperson for the company in Pittsburgh.

Similarly, the soup maker won't use produce grown on land treated with biosolids because of the "potential toxicological food safety risk posed by concentrated heavy metals in the soil," explains Anne Yourt, brand communications manager for Campbell Company of Canada.

"These concentrated heavy metals may be absorbed by the plants at levels that would be unacceptable for human consumption."

Nestle, the world's biggest food and drink company, also shies away from sludge-fertilized crops in its Gerber baby products, says a spokesperson for Nestle Canada.

Sludge is the solid waste left over at the end of the sewage treatment process. Every year nearly half of Ontario's municipal sludge is spread on 15,000 hectares of land.

The practice existed in the 1970s but began in earnest in 1996 when water quality agreements stiffened sewage treatment guidelines, thus creating more sludge. But the provincial regulations governing testing and application were last updated in 1998 and today a new range of chemical compounds is turning up in our sewer systems.

Research has shown that grains, potatoes and leafy vegetables like lettuce, spinach and swiss chard readily absorb cadmium, one of the heavy metals that may find its way into sewage, according to soil scientist Murray McBride, a specialist in heavy metals. Cadmium has been linked to loss of kidney function.

More than 30 years ago, University of Toronto researchers discovered mercury contamination of food and forage crops grown in sludged soil. Unwashed samples of tomatoes, beans, carrots, lettuce, wild barley and quack grass had higher than permissible levels of mercury, they said in a study funded by the environment ministry.

Well-scrubbed tomatoes and beans showed higher levels than samples grown in unsludged soil. For tomatoes, levels were up to 50 times higher than permissible, said the peer-reviewed report, published in 1974. Mercury, which can be particularly hazardous to pregnant women and young children, interferes with the brain and nervous system.

It's difficult to identify what produce is grown in sludge-treated soil and where. But growers in Holland Marsh, the source of about half of Ontario's carrots and onions and most of Toronto's locally produced Asian greens, don't use biosolids because the soil there is highly organic and not suitable.

"There is no sewage sludge in Holland Marsh that I'm aware of," says Art Janse, who was in charge of the area's drainage system for 36 years. "There'd be no benefit."

However, in agricultural areas where livestock graze directly on sludged pasture or eat crops grown with it, there are concerns about pollutants that accumulate in milk and meat.

McBride, director of Cornell Waste Management Institute in Ithaca, N.Y., worries about effects on dairy farms. He cites a U.K. study that showed sheep reared on sludged pastures produced smaller, hormonally affected offspring.

Bill Mitchell, spokesperson for the Dairy Farmers of Ontario, says consumers have nothing to worry about because of the rigorous testing milk undergoes. "No food that's consumed by the public is more tested than milk," he says.

Every load is tested and inspectors visit all dairy farms a minimum of once every two years to ensure quality and safety standards are met, Mitchell says, listing tests for "things you'd expect like bacteria, protein, butter fat and antibiotics" – though not such contaminants as PBDE, a flame retardant that shows up in sludge and has been found in human mother's milk.

The marquee case for sludge opponents came last February, when a federal judge ordered the U.S. Department of Agriculture to compensate a Georgia farmer whose land was poisoned in the 1980s and 1990s, and whose cows – fed on crops from those fields – died by the hundreds. Tests found their organs contained toxic levels of copper and zinc, as well as high levels of cadmium, lead and other pollutants. Sick cows recovered when fed crops from non-sludged fields.

The judge was highly critical of the U.S. Environmental Protection Agency for ignoring the farmer's concerns, for endorsing unreliable and sometimes "fudged" data and for trying to quash questioning of the EPA's biosolids program.

While sludge standards and spreading practices differ in Ontario, the province relies on testing and monitoring in other countries to set its standards, even as attitudes in Europe, at least, have shifted, including some countries banning the agricultural use of sludge.

Keeping sewage off farm fields a burning issue

Incinerating sludge, sometimes to generate power, among the alternatives to putting it on farmland

If we don't spread it, then what?

The flow of sewage and the need to do something with it is a daily dilemma. "You can't stop it," says Abdul Khan, Hamilton's director of water and wastewater treatment.

As sludge loads increase and controversy grows over the safety of spreading it on farmland, the search is on for other disposal options.

Hamilton is following the lead of Peel and York regions by moving toward burning processed human and industrial waste, after deciding land application isn't sustainable.

The city's Biosolids Master Plan, which predicts that population growth will see sludge production increase by 80 per cent over 30 years, cited a lack of available land, fewer farmers willing to take sludge due to odour and contaminant concerns, and more complex rules.

York sends sludge to Durham Region for incineration. Peel burns its sludge in two incinerators and is poised to build two more, making its Lakeview plant the largest sludge incineration facility in North America. Sludge burned to ash is sometimes used in construction materials or sent to landfills.

Peel's biosolids management review six years ago looked at pelletization, land application and composting, says Mark Schiller, water division director. Incineration, the review deemed, "was still the best way to go" in terms of environmental and cost concerns.

Toronto currently turns only a small amount of its sludge into fertilizer pellets but has plans to increase that to about half.

In Hamilton, Jim Harnum, senior director of water and wastewater, has expressed concern about toxic metals, pathogens and pharmaceutical residue in municipal sludge as a threat to land application. Hamilton sends 1,250 truckloads of sludge a year to farm fields.

The city is conducting an environmental assessment of its proposal to build an incinerator like Peel's. Other options on the table include California-based Liberty Energy's aim to generate electricity by burning sewage sludge and woody waste.

Biomass gasification involves heating organic material at low temperatures to release gases, called syn-gas or biogas, which can be burned to drive turbines and generators that make electricity.

Environmentalists favour using sludge as fuel. In Ottawa, where city council last month approved a plan by PlascoEnergy Group to use ultra-high-temperature plasma torch technology to dispose of garbage in a waste-to-energy system, anti-sludge residents hope the process can be applied to biosolids.

"Is it not possible to combine the sewage with the city garbage and still get sufficient syn-gas to generate some electricity?" Jim Poushinsky, chair of Ottawa Citizens Against Pollution by Sewage, asks in a letter to PlascoEnergy.

The company's plant, which is awaiting provincial approvals, will be the first of its kind in the world. The firm has told Poushinsky that, while the process could be modified to include sludge, for now they are concentrating on solid waste.

Meanwhile another plasma torch company, Fabgroups Technologies Inc., is working in Quebec on a system to process wet biosolids by heating them to the point where they oxidize.

Farther afield, a British scientist is developing a process that would use "gut bugs" – bacteria from the human lower intestine – to turn raw sewage into energy.

In an online newsletter, Prof. David Stuckey describes a bioreactor he'll build alongside a sewage treatment plant to break down effluent, create energy and reduce sludge by 90 per cent. His technology uses membranes to filter wastewater for potential reuse as well as bacteria that don't require oxygen to break down waste.

Stuckey recently got a $500,000 grant from an independent U.K. academy to develop his system.

"Imagine a day when mini bioreactors, located under apartment buildings, are able to convert raw sewerage from flats into valuable methane gas for use in household heating, and treated water recycled back to flush toilets," Stuckey says.

REVIEW OF STATE OF KNOWLEDGE OF BIOSOLIDS SCIENCE AND RESEARCH: CONTAMINANT INVENTORY

Draft Report – Literature Review
CCME Project # 447-2009

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ACRONYMS

ADBI	polycyclic musk fragrance, trade name Celestolide
AHMI	(or AHDI) polycyclic musk fragrance, trade name Phantolide
AHTN	polycyclic musk fragrance, trade name Tonalide
AMX	derivative nitro musk fragrance, amino Musk xylene
AP	alkylphenol
APE	alkylphenol ethoxylate
ATII	polycyclic musk fragrance, trade name Traesolide
BEHP	bis(2-ethylhexyl) phthalate
BLS	fluorescent whitening agent
BPA	bisphenol A
BFR	brominated flame retardants
DAF	dissolved air flotation
DAS 1	fluorescent whitening agent
DDE	dichlorodiphenyldichloroethylene
DDT	dichlorodiphenyltrichloroethane
DPE	diphenyl ether
DPMI	polycyclic musk fragrance, trade name Cashmeran
DSBP	fluorescent whitening agent
EE2	17α-ethinylestradiol
E1	estrone
E2	17β-estradiol
E3	estriol
EO	ethoxylate
HBCD	hexabromocyclododecane
HCH	hexachlorocyclohexane
HHCB	polycyclic musk fragrance, trade name Galaxolide
IFAS	integrated fixed-film activated sludge
LAS	linear alkylbenzene sulphonates
MA	nitro musk fragrance, trade name Musk ambrette
MEE2	mestranol
MGD	million gallons per day
MK	nitro musk fragrance, trade name Musk ketone
MM	nitro musk fragrance, trade name Musk moskene
MT	nitro musk fragrance, trade name Musk tibetene
MX	nitro musk fragrance, trade name Musk xylene
na	not analysed
nd	not detected
ng/g	nanograms/gram
N-MePFOSAA	2-(N-methylperfluorooctanesulfonamido)acetate
N-EtPFOSAA	2-(N-ethylperfluorooctanesulfonamido)acetate
NP	nonylphenol
NPE	nonylphenol ethoxylate
OP	octylphenol

PBDE	polybrominated diphenyl ethers
PCP	pentachlorophenol or personal care product
PFDA	perfluorodecanoic acid
PFDS	perfluorodecane sulfonate
PFDoDA	perfluorododecanoicacid
PFHxS	perfluorohexanesulfonate
PFNA	perfluorononanoic acid
PFOA	perfluorooctanoic acid
PFOS	perfluorooctanesulfonate
PFOSA	perfluorooctane sulfonamide
PFOSSA	perfluorooctanesulfonamidoacetate
PFTA	perfluotetradecanoic acid
PFUnDA	perfluoroundecanoic acid
RAS	return activated sludge
SRT	solids retention time
TBBPA	tetrabromobisphenol A
TNSSS	Targeted National Sewage Sludge Survey
TS	total solids
WAS	waste activated sludge

EXECUTIVE SUMMARY

INTRODUCTION

The Biosolids Task Group (BTG) established by the Canadian Council of Ministers of the Environment (CCME) is mandated to study and make recommendations on biosolids management at the national level. Wastewater treatment facilities (WWTF) across Canada generate residual wastewater solids (biosolids) that require treatment for safeguarding human health and the environment prior to their use or disposal. Options for disposal, recovery or recycling of biosolids include energy, nutrient or material recovery, landfilling, incineration, managed land application, land reclamation, and commercial product recovery (compost and pellets).

The end use of the biosolids is often governed by the constituent quality of the biosolids, such as nutrients, metals, pathogens and trace constituents. At present, there is a notable lack of sound science regarding the significance of a certain class of micro-constituents termed emerging contaminants (ECs), which include an array of pharmaceuticals, personal care products and industrial contaminants (such as plasticizers, surfactants and brominated flame retardants). While there is some documentation of ECs in biosolids, no focused study has been completed yet on an inventory of ECs in Canadian biosolids. This literature review documents the occurrence of ECs in biosolids. It will provide a basis for conducting a targeted sampling program which CCME can use to evaluate and manage the risks associated with ECs in biosolids destined for managed land application, land reclamation, production of commercial soil amendments and energy production.

METHODOLOGY

Following a computerized literature search, a citation review template was created in MS-Excel to capture the relevant data extracted form each citation. Two major categories were identified for the concentration data provided, namely for "occurrence" purposes, and for "process removal efficiency" purposes. Data classified for occurrence assessment were those in raw sludge or in treated biosolids streams which had not been adjusted in any manner, such as by spiking to elevate concentrations. Removal efficiencies reported in the original literature were used when reported. Data identified for assessment of process removal efficiencies included reported removal efficiencies based on processes deliberately spiked prior to treatment, as well as non-spiked treatment processes. The scale of the tests was identified in the spreadsheet summaries to allow assessment of possible differences between laboratory or pilot-scale studies, and those conducted at full-scale.

The major categories of substances identified in the literature review include:

- Industrial chemicals (plasticizers, pesticides, perfluorinated organic compounds, solvents, etc.)
- Alkylphenols and their ethoxylates
- Brominated flame retardants
- Hormones and sterols
- Pharmaceuticals
- Personal Care Products

- Certain metals (arsenic, silver selenium, mercury, etc.)
- Other (e.g. polyaromatic hydrocarbons, polychlorinated dioxins and furans)

At the start of the literature review, other potential categories were identified, such as nanoparticles and prions. No literature citations were found for these substances, and thus they were dropped from the literature review.

Categories of biosolids treatment processes in this review include:

- Anaerobic digestion
- Aerobic digestion
- Composting
- Lime addition
- Heat drying
- Other drying (e.g. air or solar drying)
- Other treatment
- Unknown

LITERATURE REVIEW SUMMARY

Industrial Chemicals

The compounds included in the industrial category in this review are diverse in their chemical properties and uses. The occurrence data are found more readily than are removal efficiency data. The plasticizer bis(2-ethylhexyl)phthalate has been characterized more frequently than have other phthalate esters or similar compounds. Limited data suggest it can be removed by some biosolids treatment processes, including anaerobic digestion. The plastic-associated chemical BPA is detected in most raw and digested sludges. Limited data reviewed herein indicate it is relatively unchanged by most biosolids treatment processes, based on a comparison with concentrations in other sludges. Although perfluorooctanoic acid (PFOA) and perfluorooctanesulfonate (PFOS) are the dominant species of the most frequently reported perfluorinated organic compounds, other less well recognized compounds may well be present at similar concentrations as well. Biotransformation of PFOS and PFOA in both aerobic and anaerobic environments may play a role in the presence of the compounds such as 2-(N-methylperfluorooctanesulfonamido)acetate and 2-(N-ethylperfluorooctanesulfonamido)acetate.

Relative to the other micro-constituents discussed in this review, linear alkylbenzene sulfonates (LAS) are present at very high concentrations. In limited data, sludges aerobically digested sludge from Germany and untreated sludges from Spain had lower concentrations of LAS than did anaerobically digested sludges from the same and other countries. The most commonly reported phenolic compound was pentachlorophenol (PCP). Data reviewed indicated that PCP concentrations in sludge can be reduced during anaerobic digestion. Very high pesticide residues were reported in a variety of sludge types by Ruel et al. (2008), demonstrating the persistence of these compounds. A comprehensive comparison of pesticide concentrations in sludges from other countries is lacking. Solvent data were limited but were generally identified at low concentrations in sludge samples. In general, there was almost a complete lack of removal efficiency data by different biosolids treatment processes for the industrial chemicals.

Alkylphenols and their Ethoxylates

There appear to be differences in alkylphenol (AP) and their associated ethoxylate (APE) concentrations between biosolids samples collected from different countries, possibly due to different regulations for detergent product formulation. In raw sludges or aerobically digested sludges, concentrations of mono- and di-ethoxylated species of nonylphenol (NP) may occur at concentrations approximately equal to or even slightly greater than the parent nonylphenol. Of the biosolids treatment processes examined, anaerobic digestion consistently has the highest concentrations of 4-NP, as a result of anaerobic biotransformation processes converting mono- and di-ethoxylated species to the parent compound. Composting appears to result in lower concentrations of APs and APEs than does drying or lime treatment. Limited data suggest that lime treatment may result in lower reduction of APEs compared to composting or drying processes. A composting period of between 40 and 70 days is needed to reduce the starting level of 4-NP by over 90%.

Brominated Flame Retardants

The main group of compounds in the brominated flame retardant (BFR) category are the polybrominated diphenyl ethers (PBDEs). There are apparent differences in concentrations of different PBDE isomers in North America and other countries (e.g., Europe, Kuwait, and Australia). The isomer decabromodiphenyl ether (decabromo DPE, or BDE 209) was observed in all the samples at the highest concentration of any of the isomers, followed by the penta BDE99 and tetra BDE47 isomers. Available data indicate that the concentrations of the PBDE isomers are substantially lower in the solids streams prior to secondary treatment (e.g. primary sludge), and more concentrated in the solids streams following secondary treatment (i.e, return activated sludge, and dewatered secondary or mixed sludge). Anaerobic digestion may result in a reduction of decabromodiphenyl ether, but concentrations of lower brominated congeners may increase due to this biotransformation. There was insufficient information to determine if other biosolids treatment processes can result in reduction of PBDEs. Few occurrence data were identified for other BFRs such as tetrabromobisphenol A (TBBPA) and hexabromocyclododecane (HBCD). No reduction efficiency data for the BFRs in biosolids treatment processes were observed in the literature.

Pharmaceutical Compounds

This class of micro-constituents in sludges and biosolids includes many different sub-classes with different therapeutic uses. There is a wide range of data available for the different pharmaceuticals that may be present in sludges and biosolids. Some compounds like the anti-epileptic carbamazepine have been widely characterized, while others have only one or two references in the literature. As a result of there being limited occurrence data for many pharmaceuticals, there are even fewer data available investigating the reduction of these drugs in biosolids treatment processes. Anaerobic digestion is the treatment process for which most data on pharmaceutical concentrations prior to and after treatment have been recorded. Reduction of the compounds appears to be highly specific to each class of pharmaceutical.

Hormones and Sterols

The hormones 17α-ethinylestradiol (EE2), estrone (E1) and 17β-estradiol (E2) are among the most frequently characterized compounds of this category in sludges and biosolids. Estrone (E1) is higher in concentration than these other common estrogenic compounds. The concentration of the natural hormone progesterone was the highest of the other estrogens observed. Concentrations of androgens in sludges were reported less frequently than estrogens. Concentrations of plant

sterols in sludges and biosolids were among the highest observed in this literature review, with concentrations in the tens of thousands of ng/g TS. Composting and heat drying resulted in lower concentrations of the phytosterols in the biosolids; anaerobic digestion treatment had the highest concentrations of the four treatment methods. Concentrations of the animal sterols reported in sludges varied substantially from one reference or source to the next, but were in any case among the highest concentrations observed in this review. Composting resulted in the lowest observed concentrations of both cholesterol and 3β-coprostanol in different biosolids treatment processes. Removal efficiencies up to 85% were recorded for both 17α-ethinylestradiol (EE2) and a mixture of estrone (E1) and 17β-estradiol (E2) in both thermophilic and mesophilic anaerobic digestion. Removal efficiency data for hormones and sterols in other biosolids treatment processes are scarce.

Personal Care Products

The publications reviewed have centred almost exclusively on the presence of the synthetic musk fragrance compounds, with only limited data on fluorescent whitening agents. There is negligible focus on other personal care products (PCP) compounds identified in CCME's Review of the State of Knowledge of Municipal Effluent Science and Research: Review of Effluent Substances. Polycyclic musks are present at higher concentrations than nitro musks. HHCB and AHTN are the predominant polycyclic musks, followed by ATII. The two main nitro musks identified in sludge samples were Musk ketone and Musk xylene. Full-scale anaerobic digestion does not appear to reduce concentrations of polycyclic musks in sludges, with concentrations in the digested sludges higher than in the raw sludge. Laboratory spiked anaerobic digestion studies indicate reductions in AHTN and HHCB concentrations are possible. Aerobic treatment appears to cause a reduction in concentrations of polycyclic musks. There are insufficient data reported in the literature to determine the effectiveness of the different biosolids treatment processes for reductions of the compounds.

Metals and Metalloids

The substances included in this review include a number of elements that include both metals and metalloids, such as arsenic and selenium, which will be referred to as "metals" for simplicity. The concentration database for metals and metalloids is limited because this review was focused on data from the year 2000 on, and much of the documented research on metals occurred previously. After iron and aluminum, the metals of highest concentration are zinc and copper, two metals commonly used in household plumbing. There are few data characterizing concentrations of elements such as selenium, thallium, antimony and molybdenum and others in biosolids. Organotin compounds are present in sludges at low concentrations of less than 1 mg/kg TS (less than 1,000 ng/g TS). Limited data suggest the organotin compounds are not reduced in concentration by anaerobic digestion.

Other Substances

This section brings together compounds which were not readily included in the previous sections. The major groupings include the polyaromatic hydrocarbons (PAHs) and polychlorinated polyaromatic compounds (biphenyls (PCBs); dibenzofurans (PCDFs); and dibenzo-p-dioxins (PCDDs)). The upper range of naphthalene, methylnaphthalene isomers and benzo(a)anthracene were at or above 100,000 ng/g TS in the literature review of Harrison et al. (2006), although a survey of Canadian sludges resulted in median concentrations typically lying in the range of 100 to 1,500 ng/g TS. The simplest PAHs, naphthalene and phenanthrene,

consisting of two and three fused benzene rings, respectively, have the highest median concentrations of the PAHs in a survey of Canadian sludges. Data on the effect of biosolids treatment processes on reducing concentrations of PAHs are very limited. For the lower molecular weight PAHs anthracene and phenanthrene, composted and air dried biosolids have apparent lower concentrations than biosolids produced by heat drying or after anaerobic digestion. Anaerobic digested sludge had the highest concentrations of the PAHs examined in the biosolids from different treatment processes. For the literature surveyed, the range and means of the PCDDs and PCFDs reported from different countries appear to be very similar, with mean values in the range of 0.020 ng TEQ/g TS.

ANALYSIS OF LITERATURE REVIEW

There is a great disparity in the attention devoted to the occurrence of different micro-constituents in sludges and biosolids. Some compounds have been examined comprehensively, and there is a substantial database on occurrence of these substances. Examples of the well-documented substances include the plasticizer bis(2-ethylhexyl)phthalate, the surfactant nonylphenol and some of its ethoxylates, the synthetic musk fragrances HHCB and AHTN, the flame retardants polybrominated diphenyl ethers, and the bacteriostat triclosan. In other cases however, there is a significant lack of information on concentrations of other substances in sludges and biosolids, including many antibiotics and other pharmaceuticals. The report on the State of Knowledge of Municipal Effluent Science and Research identified many classes of personal care products in treated effluents, including parabens (anti-microbial preservatives), sunscreen agents and insect repellents, for which no occurrence data in sludges or biosolids were identified. Lack of adequate analytical protocols may hinder this effort.

In much of the data reviewed, including other literature reviews, the sludges or biosolids are not specified by type (raw or treated, primary or secondary, etc) which makes the effort of determining if some processes are more beneficial than others in minimizing the concentrations of these substances. Some reviews, which summarize concentration data, group different types of sludges together without regard to the nature of the sludges. There appeared in the review to be evidence that sludges from some countries have lower concentrations than others, which may be reflective of manufacturing or use restrictions.

The treatment process most characterized for ability to reduce contaminant concentrations is the anaerobic process. For example in the studies by Carballa et al. (2006, 2007a, 2007b), concentrations of a variety of contaminants are provided for both raw sludge and sludges digested under different temperature regimes and retention times. From the review it appears that certain micro-constituents can be reduced by anaerobic digestion, while others are recalcitrant, or perhaps even increased in concentration, by anaerobic biotransformation processes.

With respect to different biosolids treatment processes, only the final treated biosolids concentrations are typically documented. Without the accompanying raw sludge concentrations, an evaluation of the effectiveness of the processes becomes more tenuous. In the data provided by Kinney et al. (2006), there appears to be evidence of some reduction in certain biosolids treatment processes such as composting or drying, but without raw sludge data, no firm conclusions can be drawn.

Based on the literature review and above assessment, the following recommendations are offered:

1. There is a need to define criteria for what is an adequate database for characterization of contaminants in sludges and biosolids, then to apply the criteria to the compiled data.
2. Where there are insufficient data, the availability of adequate analytical protocols needs to be determined. If they not available, then method development should be set as a priority.
3. If acceptable analytical protocols are available, characterization of sludges and biosolids treatment processes should follow, such as the planned follow-up field investigation accompanying this literature review project. Assessment of biosolids treatment effectiveness requires both raw and treated biosolids samples.
4. Lastly, data produced by this and similar reviews, and by the forth-coming field investigation, need to be transferred out to appropriate agencies and researchers.

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1.0 INTRODUCTION

1.1 BACKGROUND

The Biosolids Task Group (BTG) established by the Canadian Council of Ministers of the Environment (CCME) is mandated to study and make recommendations on biosolids management at the national level. Wastewater treatment facilities (WWTF) across Canada generate residual wastewater solids (biosolids) that require treatment for safeguarding human health and the environment prior to their use or disposal. Options for disposal, recovery or recycling of biosolids include energy, nutrient or material recovery, landfilling, incineration, managed land application, land reclamation, and commercial product recovery (compost and pellets).

The end use of the biosolids is often governed by the constituent quality of the biosolids, such as nutrients, metals, pathogens and trace constituents. At present, there is a notable lack of sound science regarding the significance of a certain class of micro-constituents termed emerging contaminants (ECs), which include an array of pharmaceuticals, personal care products and industrial contaminants (such as plasticizers, surfactants and brominated flame retardants). While there is some documentation of ECs in biosolids, no focused study has been completed yet on an inventory of ECs in Canadian biosolids. Consequently, the CCME has issued a Request for Proposals to document the occurrence of ECs in biosolids and to conduct a targeted sampling program which will provide a basis for the CCME to evaluate and manage the risks associated with ECs in biosolids with respect to managed land application, land reclamation, production of commercial soil amendments and energy production.

1.2 PROJECT OBJECTIVES

The objectives of this project are to:

1. Prepare a comprehensive review of research on ECs in biosolids within Canada and elsewhere based on technical literature and wastewater sector contacts;
2. Complete a survey and analyze biosolids samples with respect to ECs;
3. Identify those ECs of potential concern in Canadian biosolids;
4. Review and recommend treatment technologies that mitigate EC concentrations in biosolids;
5. Suggest Best Management Practices (BMPs);
6. Identify knowledge gaps and research needs for ECs with respect to biosolids;
7. Produce a final report of the project to the Contract and Project Authorities by October 30, 2009.

This draft report responds to Objective 1.

2.0 METHODOLOGY OF REVIEW

2.1 LITERATURE SEARCH AND IDENTIFICATION

In December of 2008, a computerised literature search was executed by Dr. Wayne Parker at the University of Waterloo with the objective of identifying citations pertaining to biosolids and contaminants.

The computerised literature search was supplemented with telephone calls to experts on the topic of ECs in biosolids. Telephone discussions were held with:

- Dr. Mel Webber of Webber Environmental, Burlington, ON
- Shirley Anne Smythe of Environment Canada, Burlington, ON
- Dr. George O'Connor of the Dept. of Soil and Water Science, the University of Florida at Gainesville
- Dr. Sally Brown of the University of Washington

2.2 LITERATURE COMPILATION

A citation review template was created in MS-Excel to capture the relevant data extracted form each citation. An initial data review session with Hydromantis and Dr. Parker assessed the nature and quality of the information extracted to mid-January. Two major categories were identified for the concentration data provided, namely for "occurrence" purposes, and for "process removal efficiency" purposes. Data classified for occurrence assessment were those in raw sludge or in treated biosolids streams which had not been adjusted in any manner, such as by spiking to elevate concentrations. Removal efficiencies reported in the original literature were used when reported. The Project Team decided not to calculate removal efficiencies from any of the original data after it appeared that values of the efficiencies calculated by the Team might differ form those in the original literature. Data identified for assessment of process removal efficiencies included reported removal efficiencies based on processes deliberately spiked prior to treatment, as well as non-spiked treatment processes. The scale of the tests was identified to allow assessment of possible differences between laboratory or pilot-scale studies, and those conducted at full-scale.

During the data review, it became clear that a number of issues needed to be addressed in interpreting the data. Analytical procedures needed to be checked to determine the basis of the matrix reported. For example, in some cases, only the liquid fraction of the sample was analyzed, while in other cases the analysis was performed on the whole sample including solids. In yet other cases, the total concentration was estimated using the analysed liquid concentration, to which was added a contaminant mass associated with the solids, estimated from a solids partition coefficient, to obtain the total concentration.

A second issue was consistency in the reported use of units of concentration. The units of measurement were variously reported on a volumetric liquid basis (e.g., ng/L), a solids mass basis (e.g., ng/g dry solids), or in other units such as ng/g of organic carbon.

The research team determined that because of the large number of potential contaminants to report, and the number of citations for reviewing, that data entry consistency was necessary. A principal concern was the manner in which non-detectable concentrations were entered in the master spreadsheet. If a compound was included in the analytical test group, but was reported in the original reference as non-detected, it was so classified in the spreadsheet. If a contaminant was not included in the analytical test group, the cell was left blank. The distinction is necessary to differentiate between compounds that were looked for but were not detected (affects the occurrence interpretation) and those that were not included for identification (no impact on occurrence assessment).

Presentation of the data in a publication required another decision regarding presentation. Mean or median values were also entered in the spreadsheet if reported in the original citations. When several concentrations within a category were documented within the same citation, a range could then be reported (e.g. n.d. – 20 ng/L).

2.3 CLASSIFICATION OF SUBSTANCES

The major categories of substances identified in the literature review include:

- Industrial chemicals (plasticizers, pesticides, perfluorinated organic compounds, solvents, etc.)
- Alkylphenols and their ethoxylates
- Flame retardants
- Hormones, steroids and sterols
- Pharmaceuticals
- Personal Care Products
- Certain metals (arsenic, silver selenium, mercury, etc.)
- Other (e.g. polyaromatic hydrocarbons, polychlorinated dioxins and furans)

At the start of the literature review, other potential categories were identified, such as nanoparticles and prions. No literature citations were found for these substances, and thus they were dropped from the literature review.

2.4 CLASSIFICATION OF BIOSOLIDS TREATMENT PROCESSES

Categories of biosolids treatment processes in this review include:

- Anaerobic digestion
- Aerobic digestion
- Composting
- Lime addition
- Heat drying
- Other drying (e.g. air or solar drying)
- Other treatment
- Unknown

3. RESULTS OF LITERATURE REVIEW

3.1 INDUSTRIAL COMPOUNDS

3.1.1 Introduction

The compounds included in this section are those which are produced or used as industrial chemical products or intermediates. This review is not intended to provide a complete historical record of concentrations of all industrial compounds in sludges and biosolids, as past surveys completed through the 1970s and through 1990s have documented these as "legacy" contaminants from the U.S. EPA's Priority Pollutant List. The intent of this review is to provide more recent data from approximately 2000 onward to the present.

The main categories of industrial compounds examined in this Section include:

- Plasticizers and Bisphenol A
- Perfluorinated organic acid and derivative compounds
- Pesticides
- Surfactants (excluding alkylphenol and ethoxylates, discussed separately)
- Chlorophenols
- Solvents, and
- Miscellaneous compounds not covered elsewhere.

3.1.2 Plasticizers and Metabolites

Plasticizers are added to polymeric materials to increase flexibility and suppleness. Phthalate and adipate esters are two common classes of plasticizers. Concentrations of phthalate esters found recently in the literature are provided in Table 1. Data for an array of phthalate esters provided by Harrison et al. (2006) in their sludge literature review and by Tan et al. (2007) for return activated sludge samples, clearly shows that bis(2-ethylhexyl) phthalate (BEHP) is the predominant compound in this class, at concentrations several orders of magnitude higher than the other phthalate esters. Concentration data from several nations provided in Table 1 focus almost exclusively on BEHP, without analysis or reporting of the other phthalate compounds. The mean concentration of BEHP in Swedish sludges at 24,500 ng/g TS appeared to be lower than the concentrations reported in sludges from other countries.

In addition to phthalate esters, other similar types of compounds are used as plasticizers. Concentrations of the compounds bis(2-ethylhexyl) terephthalate and bis(2-ethylhexyl) adipate are shown in Table 2 for two sludge samples from a Treatment facility in Montreal (Barnabé et al., 2008). Comparing concentrations of these two compounds with those of BEHP in the same sludges in Table 1 indicates that the terephthalate and adipate esters are present in similar concentrations to the BEHP. The chemicals 2-ethylhexanol and 2-ethylhexanal are metabolites of the bis(2-ethylhexyl) organic acid esters (phthalates, adipates, terephthalates, etc.). The aldehyde (2-ethylhexanal) was observed at a higher mean concentration than was the alcohol (2-ethylhexanol) in the data of Barnabé et al. (2008), particularly in the dewatered sludge sample.

Table 1. Concentrations of Phthalate Esters in Sludges and Biosolids

Sludge Source	Concentration (ng/g TS)						Reference
	Dimethyl phthalate	Diethyl phthalate	Di-n-butyl phthalate	Butylbenzyl phthalate	Bis (2-ethylhexyl) phthalate	Di-n-octyl phthalate	
Not specified (literature review)	26	18	393	201	62,480	570	Harrison et al. (2006)
Return activated sludge (plant 1)	not analysed	39.9±24.3[a]	149±80.4	25.7±9.4	9,910 ± 2,770	not analysed	Tan et al. (2007)
Return activated sludge (plant 2)	not analysed	17.2±12.0	12.6±3.4	11.0±9.5	2,200 ± 1,480	not analysed	
Norway sludges					27,000 - 115,000 (83,000)[b]		Jaganyi (2007)
Sweden sludges					25,000 - 661,000 (170,000)		
Denmark sludges					3,900 - 170,000 (24,500)		
Canadian sludge (1995-1998)					1,600 - 245,000 (160,000)		XCG (2007)
Homogenized sludge					80,000 ± 10,000		Barnabé et al. (2008)
Dewatered sludge					90,000 ± 12,000		

[a] mean ± standard deviation [b] range (median)

Table 2. Concentrations of Other Plasticizers and Metabolites in Sludges and Biosolids

Sludge Source	Concentration (ng/g TS)					Reference
	Bis (2-ethylhexyl) terephthalate	Bis (2-ethylhexyl) adipate	2-ethyl-hexanol	2-ethyl-hexanal	2-ethyl-hexanoic acid	
Homogenized sludge	45,000 ± 2,300[a]	34,000 ± 1,000	12,500 ± 900	34,000 ± 1,400	20,700 ± 400	Barnabé et al. (2008)
Dewatered sludge	104,000 ± 5,00	340,000 ± 10,000	4,500 ± 300	85,000 ± 3,400	14,600 ± 300	

[a] mean ± standard deviation

Concentrations of plasticizer and chemical intermediate compounds in treated biosolids samples are presented in Table 3. The highest concentration of BEHP was observed in the group of biosolids treatment data developed by Ruel et al. (2008), consisting of anaerobically digested, limed and dried biosolids. BEHP concentrations in the other reported literature were in the range of 15,000 to 53,000 ng/g TS. Barnabe et al. (2008) reported on concentrations of three plasticizers in dried sludge from the Montreal QC, treatment plant. Gibson et al. (2007) provided concentrations of BEHP at the inlet and outlets of composting and heat drying processes. Composting appeared to result in lower concentrations than heat drying, but the data are limited.

Table 3. Concentrations of Plasticizers and Chemical Intermediates following Biosolids Treatment Processes

Biosolids Treatment	Concentration (ng/g TS)					Reference
	Bis (2-ethyl-hexyl) phthalate	Bis (2-ethylhexyl) terephthalate	Bis (2-ethylhexyl) adipate	2-ethyl-hexanol	2-ethyl-hexanal	
Dried sludge	15,000 ± 2000[a]	12,200 ± 600	19,300 ± 1,000	nd	nd	Barnabé et al., (2008)
Anaerobic digestion, limed, drying	2,197,000 ± 11,000,000					Ruel et al., (2008)
Composting	In = 53,000 out = 15,000					Gibson et al., (2007)
Heat drying	In = 44,000 out = 34,000					
Compost	27,900 - 154,000					Williams (2007)

nd = not detected [a] mean ± standard deviation

Removal in Biosolids Treatment Processes

Removal efficiency data in biosolids treatment processes were only found for BEHP (Table 4). Composting appeared to result in effective reduction of BEHP, with removal efficiencies of 64% and 70% reported by Gibson et al. (2007) and Williams (2007), respectively. The limited data suggested heat drying and anaerobic digestion were less effective at reducing the concentration of BEHP in feed sludge than was composting.

Table 4. Removal of the Plasticizer BEHP in Biosolids Treatment Processes

Treatment Process	Removal Efficiency (%)	
composting	64%	70%
heat dry	23%	
anaerobic digestion		32%
Reference	Gibson et al. (2007)	Williams (2007)

3.1.3 Bisphenol A

Bisphenol A (BPA) is mostly used to manufacture polycarbonate plastics and epoxy resins. Uses of the compound are for food and beverage storage, and in sealants in canned food products. Entry to the wastewater system is possible through food preparation and clean-up, and through human excretion after oral intake. The primary concerns with BPA related to food and drink packaging relate to possible harmful effects on the brain, behaviour and prostate gland of foetuses, infants and children (U.S. NIH, 2009).

Bisphenol A has received considerable attention in wastewater sludges and biosolids. Lee and Peart (2002) included BPA as a target analyte in a survey of Canadian raw and digested sludges. The data are summarized in Table 5. The highest concentration of BPA in that survey (39,800 ng/g TS) was observed in raw sludge from the Toronto Highland Creek Wastewater Treatment Plant, whereas the minimum concentration was noted in a digested sludge sample from the North Toronto sewage treatment facility. Other high concentrations of BPA were observed on the digested sludges of highly urbanized centres such as Windsor (ON), Galt (ON), Hamilton (ON) and Edmonton (AB). Median concentrations of BPA in the raw and digested sludge samples were 280 and 555 ng/g TS, leading to the conclusion that BPA is not removed during sludge anaerobic digestion.

Concentrations of BPA in other sludge samples are summarized in Table 6. The range of concentrations in this table from minimum to maximum is very broad, as indicated by the range supplied by Harrison et al. (2006). The observed minimum value of 0.1 ng/g TS recorded by Harrison et al. (2006) is lower than the values of 3 – 4 ng/g TS reported by Tan et al. (2007) for return activated sludges. The high value of 32,100,000 ng/g TS from Harrison et al. (2006) is several orders of magnitude higher than maximum values found in the other literature cited in Table 5.

Biosolids Treatment Processes

Only limited data were identified which characterised the concentrations of BPA resulting from biosolids treatment processes. BPA concentrations in several treatment processes documented by Kinney et al. (2006) are summarised in Table 7. Of the various treatment processes, the concentration of BPA was lowest in the heat dried biosolids and highest in the anaerobically digested sludge. Additional data are needed to determine if these trends can be extrapolated on a more universal basis.

Table 5. Concentrations of Bisphenol A in Canadian Sludges (Lee and Peart, 2002)

Municipal Treatment Plant and Sludge Type	Bisphenol A (BPA) concentration (ng/g TS)
Edmonton (Goldbar) Raw	310
Regina Raw	280
Adelaide Raw	250
Burlington Raw	190
Ottawa Raw	450
Toronto (Ashbridges Bay) Raw	180
Toronto (Highland Creek) Raw	39,800
Toronto (Humber) Raw	1,710
Toronto (North) Raw	320
Montreal (MUC-PSI) Raw	1,060
Quebec City Raw	160
Quebec City Raw	130
Vancouver Digested	300
Vancouver Digested	440
Calgary (Bonnybrook) Digested	800
Calgary (Fish Creek) Digested	790
Edmonton (Goldbar) Digested	3,180
Regina Digested	490
Saskatoon Digested	260
Saskatoon Digested	1,170
Burlington Digested	1,860
Galt Digested	9,560
Guelph Digested	460
Hamilton Digested	4,440
Ingersoll Digested	470
Kitchener Digested	230
Ottawa Digested	640
Waterloo Digested	2,540
Windsor Digested	11,100
Toronto (Ashbridges Bay) Digested	620
Toronto (Humber) Digested	280
Toronto (North) Digested	100
Granby Digested	240
Moncton Digested	130
Truro Digested	300
Median raw	280
Median digested	555

3.1.4 Perfluorinated Organic Acid and Derivative Compounds

Perfluorinated organic compounds and derivative products have been used as stain repellents for fabrics, non-stick cookware and food wrappers, personal care products and fire-fighting foams. The major producer of the compounds in North America, the 3M Company, voluntarily phased

Table 6. Concentrations of Bisphenol A in Other Sludges and Biosolids

Sludge Source	Sludge Type	Concentration (ng/g TS)	Reference
Toronto sewage sludge	raw sludge	70 – 11,000 (680)[a]	Webber and Sidwha (2005)
	digested sludge	120 – 13,000 (1090)	
(Literature review)	not specified	0.10 – 32,100,000	Harrison et al. (2006)
Greek sludge	dewatered secondary or anaerobic digested sludge	560 – 1,750 (530)	Stasinakis et al. (2008)
Return activated sludge	Plant A	3.8±4.3[b]	Tan et al. (2007)
	Plant B	3.1±1.2	
Various	Not specified	4 – 1,363	Williams (2007)
Plant G	Waste activated sludge	1,620[c]	Kinney et al. (2006)
Plant H	Dewatered	1,090[c]	

[a] range (median) [b] mean ± standard deviation [c] ng/g organic carbon

Table 7. Concentrations of Bisphenol A following Biosolids Treatment Processes (Kinney et al., 2006)

Biosolids Treatment	Concentration (ng/g OC)
heat drying	1,680
composting	4,690 – 9,030
other drying	3,550
Anaerobic digestion	14,400

out production in the year 2000. These compounds are persistent (Sinclair and Kannan, 2006) and bioaccumulative (Swackhamer et al., 2004). The probable source of the compounds in domestic wastewater is through routine household activities such as bathing, cooking dishwashing and laundry.

Concentrations of the perfluorinated compounds in various sludges and biosolids are provided in Table 8. There are many compounds in this class as is evident from the table. In many other sludges, the predominant compounds are the perfluorooctanoic acid (PFOA) and perfluoroctanesulfonate (PFOS). The maximum concentration of PFOA found in this review was 241 ng/g TS found in a sample of a sludge from New York state (Sinclair and Kannan, 2006) and the maximum for PFOS was 160 ng/g TS in a sample of Oregon sludge (Schultz et al., 2006). Concentrations of perfluorodecanoic acid (PFDA) and perfluoroundecanoic acid (PFUnDA) were found at higher mean concentrations of 52 and 60 ng/g TS, respectively in sludge from Plant A in NYS in the testing by Sinclair and Kannan (2006), although in most other studies these compounds had much lower concentrations. In the only study in this review with perfluorodecane sulfonate (PFDS) as a target compound, Schultz et al. (2006) observed concentrations of a similar magnitude as those for PFOS. Perfluorohexane sulfonate (PFHxS) and perfluorododecanoic acid (PFDoDA) were found in relatively low concentrations compared to the predominant compounds identified above.

Table 8. Concentrations of More Common Perfluorinated Organic Acids and Derivatives in Sludges and Biosolids

Compound	Concentration (ng/g TS)									Reference
	Perfluoro-octanoic acid (PFOA)	Perfluoro-octane sulfonate (PFOS)	Perfluoro-octane sulfonamide (PFOSA)	Perfluoro-hexane sulfonate (PFHxS)	Perfluoro-nonanoic acid (PFNA)	Perfluoro-decanoic acid (PFDA)	Perfluoro-decane sulfonate (PFDS)	Perfluoro undecanoic acid (PFUnDA)	Perfluoro-dodecanoic acid (PFDoDA)	
Sewage sludge - Denmark	0.7-19.7 (4)[a]	4.8-74.1 (18.4)	0.5-3.6 (0.8)	0.4-10.7 (3.6)	0.4-8.0 (1.5)	1.2-32.0 (7.2)		0.5-4.4 (1.2)		
Bossi literature review	0.3-0.7 (0.5)	0.3-1.0 (0.6)		0.09-0.01 (0.01)	<0.6-0.2 (0.1)					Bossi et al., (2007)
Plant A: dewatered cake	39	154	24	<2.5	<13	47		6.9	12	
Plant A: solar dried sludge	8.3-219	8.2-110	<2.5-21	<2.5	<2.5-4.4	2.5-34		<2.5-7.7	<2.5-28	Loganathan et al., (2007)
Plant B: dewatered cake	15	20	<2.5	<2.5	<2.5-11	19-41		<2.5	<2.5-10	
Plant B: Ash	7.0 - 35	<2.5 - 50	<2.5 - 7.0	<2.5	<2.5	7.0 - 35		<2.5	<2.5	
NYS Plant A	69 - 241 (144)	26 - 65 (37)		<10 - 18 (<10)		25 - 91 (52)		35 - 115 (60)		Sinclair and Kannan (2006)
NYS Plant B	18 - 89 (80)	<1-0 - 34 (25)		<10		<25 - 39 (27)		<25		
Primary Sludge	<6 - 12 (7.1)	18 - 3.8 (53)		nd - 12 (3.4)	nd - 10 (4.2)	1.6 - 3.9 (2.8)	14 - 2.9 (19.4)	2.0 - 4.2 (2.6)	1.3 - 1.6 (1.5)	
Return Act.iv. Sludge	<6 - 8.2 (6.7)	31 - 55 (43)		nd	3.1 - 4.9 (3.8	7.2 - 0.8 (9.1)	94 - 140 (130)	7.7 - 0.5 (9.2)	6.1 - 7.8 (7.1)	
Thickened Sludge	<6	20 - 18 (42)		nd	nd	3.4 - 5.3 (3.9)	57 - 71 (62)	3.9 - 5.0 (4.4)	4.1 - 5.1 (4.3)	Schultz et al., (2006)
Anaerobic Digested Sludge	<3	81 - 160 (100)		nd	9.2 - 0.3 (9.9)	5.4 - 6.4 (5.9)	90 - 93 (91)	5.9 - 8.4 (6.8)	3.6 - 4.2 (3.8)	

nd = not detected [a] range (mean)

Several other perfluorinated organic compounds were identified by Schultz et al. (2006) in the study of sludges from an Oregon treatment plant. Data for most of these compounds are provided in Table 9. The magnitude of these compounds, with the exception of the perfluotetradecanoic acid (PFTA), is at least as great as for the more commonly analyzed PFOA and PFOS compounds. The presence of the compounds 2-(N-methylperfluorooctanesulfonamido)acetate (N-MeFOSAA) and 2-(N-ethylperfluorooctanesulfonamido)acetate (N-EtFOSAA) are thought to occur as metabolites of parent sulfonamido alcohols during aerobic secondary treatment with adsorption on the settled mixed liquor (Schultz et al., 2006). These data indicate that more compounds can be detected in sludge samples when the target analyte list is expanded.

Table 9. Concentrations of Additional Perfluorinated Organic Acids and Derivatives in Sludges and Biosolids (Schultz et al., 2006)

Sludge Type	Concentration (ng/g TS)			
	PFOSAA	N-MePFOSAA	N-EtPFOSAA	PFTA
Primary Sludge	<3 - 3.4 (<3)[a]	5.2 - 8.9 (6.3)	15 - 5.8 (20)	nd
Return Activated Sludge	14 - 23 (19)	99-61 (140)	90 -140 (120)	<3
Thickened Sludge	6.2 - 7.6 (6.9)	35 - 52 (41)	43 - 52 (48)	0.9 - 1.3 (1.2)
Anaerobic Digested Sludge	9.4 - 12.4 (11)	130 - 140 (130)	91 - 100 (98)	<3

PFOSSA = perfluorooctanesulfonamidoacetate
N-MePFOSAA = 2-(N-methylperfluorooctanesulfonamido)acetate
N-EtPFOSAA = 2-(N-ethylperfluorooctanesulfonamido)acetate
PFTA = perfluotetradecanoic acid
[a] range (mean)

No removal efficiency data in biosolids treatment processes were found for these chemicals. The data presented by Schultz et al. (2006) indicate that biotransformation of these compounds and precursor compounds can occur in both aerobic and anaerobic environments, complicating efforts to quantify "removals" through the treatment processes.

3.1.5 Surfactants

Because of the significant body of data on alkylphenols and their ethoxylates, a separate section has been devoted to those compounds. Otherwise, data identified in the more recent literature examine the linear alkylbenzene sulphonates (LAS). Concentration data for this class of surfactants is presented in Table 10. Note that concentration units are expressed as µg/g TS, rather than the more usual concentration units of ng/g TS for micro-constituents. Relative to the other micro-constituents discussed in this review, LAS are present at very high concentrations. In the review by Jayangi (2007), sludges aerobically digested sludge from Germany and untreated sludges from Spain had lower concentrations of LAS than did anaerobically digested sludges from the same and other countries. There are insufficient data to determine whether aerobic digestion might result in lower concentrations of LAS in the treated sludge than anaerobic digestion.

In this review, no data were identified on treatment of removal of these compounds in biosolids treatment processes.

Table 10. Concentrations of Linear Alkylbenzene Sulfonates in Sludges and Biosolids

Sludge Source	Concentration (µg/g TS)	Reference
sludges (not specified)	1,000 - 10,000	Fent (1996)
Norway – sludges (not specified)	1 - 424	Jaganyi (2007)
Denmark – sludges(not specified)	11 – 16,100	
Germany – anaerobically digested sludges	1,600 – 11,800	
Germany – aerobically digested sludges	182 – 432	
Italy - anaerobically digested sludges	11,500 – 14,000	
Spain - anaerobically digested sludges	12,100 – 17,800	
Spain – untreated sludges	40 – 700	
Switzerland - anaerobically digested sludges	2,900 - 11,900	
UK - anaerobically digested sludges	9,300 – 18,800	
primary sludge	5,340 -- 6,310	Angelidaki et al. (2004)
Anaerobically digested (literature)	2,000 - 30,200	
Aerobically digested (literature)	100 - 2,900	
Air-dried digested (literature)	150 - 160	

3.1.6 Phenolic Compounds

This literature review has not focused on the longer-term historical data of phenolic compounds in sludges and biosolids because, as legacy contaminants, much has been published on their concentrations prior to 2000. A substantial body of occurrence data of phenolics in sludges was compiled by Monteith (1987). The focus of this review is directed more at micro-constituents or compounds of emerging concern rather than on the legacy contaminants.

Compounds included in this category include chlorinated phenols and para-cresol, a simple alkylphenol. Chlorinated phenols are used as disinfectants, biocides, preservatives, dyes, pesticides and industrial and medical organic chemicals (CCREM, 1987). Pentachlorophenol (PCP) is used as a wood preservative. Cresol solutions are used as household cleaners, disinfectants and deodorizers as well as chemical intermediates in insecticide production. In a survey of Canadian municipal raw and anaerobically digested sludges (Table 11), PCP concentrations ranged from a high value of 131 ng/g TS in Edmonton raw sludge to a low value of 7.1 ng/g TS in a digested sludge sample from the Toronto Ashbridges Bay Wastewater Treatment Plant (Lee and Peart, 2002). Median concentrations in the raw and digested sludge samples were 29 and 18 ng/g TS, indicating the potential for a reduction in the PCP concentrations by anaerobic digestion.

Table 11. Pentachlorophenol (PCP) Concentrations in Canadian Municipal Sludges (Lee and Peart, 2002)

Municipal Treatment Plant and Sludge Type	Pentachlorophenol (PCP) concentration (ng/g TS)
Edmonton (Goldbar) Raw	131
Regina Raw	16.4
Adelaide Raw	56.6
Burlington Raw	27.7
Ottawa Raw	28.9
Toronto (Ashbridges Bay) Raw	45.8
Toronto (Highland Creek) Raw	22.9
Toronto (Humber) Raw	44.5
Toronto (North) Raw	20.1
Montreal (MUC-PSI) Raw	28.9
Quebec City Raw	80
Quebec City Raw	27.9
Vancouver Digested	99.4
Vancouver Digested	239
Calgary (Bonnybrook) Digested	10.5
Calgary (Fish Creek) Digested	71.9
Edmonton (Goldbar) Digested	119
Regina Digested	15.6
Saskatoon Digested	19.7
Saskatoon Digested	35.6
Burlington Digested	12.5
Galt Digested	8.4
Guelph Digested	15.1
Hamilton Digested	10.6
Ingersoll Digested	18.3
Kitchener Digested	17.8
Ottawa Digested	15.1
Waterloo Digested	85.1
Windsor Digested	13.5
Toronto (Ashbridges Bay) Digested	7.1
Toronto (Humber) Digested	11.1
Toronto (North) Digested	88.8
Granby Digested	54.5
Moncton Digested	58.3
Truro Digested	99.4
median raw	28.9
median digested	18.1

Concentrations of phenolic compounds in other sludges are presented in Table 12. The mean concentration of PCP in a variety of sludges and biosolids reported by Ruel et al. (2008) is substantially higher than the range of values reported by Lee and Peart (2002). The mean concentration of trichlorophenols, used as a chemical intermediate in pesticide production, was

much higher at 70,400 ng/g TS, although this value may be skewed upward by one or two high concentrations, based on the very high value of the standard deviation of the mean.

Table 12. Concentrations of Phenolic Compounds in Sludges and Biosolids

Compound	Sludge Source	Concentration (ng/g TS)	Reference
Para-cresol	African - Sewage sludges	350 - 5,370	Jaganyi (2007)
Trichlorophenols	anaerobic digestion, limed, drying	70,400 ± 297,000[a]	Ruel et al. (2008)
pentachlorophenol	anaerobic digestion, limed, drying	600 ± 1,800	

[a] mean ± standard deviation

3.1.7 Pesticides

This literature review has not focused on the longer-term historical data of pesticides in sludges and biosolids because, as legacy contaminants, much has been published on their concentrations prior to 2000. A substantial body of occurrence data of pesticides in sludges was compiled by Monteith (1987). The focus of this review is directed more at micro-constituents or compounds of emerging concern rather than on the legacy contaminants.

Pesticide concentration data from a number of review papers are summarized in Table 13. Note that these are predominantly chlorinated hydrocarbon pesticides. Concentrations of the pesticides appear to vary widely in the different source sludges. The data presented by Ruel et al. (2008) are among the highest observed. The limited data from Canadian sources for hexachlorobenzene and total DDT and metabolites (XCG, 2007) indicate that pesticide concentrations in sludges are lower than reported elsewhere, while the concentration of aldrin was of a similar magnitude as in the other reported study.

3.1.8 Solvents

As with the pesticide group, many solvents were included in the original U.S. EPA Priority Pollutant list of the 1980s and have received significant attention previously. Because the focus of this review is directed more at micro-constituents or compounds of emerging concern in sludges and biosolids than on the legacy contaminants, the review is limited in scope.

Concentration data for some common industrial solvents in sludge and biosolids are presented in Table 14. Concentrations of solvents reported in Canadian sludges (XCG, 2007) are less than 500 ng/g TS. Concentrations reported in different treated biosolids by Ruel et al. (2008) were present at substantially higher concentrations than those reported by XCG (2007).

Table 13. Concentrations of Pesticide Compounds in Sludges and Biosolids

Compound	Concentration (ng/g TS)					
	34 Canadian stabilized sludges (1987)	Canadian sludge (1995-1998)	anaerobic digestion, limed, drying	Swiss sludges	UK sludges	French sludges
Aldrin	300		4100 ± 7000[a]		10 - 200 (30)[b]	
Chlorpyrifos			500[c]			
Dieldrin			26,300 ± 30,000	100 - 2,000	10 - 5,300 (500)	
Diuron						11.2 - 46.6
Total pp'-DDT & op'-DDT & pp'-DDE		17		100 - 500		
Endosulfan			300[c]			
Endrin			500 ± 500		10 - 700 (100)	
Heptachlor			4,900 ± 3,300			
Hexachloro-benzene	10	5.4	1,200 ± 1,500			
Hexachloro-cyclohexane; Lindane; γ-HCH			12,300 ± 22,000	100 - 900	10 - 70,000 (400)	
Reference	XCG (2007)		Ruel et al. (2008)	Fent (1996)	Rogers (1996)	Ghanem et al. (2007)

[a] mean ± standard deviation [b] range (mean)
[c] no standard deviation calculated due to analytical difficulties

Table 14. Concentrations of Solvents in Sludges and Biosolids

Compound	Concentration (ng/g TS)		
	Canadian sludges (1995-1998)	34 Canadian stabilised sludges (1987)	anaerobic digestion, limed, drying
Benzene	0.04 - 483 (45)[a]		1,000
1-2-Dichloroethane	nd		
Dichloromethane	5 - 19 (8)		
Carbon tetrachloride	nd		
Trichloroethylene	0.3 - 380 (167)		
1,1,1- Trichloroethane	nd	200	
Tetrachloroethylene	nd - 231 (59)	300	8,100
Reference	XCG (2007)		Ruel et al., (2008)

nd = not detected [b] range (mean)

3.1.9 Miscellaneous Compounds

Few compounds are included in this group. The substance 4-chloroaniline is used as a chemical intermediate in the production of a number of products, including agricultural chemicals, azo dyes and pigments, and pharmaceutical and personal care products. In the U.S. EPA's TNSSS, 4-chloroaniline was found at a median concentration of 513 ng/g TS (U.S. EPA, 2009)

3.1.10 Section Summary

The compounds included in this review are diverse in their chemical properties and uses. The occurrence data are found more readily than are removal efficiency data. The main summary points follow.

1. The plasticizer BEHP has been characterized more frequently than have other phthalate esters or similar compounds. Limited data suggest it can be removed by some biosolids treatment processes, including anaerobic digestion.
2. The plastic-associated chemical BPA is detected in most raw and digested sludges. Limited data reviewed indicate it is relatively unchanged by most biosolids treatment processes, based on a comparison with concentrations in other sludges.
3. Although the perfluorinated organic compound perfluorooctanoic acid (PFOA) and perfluorooctanesulfonate (PFOS) are found in the highest concentrations of those reported most frequently, other less well recognized compounds may well be present at high concentrations as well. Biotransformation in both aerobic and anaerobic environments may play a role in the presence of the compounds such as 2-(N-methylperfluorooctanesulfonamido)acetate and 2-(N-ethylperfluorooctanesulfonamido)acetate.
4. Relative to the other micro-constituents discussed in this review, linear alkylbenzene sulfonates are present at very high concentrations. In limited data, aerobically digested sludge from Germany and untreated sludges from Spain had lower concentrations of LAS than did anaerobically digested sludges from the same and other countries.
5. The most commonly reported phenolic compound was pentachlorophenol. Data reviewed indicated that PCP concentrations in sludge can be reduced during anaerobic digestion.
6. Very high pesticide residues were reported in a variety of sludge types by Ruel et al. (2008), demonstrating the persistence of these compounds. A comprehensive comparison of pesticide concentrations in sludges from other countries is lacking.
7. Solvent data were limited but were generally identified at low concentrations in sludge samples.
8. In general, there was almost a complete lack of removal efficiency data by different biosolids treatment processes for the industrial chemicals.

3.2 ALKYLPHENOL AND THEIR ETHOXYLATES

3.2.1 Introduction

Alkylphenol ethoxylates (APEs) are among the most commonly used surfactants (surface active agents) around the world. Nonylphenol ethoxylates account for approximately 80% of the total use, while octylphenol ethoxylates represent most of the remaining 20% (Melcer et al., 2007).The predominant uses of APEs are in pulp and paper production, textile manufacturing and in the production of crop protection chemicals (Melcer et al., 2007). APEs enter municipal wastewater treatment facilities in industrial wastewater discharges to municipal sewers, as well as being present in domestic sewage. During biological (secondary or tertiary) wastewater treatment, alkylphenols (APs) with longer polyethoxylate chains are biotransformed to mostly mono- or di-ethoxylated APs, or to the parent AP itself (Melcer et al. 2007). Biological treatment also results in formation of carboxylated forms of the APEs, which are more soluble than the mono- or di-ethoxylated APs. As the polyethoxylate chain decreases, the compound becomes more hydrophobic (less water soluble), causing the compound to adsorb onto wastewater and sludge particles (Melcer et al. 2007). As a result, wastewater sludge streams tend to concentrate the parent AP and mono- or di-ethoxylated APs.

3.2.2 Occurrence Data

Raw and anaerobically digested municipal sludges from Canadian municipalities were surveyed for alkylphenols (AP) and their ethoxylates by Lee and Peart (2002). The results are provided in Table 15. In raw sludges, concentrations of 4-nonylphenol (NP) tended to dominate, but in a number of samples, the mono-ethoxylated NP and di-ethoxylated NP were of similar or greater magnitude (e.g., London Adelaide, Quebec City, Montreal). After anaerobic digestion, the concentration of 4-NP was substantially increased compared to levels in corresponding raw sludges. The median value of 4-NP in digested sludge samples was 413 μg/g TS, compared to a median concentration of 91 μg/g TS in the raw sludge samples. Concentrations of the di-ethoxylated NP and higher congeners were lower in digested sludges than in the raw sludges, suggesting transformation of the higher-ethoxylated species to the parent 4-NP and the mono-ethoxylated NP. Another AP compound, 4-tert-octyl phenol was reported in the study. The median concentration of 10.4 μg/g TS in the digested sludge was higher than the median value of 4.75 μg/g in the raw sludge.

Concentrations of APs and their ethoxylates in Toronto sludges reported by Webber and Sidwha (2005) are presented in Table 16. Concentrations of 4-NP were higher in digested sludges than in the raw sludge samples. Data from another Canadian survey (XCG Consultants, 2007) reported that 4-NP in mixed raw and digested sludges was the predominant form, approximately an order of magnitude higher than the mono-ethoxylated form of higher ethoxylated species (Table 16).

Table 15. Concentrations (μg/g TS) of Alkylphenol (AP) and Ethoxylates (EO) in Canadian Municipal Sludges (Lee and Peart, 2002)

Municipal Treatment Plant and Sludge Type	4-nonyl phenol (NP)	NP mono-EO (NP1EO)	NP di-EO (NP2EO)	NP tri-EO (NP3EO)	Higher NP-EOs NP(4-17)EO	4-tert-octyl phenol
Edmonton (Goldbar) Raw	237	107	22	5.3	28.4	6.9
Regina Raw	97.7	26.9	11.8	20.1	194	4
Adelaide Raw	20.1	60.9	60.2	23.3	10.6	2.9
Burlington Raw	90.3	98.7	38.5	9.6	44.7	8
Ottawa Raw	53.6	13.9	10.6	8.1	43.5	4.3
Toronto (Ashbridges Bay) Raw	126	129	73.7	11.9	3.5	8.6
Toronto (Highland Creek) Raw	112	94.9	40.5	13.8	15.7	13.6
Toronto (Humber) Raw	125	62.9	91.7	28.9	13	5.2
Toronto (North) Raw	92.7	69.4	34.6	<2	<2	5.4
Montreal (MUC-PSI) Raw	11.3	27.9	35.7	43.1	467	2.6
Quebec City Raw	12.4	24.6	19.8	15.1	141	2.9
Quebec City Raw	33.1	20.5	24.1	12.4	145	3.3
Vancouver Digested	457	124	26.6	17.7	47.6	10.7
Vancouver Digested	468	74.1	31.6	7.7	4.5	10.4
Calgary (Bonnybrook) Digested	413	154	33.1	<2	8.1	6.2
Calgary (Fish Creek) Digested	393	154	20.5	16.5	5.1	6
Edmonton (Goldbar) Digested	848	160	36.8	6	11.1	11.3
Regina Digested	568	228	1.8	1.9	11.5	10.8
Saskatoon Digested	26.5	39.3	39.3	6.8	2.1	1.9
Saskatoon Digested	139	97.2	24.8	4.5	<2	3.8
Burlington Digested	435	66	3.2	<2	17.7	13.1
Galt Digested	1210	126	24.1	12.4	23	20.5
Guelph Digested	1230	130	36.4	8.4	120	43.9
Hamilton Digested	403	114	26.4	6.9	5.5	15.6
Ingersoll Digested	232	32	6.7	3.9	67	8.5
Kitchener Digested	617	19.8	3	<2	<2	11.6
Ottawa Digested	298	83.7	11	2	<2	7.2
Waterloo Digested	518	146	38	4.1	5.9	8.2
Windsor Digested	203	307	127	34.7	139	13
Toronto (Ashbridges Bay) Digested	450	36.8	4.7	1.5	<2	12.8
Toronto (Humber) Digested	495	53.2	16.8	4.6	25.1	12.3
Toronto (North) Digested	233	28	2.2	<2	<2	6.5
Granby Digested	18.3	46.8	64.8	8.1	7.7	1.3
Moncton Digested	4.6	29.8	17.8	10.9	55.7	0.8
Truro Digested	18.3	30.4	68	15.8	9.9	2.1
Median raw	91.5	61.9	35.15	13.8	43.5	4.75
Median digested	413	83.7	24.8	6.9	11.3	10.4

Table 16. Supplementary Canadian Biosolids Concentration Data for APEs

Sludge Source	Sludge Type	Parameter	Concentrations (μg/g TS)				Reference
			4-nonyl phenol (NP)	NP1EOs and NP2EOs	Nonylphenol (1-17) ethoxylates[a] NP(4-17)EO[b]	4-tert-octyl phenol	
Toronto sewage sludge	raw sludge	range	15-311	not anal.	57-497[a]	0.8-14	Webber and Sidwha (2005)
		mean	55	not anal.	187[a]	3.7	
	digested sludge	range	85-514	not anal.	18-342[a]	1.9-13	
		mean	244	not anal.	83[a]	6.7	
Guelph Sludge (stabilized)	not specified		450	not anal.	325[b]	not anal.	XCG (2007)
Canadian sludges (1995-1998)	Raw and digested sludges	Range	4.6 – 1,230	2.1 – 467	1.5 – 43[b]	not anal.	
		Mean	305	58	12[b]	not anal.	
		Median	232	18	9[b]	not anal.	

Surveys of nonylphenol in sludges from other countries have been summarized in other literature reviews. Concentrations of NP for many types of sludges fall in the range of 500 to 2,500 μg/g TS, with the highest maximum value of 7,214 reported from Sweden although minimum levels may be as low as approximately 25 μg/g TS (Table 17). One very low value of 0.0195 μg/g TS for Norwegian final sludge was reported by Soares et al. (2008). The data in the table appear to indicate there may be differences in concentrations of 4-NP in biosolids samples, with some countries such as Italy (Soares et al., 2008), Denmark (Jayangi 2007) and France (Ghanem et al., 2007) having lower concentration ranges than other countries. Changes in formulation of household laundry detergents may be responsible; Ahel et al. (2000) reported that effluent concentrations of NPEs declined after Switzerland imposed a ban on use of NPEs in laundry detergents. Data provided by Tan et al. (2007) for return activated sludge indicated the concentration of 4-NP in this solids stream was low in comparison to other sludges.

Ruel et al. (2008) reported concentrations of total polyethoxylated nonylphenols in French biosolids as 44 μg/g TS, but noted analytical problems with the sludge matrix. These difficulties were reflected in the high standard deviation value of 970 μg/g TS (Table 18). In the same sampling survey, the mean value of total octylphenols was only 2.6 μg/g TS. Low concentrations of the mono- and di-ethoxylated forms of nonylphenol (maximum values of 41 and 25 μg/g TS, respectively) were found by Stasinakis et al. (2008). The literature survey of Harrison et al. (2006) however, indicated concentrations of alkylphenol ethoxylates were observed at up to 7,214 μg/g TS. Very low concentrations of 4-tert-octylphenol and 4-cumylphenol (less than 0.05 μg/g TS each) were documented by Tan et al. (2007) in return activated sludge samples.

Table 17. Concentrations of Nonylphenol in Sludges from Other Countries

Country	Sludge Treatment	4-nonylphenol (μg/g TS)	Reference
Switzerland	Anaerobic Digestion	450-2530	Soares et al. (2008)
	Aerobic Digestion	120-650	
	Final sludge	540-1000	
Germany	Aerobic Digestion	80-500	
	Unidentified	128.2	
USA	Anaerobic Digestion	754	
	Heat treatment	496	
	Lime treatment	470	
	Composting	64	
Italy	Raw Sludge before anaerobic digestion	242	
	Sludge after anaerobic digestion	308	
Norway	Final sludge	0.0195	
Norway	Biosolids	25-2298	Jayangi (2007)
Sweden	Biosolids	23-7214	
Denmark	Biosolids	0.3-537	
Africa	Biosolids	0.15-2790	
Australia	Return Activated sludge Plant #1	0.429±0.238 [a]	Tan et al. (2007)
	Return Activated sludge Plant #2	0.0205±0.005	
France	Mixed sludge sources (anaerobically digested, composted, limed or dried) in different conventional secondary or tertiary plants	132 ± 730	Ruel et al. (2008)
U.S.A.	Aerobically digested biosolids	nd-180	Xia et al. (2005)
	Anaerobically digested biosolids	300-1300	
Greece	Dewatered anaerobically digested or dewatered secondary sludge	<0.04 -0.45	Stasinakis et al. (2008)
France	Unknown/not specified prior to pelletization	16.5 – 125	Ghanem et al. (2007)
	Unknown/not specified prior to composting	75.6-173	
	Unknown/not specified prior to lime treatment (Plant 2)	49.6-136	
	Unknown/not specified prior to lime treatment (Plant 3)	89.8-217	
U.S.A.	Digested sludge (aerobic and anaerobic)	13-898	Heidler and Halden (2008)
Mexico and UK	Anaerobically digested sludge before composting	114	Gibson et al. (2007)

nd=not detected. [a] mean ± standard deviation

Table 18. Concentrations of Nonylphenol Ethoxylates and Other Alkylphenol in Sludges

Biosolids Type	Concentration (μg/g TS)						Reference
	Nonylphenol monoethoxylate (NP1EO)	Nonylphenol diethoxylate (NP2EO)	Nonylphenol (1-17) ethoxylates NP(1-17)EO	Alkylphenol carboxylates	4-tert-octylphenol	4-Cumylphenol	
dewatered anaerobically digested or dewatered secondary sludge	1.01 - 41.3	<0.96 - 24.7					Stasinakis et al. (2008)
mixed sludge sources (anaerobically digested, composted, limed or dried) in different conventional secondary or tertiary treatment plants			44 ± 970 [a]		2.6 ± 4 (as Octyl phenols)		Ruel et al. (2008)
Unknown/not specified			nd–7214	10–14			Harrison et al. (2006)
Return activated sludge Plant #1					0.035±0.0029 [a]	0.0015±0.0013	Tan et al. (2007)
Return activated sludge Plant #2					0.0056±0.0024 [a]	BDL	

nd=not detected; BDL=below detection limit.
[a] mean ± standard deviation

3.2.3 Effect of Sludge Treatment Processes on Occurrence of Alkylphenols and Ethoxylates

A number of published studies have compared concentrations of the alkylphenols resulting from different types of sludge treatment processes (Table 19). The different studies reviewed here have been grouped by the biosolids treatment process.

In Kinney (2006), concentrations are expressed μg/g OC, rather than the more common units of μg/g total solids (TS). Assuming that carbon in the solids is organic carbon, and that C is approximately 50 % of activated sludge biomass (Metcalf & Eddy, 1991), then concentrations expressed on an organic carbon basis would be approximately halved if expressed on a total solids basis. Concentrations of 4-nonylphenol are substantially higher following anaerobic digestion than after other forms of treatment (e.g., Kinney et al., 2006). Giger et al. (1984) were among the early researchers to propose that the hydrophobic NP1EO and NP2EO compounds associated with wastewater solids were degraded biologically to NP in anaerobic digestion. Studies by Hale and LaGuardia (2002) and LaGuardia et al. (2004) indicate that composting and drying result in lower levels of nonylphenol and ethoxylates, but that concentrations following lime treatment may not be much different than after anaerobic digestion. Composting appeared to result in the most consistent low concentrations of the tested alkylphenols and surfactants, presumably due to a prolonged retention time for aerobic biodegradation at elevated temperatures during the curing period.

3.2.4 Removal Efficiencies of Biosolids Treatment Processes

Fewer studies actually reported removal efficiencies of APs and their ethoxylates. The effect of different durations of the composting period on the reduction of 4-nonylphenol has been studied by Xia et al. (2005) and Das and Xia (2007). Their results, summarised in Table 20, indicate that a composting period of between 40 and 70 days is needed to reduce the starting level of 4-NP by over 90%. A higher proportion of wood shavings mixed with the biosolids resulted in a faster rate of reduction of the 4-NP.

In Table 21, Gibson et al. (2007) found that composting resulted in a higher removal efficiency of 4-NP than did heat drying, in general agreement with most other observed data in Table 19. Conversely, Ghanem et al. (2007) noted only an 18% reduction in 4-NP, compared to removal efficiency of 72% for drying by pelletization. Removals by lime treatment were variable.

Table 19. Concentrations of Alkylphenols and their Ethoxylates after Biosolids Treatment

Biosolids Treatment	Concentration (μg/g TS unless otherwise specified)					Reference
	4-nonylphenol (NP)	Nonylphenol mono-ethoxylate (NP1EO)	Nonylphenol diethoxylate (NP2EO)	Total of nonylphenols (NPs), NP1EOs and NP2EOs	Octylphenols (OP)	
Anaerobically digested WAS	1,520	79.4	0.793		3.50 μg/g OC	Kinney et al. (2006)
anaerobically digested biosolids				758-981		Hale and LaGuardia (2002)
Anaerobically digested biosolids-A	683	28.4	<1.5		9.9	LaGuardia et al. (2004)
Anaerobically digested biosolids-B	720	25.7	<1.5		12.6	
Anaerobically digested biosolids-C	779	102	32.6		11	
Anaerobically digested biosolids-D	701	55.8	<1.5		11.7	
Anaerobically digested biosolids-E	887	64.9	22.7		6.7	
Fresh biosolids (anaerobic)	300-1300					Xia et al. (2005)
Anaerobically digested sludge STP 1	1840 ± 68 [a]					Pryor et al. (2002)
Anaerobically digested sludge STP 2	1790 ± 68					
Anaerobically digested sludge STP 3	1480 ± 38					
Anaerobically digested sludge STP 4	1240 ± 161					
Anaerobically digested sludge STP 5	1130 ± 188					
anaerobically digested biosolids	900					Brown et al. (in press)
composted WAS	2.18-491 μg/g OC	3.96-17.2 μg/g OC	2.85-7.01 μg/g OC		0.90-4.21 μg/g OC	Kinney et al. (2006)
composted biosolids				6.1-176		Hale and LaGuardia (2002)
Composted biosolids-A	5.4	0.7	<1.5		<0.5	LaGuardia et al. (2004)

Table 19 (cont'd)

Biosolids Treatment	Concentration μg/g TS unless otherwise specified					Reference
	4-nonylphenol (NP)	Nonylphenol mono-ethoxylate (NP1EO)	Nonylphenol diethoxylate (NP2EO)	Total of nonylphenols (NPs), NP1EOs and NP2EOs	Octylphenols (OP)	
Composted biosolids-B	172	2.5	<1.5		1.5	LaGuardia et al. (2004)
Composted biosolids-C	14.2	<0.5	<1.5		<0.5	
Anaerobically digested sludge after composting	14					Gibson et al. (2007)
Composted biosolids	nd-120					Xia et al. (2005)
Unknown/not specified, after composting	119.9 ± 14.4					Ghanem et al. (2007)
air dried WAS	229 μg/g OC	23.9 μg/g OC	44.6 μg/g OC		2.71 μg/g OC	Kinney et al. (2006)
Unknown/not specified, after drying to pellets	17.3 ± 6.2					Ghanem et al. (2007)
Heat dried WAS	261 μg/g OC	44.6 μg/g OC	89.0 μg/g OC		0.414 μg/g OC	Kinney et al. (2006)
Heat dried biosolids				nd-544		Hale and LaGuardia (2002)
Heat dried biosolids-A	496	33.5	7.4		7.5	LaGuardia et al. (2004)
Anaerobically digested sludge after heat drying	212					Gibson et al. (2007)
Limed biosolids				529-932		Hale and LaGuardia (2002)
Limed biosolids-A	820	81.7	25.3		5.3	LaGuardia et al. (2004)
Limed biosolids-B	119	154	254		2	
Unknown/not specified after lime treatment (Plant 2)	62.5 ± 14.1					Ghanem et al. (2007)
Unknown/not specified after lime treatment (Plant 3)	130 ± 35.8					

nd=not detected [a] mean ± standard deviation

Table 20. Removal Efficiencies of 4-NP by Biosolids Composting

Biosolids:Wood shaving ratio	% Reduction of 4-NP after Time of Composting in Days			
	8	20	40	70
43:57	76%	80%	90%	no data
65:35	22%	70%	78%	92%
84:16	7%	60%	70%	92%
Reference	Xia et al. (2005)	Das and Xia (2007)		

Table 21. Removal Efficiencies of APs and Ethoxylates by Biosolids Treatment Processes

Constituent	Sludge Type	Removal Efficiency %	Reference	Country
4-nonylphenol	Unknown/not specified, after drying to pellets	72	Ghanem et al. (2007)	France
	Unknown/not specified, after composting	18		
	Unknown/not specified after lime treatment (Plant 2)	31		
	Unknown/not specified after lime treatment (Plant 3)	19		
	Composted anaerobically digested sludge	88	Gibson et al. (2007)	Mexico and UK
	Heat dried anaerobically digested sludge	39		
AP Polyethoxylates	Aerobic/Anaerobic	40-100	Lindberg (2005)	Sweden

3.2.5 Section Summary

The important points from this section follow.

1. There appear to be differences in APE and AP concentrations between biosolids samples collected from different countries, possibly due to different regulations for detergent product formulation.
2. In raw sludges or aerobically digested sludges, concentrations of mono- and di-ethoxylated species of NP may occur at concentrations approximately equal to or even slightly greater than the parent nonylphenol.
3. Of the biosolids treatment processes examined, anaerobic digestion consistently has the highest concentrations of 4-NP, as a result of anaerobic biotransformation processes converting mono- and di-ethoxylated species to the parent compound.
4. Composting appears to result in lower concentrations of APs and APEs than does drying or lime treatment. Limited data suggest that lime treatment may result in lower reduction of APEs compared to composting or drying processes.
5. A composting period of between 40 and 70 days is needed to reduce the starting level of 4-NP by over 90%.

3.3 BROMINATED FLAME RETARDANTS

3.3.1 Introduction

Polybrominated diphenyl ethers (PBDEs) are compounds used as flame retardants in a wide variety of applications. They have been historically sold as commercial mixtures having a predominant homolog class (compounds with the same number of bromine substituents located at different locations on the diphenyl ether structure. The main commercial classes of the PBDEs sometimes referred to generically as brominated flame retardants (BFRs) are the pentabromo-, octabromo- and decabromo diphenyl ethers (US DHHS, 2004). PBDEs cannot be manufactured or used in Canada, and use of the pentabromo- and octabromo- diphenyl ethers is prohibited in Canada (Canada Gazette, 2008). Only the decabromo diphenyl ether (DPE) product is allowed for use in Canada.

PBDEs are added to plastics to reduce flammability and fire damage; products incorporating these retardants are used in domestic, commercial and industrial settings, and include polyurethane furniture foam, carpets, high impact cases, circuit boards, appliances and electrical equipment (USGS, 2004). As the products age, the PBDEs can dissociate from the host plastic to become part of indoor dust. Cleaning by wet mopping of floors and washing of dusting cloths or floor-mats is therefore a probable source of entry to wastewater treatment facilities. At wastewater treatment facilities, the PBDEs, because of high octanol:water partition coefficients, are expected to sorb strongly to wastewater solids, and thus end up mainly in the residual wastewater solids.

The environmental and health concerns with PBDEs centre on their persistence, potential toxicity and ability to bioaccumulate. Elevated concentrations of the compounds have been found in human breast milk, particularly in North America (USGS, 2004), and in Arctic mammals near the top of the food chain (ringed seals and beluga whales) (Environment Canada, 2005). In humans, these compounds can disrupt thyroid hormone activity due to the similarity of PBDE metabolites to the hormone thyroxin, and may impair neurodevelopment (USGS, 2004).

3.3.2 Occurrence Data

Among the most detailed concentration data of PBDEs in Canadian wastewater solids is that of Rayne and Ikonomu (2005) for the Kelowna, BC wastewater treatment plant. Isomer concentrations were documented for primary sludge, anaerobically fermented primary sludge, return activated sludge dissolved air flotation float solids, and dewatered centrifuge cake. The data are produced in Table 22, converted to a dry solids basis for comparison with other literature concentration data. The isomer decabromo DPE (BDE 209) was observed in all the samples at the highest concentration of any of the isomers, followed by the penta BDE99 and tetra BDE47 isomers. The data indicate that the concentrations of the PBDE isomers are substantially lower in the solids streams prior to secondary treatment (primary sludge before and after anaerobic fermentation), and more concentrated in the solids streams following secondary treatment (i.e, return activated sludge, dissolve air flotation (DAF) float and centrifuge cake).

Table 22. PBDE Concentrations in Kelowna, BC Wastewater Solids Streams (from Rayne and Ikonomou, 2005)

BDPE Isomer	Concentration (ng/g TS)				
	Primary sludge	Fermented Primary sludge	Return Activated Sludge	DAF sludge	Biosolids
2,4-Dibromodiphenyl Ether (di BDE7)	0.0070	0.0059	0.0620	0.0898	0.0806
2,4' + 3,3'- Dibromodiphenyl Ether (di BDE8/11)	0.0070	0.0151	0.0728	0.0820	0.0764
3,4'- Dibromodiphenyl Ether (di BDE13)	0.0070	0.0189	0.0661	0.0980	0.0753
4,4'- Dibromodiphenyl Ether (di BDE15)	0.0651	0.0551	0.477	0.408	0.348
2,2',4- Tribromodiphenyl Ether (tri BDE-17)	0.379	0.343	5.70	8.57	4.88
2,3',4- Tribromodiphenyl Ether (tri BDE25	0.0279	0.0415	0.326	0.422	0.279
2,4,4'-Tribromodiphenyl Ether (tri BDE 28)	1.15	1.05	8.71	10.74	7.67
2,2',4,4'-Tetrabromodiphenyl Ether (tetra BDE47)	58.46	57.97	469.50	553.22	401.95
2,2',4,5'-Tetrabromodiphenyl Ether (tetra BDE49)	1.67	1.67	13.80	17.10	11.82
2,3',4,4'-Tetrabromodiphenyl Ether (tetra BDE66)	1.33	1.35	11.21	12.63	9.17
2,3',4',6-Tetrabromodiphenyl Ether (tetra BDE-71)	0.160	0.132	1.72	3.76	1.47
3,3',4,4'-Tetrabromodiphenyl Ether (tetra BDE77)	0.0070	0.0164	0.0459	0.0258	0.106
2,2',4,4',5-Pentabromodiphenyl Ether (penta BDE85)	3.19	2.44	25.47	30.53	22.16
2,2',4,4',5-Pentabromodiphenyl Ether (penta BDE99)	71.81	61.70	603.46	738.35	523.98
2,2',4,4',6-Pentabromodiphenyl Ether (penta BDE100)	10.65	10.67	87.50	111.50	79.64
2,3,3',4,4'-Pentabromodiphenyl Ether (penta BDE105)	0.0139	0.0065	0.129	0.0426	0.118
2,3',4,4',6-Pentabromodiphenyl Ether(penta BDE119)	0.0488	0.0474	0.503	0.608	0.437
3,3',4,4',5-Pentabromodiphenyl Ether (penta BDE126)	0.0581	0.0384	0.167	0.0426	0.192
2,2',3,4,4',5'-Hexabromodiphenyl Ether (hexa BDE 138)	0.990	0.842	8.04	8.63	6.73
2,2',3,4,4',6'-Hexabromodiphenyl Ether (hexa BDE140)	0.204	0.206	1.99	2.19	1.67
2,2',4,4',5,5'-Hexabromodiphenyl Ether (hexa BDE 153)	10.89	8.90	86.29	98.47	78.06
2,2',4,4',5,6'-Hexabromodiphenyl Ether (hexa BDE 154)	5.61	4.99	51.18	57.36	43.89
2,2',4,4',6,6'-Hexabromodiphenyl Ether (hexa BDE155)	0.265	0.228	2.08	2.54	1.82
2,2',3,4,4',5,6-Heptabromodiphenyl Ether (hepta BDE181)	0.0163	0.0242	0.0796	0.0949	0.1214
2,2',3,4,4',5',6-Heptabromodiphenyl Ether (hepta BDE 183)	2.41	1.35	9.43	10.72	8.16
2,3,3',4,4',5,6-heptabromodiphenyl ether (BDE-190)	0.100	0.080	0.442	0.512	0.480
2,2',3,3',4,4',5,5',6-Nonabromodiphenyl Ether (nona BDE206)	2.04	2.31	11.91	14.93	11.06
2,2',3,3',4,4',5,6,6'-Nonabromodiphenyl Ether (nona BDE207)	2.17	2.99	14.64	18.94	14.44
2,2',3,3',4,5,5',6,6'-Nonabromodiphenyl Ether (nona BDE208)	0.251	0.407	2.03	2.30	1.93
2,2',3,3',4,4',5,5',6,6'-Decabromodephenyl Ether (deca BDE209)	122.44	146.15	553.46	691.20	558.66
TOTAL PBDEs	300.10	310.65	1989.45	2421.57	1809.15

Data for PBDEs in wastewater solids from the Windsor Little River treatment plant are provided by Song et al. (2006). The concentrations of eight major isomers monitored in the study are summarized in Table 23.

Table 23. Concentrations of Major PBDE Isomers in Windsor Little River WW Solids (Song et al., 2006)

PBDE Isomer	Concentration, ng/g TS mean ± std. dev.	
	Primary sludge	Waste activated sludge
2,4,4'-Tribromodiphenyl Ether (tri BDE 28)	8.0 ± 3.1[a]	14 ± 6
2,2',4,4'-Tetrabromodiphenyl Ether (tetra BDE47)	586 ± 207	963 ± 415
2,2',4,4',5-Pentabromodiphenyl Ether (penta BDE99)	757 ± 272	1247 ± 516
2,2',4,4',6-Pentabromodiphenyl Ether (penta BDE100)	122 ± 42	167 ± 129
2,2',3,4,4',5'-Hexabromodiphenyl Ether (hexa BDE 138)	9.1 ± 4.3	17 ± 6
2,2',4,4',5,5'-Hexabromodiphenyl Ether (hexa BDE 153)	84 ± 27	109 ± 82
2,2',4,4',5,6'-Hexabromodiphenyl Ether (hexa BDE 154)	49 ± 19	71 ± 54
2,2',3,4,4',5',6-Heptabromodiphenyl Ether (hepta BDE 183)	12 ± 6	22 ± 10

[a] mean ± standard deviation

Decabromo (BDE209) was not monitored in this study. The two isomers detected at the highest concentrations in both sludge types were the penta BDE99 and the tetra BDE47. As was observed with the data from Rayne and Ikonomou (2005), the WAS sample exhibited higher concentrations of the monitored isomers than did the primary sludge.

One of the more comprehensive studies on PBDEs in municipal biosolids was completed in Australia (Clarke et al., 2008), comparing concentrations in eight urban and eight rural biosolids samples. Table 24 presents the concentration data for the different PBDE isomers included in the analytical method for the urban wastewater treatment plants sampled. Decabromo DPE (BDE209) was present at the highest concentration (mean 881 ng/g TS). It is of interest that three of the four plants identified as having anaerobic digestion as part of the treatment have concentration of decabromo DPE, and indeed many of the isomers, higher than the mean value. Because of the limited sample size, however, it is not clear whether this represents an actual phenomenon that occurs with anaerobic digestion, or whether it is an artifact of the limited sample size. The apparent higher concentration of the decabromo DPE in the anaerobically digested sludge samples is contrary to expected dehalogenation reaction which occurs under anaerobic conditions. For example, Shin et al. (2006) observed a decline of decabromo DPE (BDE209) under batch anaerobic test at both 35 and 55 ^{o}C, with concomitant increases in lower brominated congeners. Because the concentrations of many of the lower brominated isomers in anaerobically digested sludges are higher than the mean value of urban biosolids samples, it is likely the observation is due to the randomness of the samples rather than due to higher levels attributed to the anaerobic digestion process.

Results for the eight rural samples in the Australian survey are provided in Table 25. Only one of the rural plants utilized anaerobic digestion as a biosolids treatment process, while most relied instead on dewatering and drying of the biosolids. Decabromo DPE was present in all the

Table 24. PBDE Concentrations in Biosolids from Australian Urban Municipalities (Clarke et al., 2008)

	Concentration of PBDE congeners (ng/g TS) by designated plant and sludge treatment								
	U1	U5	U6	U8	U2	U3	U4	U7	
BDPE Isomer	An Dig + Dewater	An Dig + Dewater	An Dig + Dewater	An Dig + Dewater	Dewater	DAF filtration	IFAS	Dewater + Lime	Mean ± Std Dev.
2,2',4-Tribromo DPE (BDE17)	0.96	0.16	7.75	1.85	2.7	2.7	0.27	0.46	2.1 ± 2.5
2,4,4'-Tribromo DPE (BDE28) + (BDE33)	2	<0.2	4.55	5.2	3.1	25	1.1	0.85	6±8.6
2,2',4,4'-Tetrabromo DPE (tetra BDE47)	120	17	205	285	180	36	72	45	120±95
2,2',4,5'-Tetrabromo DPE (BDE49)	3.8	1.9	7.95	8.45	5.6	2.3	2.3	1.5	4.2±2.8
2,3',4,4'-Tetrabromo DPE (BDE66)	3.3	0.59	7.15	7.7	6.1	1.4	2.9	1.5	3.8±2.8
3,3',4,4'-Tetrabromo DPE (BDE77)	0.049	<0.004	0.58	0.092	0.055	0.0099	<0.01	<0.03	0.2±0.2
2,2',4,4',5-Pentabromo DPE (BDE85)	4.8	1	8.8	11.5	6.7	1.1	3.1	1.8	4.9±3.9
2,2',4,4',5-Pentabromo DPE (BDE99)	130	22	230	315	190	31	84	48	131±106
2,2',4,4',6-Pentabromo DPE (BDE100)	26	4.4	47.5	63.5	39	8.6	16	9.6	27±21.0
2,3',4,4',6-Pentabromo DPE (BDE119)	<0.9	0.04	0.695	0.465	<1	<0.1	<0.4	0.11	0.33±0.31
2,2',3,4,4',5'-Hexabromo DPE (BDE138)			3.3	2.7				1.9	2.6±0.7
2,2',3,4,4',6-Hexabromo DPE (BDE139)	1.5	0.31	2.8	3.15	2	0.49	0.82	0.42	1.4±1.1
2,2',3,4,4',6'-Hexabromo DPE (BDE140)	0.45	0.16	1.27	0.84	0.71	0.18	0.29	0.13	0.5±0.41
2,2',4,4',5,5'-Hexabromo DPE (BDE153)	13	4.9	23	28	20	4.8	8.2	4.4	13.3±9.3
2,2',4,4',5,6'-Hexabromo DPE (BDE154)	10	3.2	19.5	24.5	16	4.3	6.1	3.9	10.9±8.1
2,2',3,3',4,4',6-Heptabromo DPE (BDE171)	<0.09	0.41	3.87	0.375	<0.2	0.097	<0.4	0.099	0.97±1.63
2,2',3,4,4',5,5'-Heptabromo DPE (BDE180)	0.37	0.81	3.95	0.615	1.7	0.14	0.29	0.11	1±1.3
2,2',3,4,4',5',6-Heptabromo DPE (BDE183)	9.6	15	13	10	19	3.9	5.1	1.9	9.7±5.9
2,2',3,4,4',6,6'-Heptabromo DPE (BDE184)	0.16	0.2	2.23	0.41	0.39	0.094	0.11	0.064	0.46±0.73
2,2',3,3',4,4',5',6-Octabromo DPE (BDE196)	4.7	<2	7.4	4.2	7.7	<0.2	<1	1.6	5.1±2.5
2,2',3,3',4,4',6,6'-Octabromo DPE (BDE197)	2.9	8.4	8.75	4.3	3.6	0.89	1.1	0.85	3.8±3.2
2,2',3,3',4,4',5,5'-Octabromo DPE (BDE201)	1.1	14	4.85	1.3	<4	<1	<0.7	0.38	4.3±5.7
2,2',3,4,4',5,5',6-Octabromo DPE (BDE203)	<3	40	8.35	5.1	<3	<1	<2	1.3	13.7±17.8
2,2',3,3',4,4',5,5',6-Nonabromo DPE (BDE206)	32	98	30	27.5	9.7	3.1	4.5	6	26±31
2,2',3,3',4,4',5,6,6'-Nonabromo DPE (BDE207)	13	110	19.5	12.5	12	5.7	6	6.3	23±35
2,2',3,3',4,5,5',6,6'-Nonabromo DPE (BDE208)	7.9	97	15.7	7.95	6.5	2.7	2.8	3.7	18±32
2,2',3,3',4,4',5,5',6,6'-Decabromo DPE (BDE209)	1170	3780	530	910	360	93	81	130	880±1200

An Dig = Anaerobic Digestion DAF=dissolved air flotation IFAS= integrated fixed-film activated sludge

Table 25. PBDE Concentrations in Biosolids from Australian Rural Municipalities (Clarke et al., 2008)

BDPE Isomer	Concentration of PBDE congeners (ng/g TS) by designated plant and sludge treatment								
	R1	R2	R3	R4	R5	R6	R7	R8	
	Dewater	Dewater	Dewater	An Dig + Dewater	Lime	Dewater + Lime	Land Dried	Solar Dried	Mean ± Std Dev.
2,2',4-Tribromo DPE (BDE17)	4.3	0.25	12	2.6	0.69	0.4	0.0065	3.6	3.2 ± 4.5
2,4,4'-Tribromo DPE (BDE28) + (BDE33)	8.1	0.92	2.6	2.4	1.2	1.4	<0.06	11	3.7± 4.1
2,2',4,4'-Tetrabromo DPE (tetra BDE47)	170	74	120	140	56	89	<0.4	410	160 ± 140
2,2',4,5'-Tetrabromo DPE (BDE49)	16	1.9	6.4	5.6	2	3.1	0.035	23	6.7 ±8.3
2,3',4,4'-Tetrabromo DPE (BDE66)	8.4	1.9	4.2	4.8	1.7	2.8	0.017	14	4.6 ±4.9
2,3',4',6-Tetrabromo DPE (BDE71)	1.6	0.17	8	1.9	<4	0.18	<0.009	1.4	2.9 ±3.5
3,3',4,4'-Tetrabromo DPE (BDE77)	0.1	0.027	0.06	0.069	<0.03	0.06	<0.004	0.16	0.09 ±0.05
2,2',4,4',5-Pentabromo DPE (BDE85)	5.1	5.8	3.9	5.8	1.8	4.2	0.013	14	5 ±4.9
2,2',4,4',5-Pentabromo DPE (BDE99)	210	120	130	170	51	130	0.37	400	150 ±140
2,2',4,4',6-Pentabromo DPE (BDE100)	41	21	24	32	11	21	<0.08	94	36 ± 33
2,3',4,4',6-Pentabromo DPE (BDE119)	0.28	0.14	0.28	0.29	<0.6	0.21	<0.002	0.68	0.37 ±0.21
2,2',3,4,4',5'-Hexabromo DPE (BDE138)	4.2	4.7	3.9	6.1	nd	4.2		11	6.3 ±3.3
2,2',3,4,4',6-Hexabromo DPE (BDE139)	1.6	1.9	1.1	1.5	0.4	1.3	<0.002	3.9	1.6 ±1.3
2,2',3,4,4',6'-Hexabromo DPE (BDE140)	0.61	0.54	0.47	0.59	0.16	0.36	<0.01	1.1	0.54±0.35
2,2',4,4',5,5'-Hexabromo DPE (BDE153)	23	14	13	17	4.6	13	0.064	35	13.8±12.1
2,2',4,4',5,6'-Hexabromo DPE (BDE154)	19	9.8	12	15	3.8	8.4	0.04	33	12±11.6
2,2',3,3',4,4',6-Heptabromo DPE (BDE171)	0.38	0.11	0.17	0.27	0.13	0.2	<0.009	0.47	0.25±0.13
2,2',3,4,4',5,5'-Heptabromo DPE (BDE180)	0.57	0.17	0.26	0.41	0.16	0.33	<0.003	0.67	0.37±0.19
2,2',3,4,4',5',6-Heptabromo DPE (BDE183)	13	3.3	3.7	11	3.3	7.3	0.083	11	6.1±4.5
2,2',3,4,4',6,6'-Heptabromo DPE (BDE184)	0.67	0.098	0.2	0.47	0.075	0.19	<0.002	0.38	0.26±0.16
2,3,3',4,4',5',6-heptabromo DPE (BDE191)	0.2	0.047	0.14	0.092	0.053	0.082	<0.005	0.22	0.12±0.07
2,2',3,3',4,4',5',6-Octabromo DPE (BDE196)	6.4	2.2	4.7	4.2	3	4.3	<0.3	6.5	4.5±1.3
2,2',3,3',4,4',6,6'-Octabromo DPE (BDE197)	6.6	1.4	2.2	5.4	1.5	3	0.022	4.3	2.7±1.9
2,2',3,3',4,4',5,5'-Octabromo DPE (BDE201)	2.8	0.44	1.8	1.2	0.59	1	0.015	2.7	1.2±0.9
2,2',3,4,4',5,5',6-Octabromo DPE (BDE203)	7.8	2.4	5.7	4.5	2.3	3.7	<0.03	8.7	5±2.4
2,2',3,3',4,4',5,5',6-Nonabromo DPE (BDE206)	28	7.6	31	8.5	8.2	7.9	0.093	31	14±13
2,2',3,3',4,4',5,6,6'-Nonabromo DPE (BDE207)	21	5.9	20	8.7	7.4	9.9	0.094	19	11±7.5
2,2',3,3',4,5,5',6,6'-Nonabromo DPE (BDE208)	14	3.4	10	4.4	3.9	5.7	0.064	14	6.3±4.9
2,2',3,3',4,4',5,5',6,6'-Decabromo DPE (BDE209)	990	280	1210	260	250	180	3.4	1050	490±510

rural biosolids samples at the highest concentration of any isomer, as was the case with the urban biosolids samples. The sample derived for solar drying had the highest concentrations of almost all isomers in the rural biosolids samples. There are insufficient data to determine whether it is the sludge itself or the treatment method that results in the high concentrations relative to the other rural samples.

Based on statistical analysis, Clarke et al. (2008) concluded that the sum of the PBDE isomers were not statistically different between the urban and rural biosolids. The authors further concluded that the lower brominated isomers, representative of the Penta-DPE commercial formula were consistent between urban and rural biosolids, and thus were likely of domestic sources. Greater variability between the concentrations of decabromo DPE in urban and rural biosolids was observed however, suggesting to the authors that industrial sources were likely the main contributors to the presence of decabromo DPE in biosolids.

The recently published US EPA's Targeted National Sewage Sludge Survey documented concentrations of many target analytes including PBDEs. Biosolids from a total of 74 municipal treatment plants in 35 states were included in this comprehensive national survey. The data were statistically analyzed to determine median, mean and standard deviations for the target contaminants. The results for the PBDEs are presented in Table 26. The sludges represent a wide range of process types, geographic locations and treatment plant capacities, although all facilities tested had a treatment capacity greater than 3780 m^3/d (1 MGD) with a minimum of secondary treatment (US EPA 2009).

Table 26. PBDE Concentrations in Sludges and Biosolids Based on U.S. EPA's Targeted National Sewage Sludge Survey (US EPA 2009)

PBDE Isomer	Concentration (ng/g TS)		
	median	mean	std dev
2,4,4'-Tribromodiphenyl Ether (tri BDE28) + tri BDE33	8.90	15.35	24.07
2,2',4,4'-Tetrabromodiphenyl Ether (tetra BDE47)	570.4	709.2	523.8
2,3',4,4'-Tetrabromodiphenyl Ether (tetra BDE66)	12.00	17.40	18.55
2,2',4,4',5-Pentabromodiphenyl Ether (penta BDE85)	23.00	27.94	22.00
2,2',4,4',5-Pentabromodiphenyl Ether (penta BDE99)	574.6	716.4	533.4
2,2',4,4',6-Pentabromodiphenyl Ether (penta BDE100)	120.0	150.4	143.8
2,2',3,4,4',5'-Hexabromodiphenyl Ether (hexa BDE138)	7.00	10.75	12.63
2,2',4,4',5,5'-Hexabromodiphenyl Ether (hexa BDE153)	54.12	68.33	52.69
2,2',4,4',5,6'-Hexabromodiphenyl Ether (hexa BDE154)	46.50	59.90	57.92
2,2',3,4,4',5',6-Heptabromodiphenyl Ether (hepta BDE183)	10.00	16.66	20.47
2,2',3,3',4,4',5,5',6,6'-Decabromodephenyl Ether (deca BDE209)	1,163	2,181	3,463

The data show that decabromo BDE209 is present at the highest concentration in the sludges tested, almost twice the concentration of the isomers with next highest concentrations, the penta BDE99 and tetra BDE47.

Occurrence data retrieved from the literature for other countries are summarized in Table 27.

Table 27. Occurrence data for PBDEs in Biosolids Samples from Other Countries

Biosolids Source	2,2',4,4'-Tetrabromo diphenyl Ether (BDE47)	2,2',4,4',5-Pentabromo diphenyl Ether (BDE99)	2,2',4,4',6-Pentabromo diphenyl Ether (BDE100)	2,2',4,4',5,5'-Hexabromo diphenyl Ether (BDE 153)	2,2',4,4',5,6'-Hexabromo diphenyl Ether (BDE 154)	2,2',3,4,4',5',6-Heptabromo diphenyl Ether (BDE 183)	2,2',3,3',4,4',5,5',6,6'-Decabromo diphenyl Ether (deca BDE209)	Reference
Palo Alto STP CA, U.S.	722-778	894-973	158-172	83-91	61-72		not anal.	from Song et al. (2006)
11 U.S. sites	359-754	931-1157	89-255	56-199	58-172	85-4890		from Gevao et al. (2008)
European STPs	15-91	19-120	3.5-28	1.0-15.5	0.7-14.8		not anal.	from Song et al. (2006)
Spain 5 sites	17.0-40.9 (22.9)[a]	25.0-50.9 (26.9)	5.60-11.0 (6.29)	3.31-5.70 (3.68)	2.47-4.08 (3.42)	3.66-29.6 (3.90)	80.6-1082 (393)	Eljarrat (2008)
Spain 6 sites	1.8-83.6	23.4-64.2	0.2-14	1.2-7	1.1-5.8	8.5-275		from Gevao et al. (2008)
Kuwait Treatment plant 1	0.24-2.72 (0.97)	0.61-5.96 (1.95)	0.06-0.85 (0.29)	0.04-0.62 (0.19)	0.06-1.05 (0.31)	0.04-0.78 (0.21)	4.8-157.5 (48.5)	Gevao et al. 2008
Kuwait Treatment plant 2	0.95-7.81 (4.16)	2.04-14.74 (8.4)	0.82-2.3 (1.34)	0.24-1.37 (0.82)	0.35-1.98 (1.18)	0.15-0.86 (0.44)	16.4-1595.6 (360.4)	Gevao et al. 2008
Kuwait Treatment plant 3	0.97-2.35 (1.86)	1.53-4.84 (3.8)	0.28-0.75 (0.4)	0.16-0.54 (0.40)	0.18-0.81 (0.58)	0.11-0.50 (0.33)	28.4-286.8 (136.5)	Gevao et al. 2008
Sweden 14 sites	<2-80	<2-104	<2-25	<dl-16.4	<dl-10.4	785-18032		from Gevao et al. (2008)
Stockholm, Sweden	39-91	48-120	11-28	not anal.	not anal.	not anal.		from Gevao et al. (2008)
Klippen, Sweden	22	18	5.4	not anal.	not anal.	not anal.		from Gevao et al. (2008)
Rimbo, Sweden	53	53	13	not anal.	not anal.	not anal.		from Gevao et al. (2008)
Bjergmarken, DN	96.8	86.2	19.1	7.8	6.1	248		from Gevao et al. (2008)

[a] median value in parentheses; dl = detection limit

The data indicate that the isomer concentrations are much higher in biosolids samples from the U.S. than from samples from European facilities or from the sited in Kuwait. The mean concentration data from the US EPA's sludge survey in Table 26 are similar to the U.S. concentration values in Table BDE6. Canadian concentration data presented in Tables 22 and 23 are more similar to the U.S. values, while the Australian data found in Tables 24 and 25 lie between the North American and European/Kuwaiti data.

3.3.3 Effect of Treatment Processes

LaGuardia et al. (2004) compared PBDE concentrations in four biosolids treatment processes, including composting, lime treatment, heat drying, and anaerobic digestion. The concentration data from these treatments are provided in Table 28.

Table 28. Comparison of PBDE Concentrations in Four Biosolids Treatment Processes (LaGuardia et al., 2004).

Treatment Process	2,2',4,4'-Tetrabromo diphenyl Ether (BDE47)	2,2',4,4',5-Pentabromo diphenyl Ether (BDE99)	2,2',4,4',6-Pentabromo diphenyl Ether (BDE100)	2,2',4,4',5,5'-Hexabromo diphenyl Ether (BDE153)	2,2',4,4',5,6'-Hexabromo diphenyl Ether (BDE154)	2,2',3,3',4,4',5,5',6,6'-Decabromo diphenyl Ether (BDE209)
Compost-A	498	743	106	55.6	98.8	308
Compost-B	754	1157	167	87.9	121	1460
Compost-C	536	516	112	71.8	58.2	368
Lime-A	359	513	88.5	64.3	82.6	553
Lime-B	525	584	200	179	172	84.8
Heat-A	518	714	115	58.8	95.2	1940
Heat-B	673	815	255	119	169	4890
AD-A	605	572	125	68.9	57.2	347
AD-B	421	391	113	116	61	340
AD-C	686	648	129	67.7	61.9	40
AD-D	674	613	176	80.6	74.5	389

The data suggest that for the hexabromo and lower brominated congeners, the different treatment processes had relatively little effect on the observed concentrations. The data are less clear with respect to the decabromo isomer. The sites using anaerobic digestion appeared to have substantially lower concentrations than the other treatment processes. The two sites using heat drying had biosolids with the highest decabromo DPE concentrations observed in the survey. Two of three composted samples exhibited relatively low concentrations of the isomer, as did the two limed samples. More sites and tests would be required to provide an adequate basis for statistical analysis.

A study be Gerecke et al. (2006) at a full-scale Swiss treatment plant was of interest because it compared samples of PBDEs before and after anaerobic digestion, and also investigated two other type of brominated flame retardant. Data are provided in Table 29.

All three classes of flame retardants were reduced by full-scale anaerobic digestion. The

decabromo isomer was present at much higher concentrations than the other two flame retardants. The tetrabromobisphenol A (TBBPA) class was reduced by the largest extent while the decabromo DPE was reduced by the least extent. In laboratory studies, Gerecke et al. (2006) determined compound half-lives of less than a day for the hexabromocyclododecane (HBCD) and TBBPA, compared to 700 days for the decabromo DPE. Shin et al. (2006) also found in laboratory studies with batch reactors that concentrations of decabromo DPE were reduced by anaerobic treatment, with a faster reduction occurring at thermophilic than at mesophilic temperatures. Such observations may well explain why anaerobic treatment resulted in lower concentration in biosolids than did composting, heat treatment or liming in the study by LaGuardia et al. (2004).

Table 29. Concentrations of Brominated Flame Retardants In Sludge Before and After Anaerobic Digestion (Gerecke et al., 2006)

Flame retardant	Concentration nmol/L		Concentration ng/g	
	raw	digested	raw	digested
2,2',3,3',4,4',5,5',6,6'-Decabromodephenyl Ether (BDE209)	76	49	2,490	1,605
Hexabromocyclododecane (HBCD) (6 stereoisomers)	2.1	1.2	66.32	37.89
Tetrabromobisphenol A + Tetrabromobisphenol (dimethyl) (TBBPA)	1.9	0.3	34.20	5.40

No reduction efficiency data for the BDPEs in biosolids treatment processes were observed in the literature.

3.3.4 Section Summary

The main points of interest for this section follow.

1. There are apparent differences in concentrations of PBDE isomers in North America and other countries (e.g., Europe, Kuwait, and Australia).
2. The isomer decabromo DPE (BDE 209) was observed in all the samples at the highest concentration of any of the isomers, followed by the penta BDE99 and tetra BDE47.
3. Available data indicate that the concentrations of the PBDE isomers are substantially lower in the solids streams prior to secondary treatment (e.g. primary sludge), and more concentrated in the solids streams following secondary treatment (i.e, return activated sludge, and dewater secondary or mixed sludge).
4. Anaerobic digestion may result in a reduction of decabromo DPE, but concentrations of lower brominated congeners may increase.
5. There is insufficient information to determine if other biosolids treatment processes can result in reduction of PBDEs.
6. Few occurrence data were identified for other BFRs such as HBCD and TBBPA.
7. No reduction efficiency data for the BDPEs or other brominated flame retardants in biosolids treatment processes were observed in the literature.

3.4 PHARMACEUTICAL COMPOUNDS

3.4.1 Introduction

This class of micro-constituents in sludges and biosolids includes many different sub-classes with different therapeutic uses. This report will follow to a great extent the classification used by Gielen (2007) for investigating interactions of pharmaceuticals in New Zealand sludge with soils. The classes of pharmaceuticals investigated and reported herein are provided in Table 30. By far, the most compounds identified belonged to the general class of antibiotics.

Table 30. Categories and Pharmaceuticals Identified in this Review

Antibiotics		**Nervous system**	**Analgesic**	**Blood**
sulfonamides	macrolides	anti-epileptic	Acetaminophen	anti-lipid
Sulfadimethoxine	Clarithromycin	Primidone	NSAID	Bezafibrate
Sulfamethazine	Erythromycin	Carbamazepine	Diclofenac	Clofibric Acid
Sulfamerazine	Azithromycin	anti-depressant	Naproxen	Gemfibrozil
Sulfametazine	Ormetoprim	fluoxetine	Ibuprofen	Anti-coagulant
Sulfadiazine	Virginiamycin	anti-psychotic	Indometacin	Warfarin
Sulfisoxazole	Tylosin	Chlorpromazine	Mefenamic acid	
Sulfachloro-pyridazine	Roxithromycin	Thioridazine	Ketoprofen	**Other**
Sulfanilamide	tetracyclines	anti-anxiety		anti-parasitic
Sulfadimidine	Doxycycline	Diazepam	**Alimentary**	Thiabendazole
Sulfonamide	Chlortetracycline	Amitriptyline	gastric	Carbadox
Sulfamethizole	Minocycline	Paroxetine	Cimetidine	Anti-fungal
fluoroquinolones	Oxytetracycline	stimulants	Ranitidine (hydrochloride)	Miconazole
Norfloxacin	Demeclocycline	Methamphetamine	Famotidine	Other
Enrofloxacin	beta-lactams	Amphetamine	Omeprazole	Digoxigenin
Lomefloxacin	Cloxacillin	Caffeine	Diabetic	Cotinine
Sarafloxacin	Oxacillin	1,7-Dimethylxanthine	Glibenclamide	Norgestimate
Flumequine	Penicillin G		Metformin (hydrochloride)	Salicylic Acid
Ciprofloxacin	Penicillin V	**Respiratory/allergy**		
Ofloxacin	Dicloxacillin	anti-histamine	**Cardiac**	
Clinafloxacin	bacteriostats	Diphenylhydramine	Digoxin	Atenolol
lincosamides	Triclosan	Diphenhydramine	Hydrochlorothiazide	Propranolol
Clindamycin,	Triclocarban	Loratidine	Chlorpromazine	Diltiazem
Lincomycin	Trimethoprim	anti-bronchospasm	Thioridazine	
	Chloramphenicol	Albuterol		

3.4.2 Antibiotics

Antibiotic pharmaceuticals consist of many classes of compounds applied to inhibit or kill pathogenic bacteria. The main classes of antibiotics observed in this literature review are found in Table 31.

Tetracycline Antibiotics

Data on the occurrence of tetracycline compounds in sludge and biosolids are sparse. The recent U.S. EPA Targeted National Sewage Sludge Survey (TNSSS) (EPA, 2009) provides one of the

Table 31. Classes of Antibiotics and Compounds Noted in Literature Review of Biosolids

Tetracyclines	Sulfonamides	Fluoroquinolones	Macrolides	Beta-lactams
Tetracycline	Sulfadimethoxine	Norfloxacin	Clarithromycin	Cloxacillin
4-Epitetracycline[a]	Sulfamerazine	Enrofloxacin	Erythromycin	Oxacillin
4-Epianhydrotetracycline[a]	Sulfamethazine/	Lomefloxacin	Azithromycin	Penicillin G
Anhydrotetracycline[a]	Sulfametazine	Sarafloxacin	Ormetoprim	Penicillin V
Chlortetracycline	Sulfadiazine	Flumequine	Virginiamycin	Dicloxacillin
4-Epichlortetracycline[b]	Sulfisoxazole	Ciprofloxacin	Tylosin	
4-Epianhydrochlor-tetracycline[b]	Sulfachloro-pyridazine	Ofloxacin	Roxithromycin	
Anhydrochlortetracycline[b]	Sulfanilamide	Clinafloxacin		
Isochlortetracycline[b]	Sulfadimidine			Bacteriostats
Oxytetracycline	Sulfonamide	Quinolones		Trimethoprim
4-Epioxytetracycline[c]	Sulfamethizole	Oxolinic acid	Lincosamides	Chloramphenicol
Demeclocycline			Clindamycin,	Hexachlorophene
Doxycycline		Cephalosprins	Lincomycin	Triclosan
Minocycline		Cefotaxime		Triclocarban

[a] metabolite of Tetracycline [b] metabolite of Chlortetracycline [c] metabolite of Oxytetracycline

most complete pictures of the compounds there, as indicated in Table 32. Tetracycline and its metabolite 4-epitetracycline, doxycycline and minocycline were found in the highest concentrations. Other than the EPA data, few other studies have reported concentrations.

Earlier studies by Lindberg et al. (2005) and the review by Harrison et al. (2006) indicate a high Limit of Quantitation (LOQ) for doxycycline (approx. 1200 – 1500 ng/g TS). The more recent EPA (2009) study cites a median concentration of 424 ng/g TS, suggesting a significant improvement in analytical procedures in the interim.

Wu et al. (2008) observed the reduction of concentrations of tetracycline and doxycycline over time in aerobically digested biosolids held under different storage conditions (Table 33). Concentrations of tetracycline declined so readily that the tests were terminated after 8 days. Conditions of aerobic or anaerobic storage had no effect on the reduction in tetracycline, nor did the presence or absence of daylight. The data indicated that doxycycline required a substantially longer time for reduction in concentration than did tetracycline, with aerobic conditions providing a more favourable environment for reduction of doxycycline than anaerobic conditions. Presence or absence of daylight made no appreciable difference in the reduction of doxycycline.

Table 32. Concentrations of Tetracycline Antibiotics in Sludges and Biosolids

Tetracycline Compound	Not specified (sludge survey)	Not specified (literature survey)	Unknown sludge (An urban WWTP)	Unknown sludge (A rural WWTP)	Anaerobic & aerobic digested
Tetracycline	1278 (630) [a]		<LOQ - 15.8	19.8	
4-ETC	1135 (620)				
4-Epianhydro-tetracycline	251 (140)				
Anhydrotetracycline	263 (153)				
Chlortetracycline	55.1 (39.7)		<LOQ - 14.7		
4-Epichlortetracycline	119 (100)				
4-Epianhydrochlortetracycline	421 (397)				
Anhydrochlortetracycline	126 (105)				
Isochlortetracycline	83.4 (39.6)				
Oxytetracycline	57.9 (43.2)				
4-Epioxytetracycline	45.3 (41.5)				
Demeclocycline	106 (99.2)				
Doxycycline	877 (424)	<1200–1500			<LOQ - 1500
Minocycline	660 (432)				
Reference	U.S. EPA (2009)	Harrison et al. (2006)	Spongberg and Witter (2008)		Lindberg et al. (2005)

[a] mean (median) LOQ = limit of quantitation

Table 33. Effect of Storage Treatment on Tetracyclines in Aerobically Digested Biosolids (Wu et al., 2008)

Storage Treatment	Storage Time (days)	Concentration (ng/g TS)	
		Tetracycline	Doxycycline
Dark, Anaerobic	0	1500	1500
	7	900	1400
	35	no data [a]	1000
	77	no data	1100
Dark, Aerobic	0	1600	1500
	7	800	1500
	35	no data [a]	750
	77	no data	600
Daylight, Aerobic	0	1600	1500
	7	700	1600
	35	no data [a]	800
	77	no data	700

[a] study terminated after 8 days

Sulfonamide Antibiotics

Occurrence data for the sulphonamide class of drugs is scattered through the technical literature, with most attention devoted to one or two compounds (sulfamethoxazole). The most complete

data set is derived for the U.S. EPA's TNSSS, in which sulfanilamide was detected at the highest concentration, approximately one to two orders of magnitude higher than the remaining drugs in this class (Table 34). In the literature survey by Jones-Lepp and Stevens (2007), maximum concentrations of sulfametazine and sulfapyridineb were 160 and 197 ng/g TS, respectively. Many of the sulphonamide class tested by Spongberg and Witter (2008) were beneath the limit of quantitation.

Several researchers included analysis of sulfamethoxazole in anaerobically digested sludge samples, but reported it at non-detectable levels (Göbel et al., 2005; Heidler and Halden 2008).

Table 34. Concentrations of Sulfonamide Antibiotics in Sludges and Biosolids

Sulfonamide Compound	Concentration (ng/g TS)						
	Survey of biosolids	Unknown sludge (An urban WWTP)	WAS (5 WWTPs)	Biosolids class A + sludge	Anaerobic digested sludge	Primary Sludge	Anaerobic Sludge
Sulfadiazine	13.6 (9.8) [a]						
Sulfachloropyridazine	12.0 (9,8)						
Sulfadimethoxine	3.57 (2.01)	< LOQ - 8.15					
Sulfametazine	7.38 (4.0)	<LOQ - 26.7		nd-160			
Sulfamethizole	4.72 (3.97)	<LOQ					
Sulfamethoxazole	21.59 (4.32)					10±10[b]	5 ±5
Sulfanilamide	536 (99.2)						
Sulfathiazole	10.7 (9.8)	<LOQ					
Sulfapyridine				nd-197	1,000±100		
Sulfisoxazole		<LOQ - 21.9					
Sulfadimidine			nd - 31				
Sulfonamide			nd - 20				
Reference	U.S. EPA (2009)	Spongberg and Witter (2008)	Xu et al. (2007)	Jones-Lepp and Stevens (2007)	Göbel et al. (2005)	Radjenovic et al. (2009)	

[a] mean (median) [b] mean ± standard deviation
LOQ = limit of quantitation nd = not detected

Treatment
Sulfamethoxazole was found to be highly amenable to anaerobic digestion in several laboratory-scale studies by Carballa and colleagues. This sulfa drug was so readily degradable in anaerobic digestion (99% removal) that no difference in removal efficiency due to operating temperature or solids retention time could be discerned (Caraballa et al. 2006, 2007a) (Table 35). Similarly, no difference in the effect of pre-ozonating the sludge prior to digestion could be observed because the compound was effectively eliminated completely (Carballa et al., 2007b).

Fluoroquinoline and Quinoline Antibiotics
The most complete identification of fluoroquinolone compounds in biosolids comes from the U.S. EPA TNSSS (Table 36). The drugs found at the highest levels were ciprofloxacin and

Table 35. Effect of Anaerobic Digestion Conditions on Removal Efficiency of Sulfamethoxazole

Anaerobic Temperature	Condition	Removal Efficiency %)	Reference
mesophilic	SRT = 20 d	(99 ± 1)[a]	Carballa et al. (2006)
mesophilic	SRT = 30 d	99	Carballa et al. (2007a)
mesophilic	SRT = 20 d	99	
mesophilic	SRT = 10 d	99	
thermophilic	SRT = 20d	98	
thermophilic	SRT = 10 d	99	
thermophilic	SRT = 6 d	99	
mesophilic	Non-ozonated	(100 ± 1)	Carballa et al. (2007b)
mesophilic	Ozonated	(100 ± 1)	
thermophilic	Non-ozonated	(100 ± 1)	
thermophilic	Ozonated	(100 ± 1)	

[a] mean ± standard deviation SRT = solids retention time

ofloxacin, at median concentrations of 5,370 and 3,110 ng/g TS, respectively. These two pharmaceuticals have been identified most frequently in the literature, along with norfloxacin. The remaining fluoroquinolines and quinolines in the EPA survey had median concentrations under 50 ng/g TS.

Table 36. Concentrations of Fluoroquinolone and Quinolone Antibiotics in Sludges and Biosolids

Fluoroquinoline /Quinoline	Concentration (ng/g TS)					
	Sludge Survey	Anaerobic digestion (n=5 WWTPs)	Aerobic digestion (n=1 WWTP)	Literature Review	Unknown sludge (An urban WWTP)	Unknown sludge (A rural WWTP)
Ciprofloxacin	10,500 (5,370) [a]	1,400 – 4,800	500 - 900	50 – 4,800	<LOQ - 46.3	8.3
Clinafloxacin	75.6 (40.4)					
Enrofloxacin	27.9 (19.8)					
Flumequine	10.6 (9.87)					
Lomefloxacin	22.9 (19.8)					
Norfloxacin	275 (109)	900 – 4,200	100 - 400	10 – 4,200		
Ofloxacin	8,570 (3,110)	<LOQ–2,000	100 - 700	<10 – 2,000		
Oxolinic acid	4.7 (4.0)					
Sarafloxacin	294 (91.9)					
Reference	U.S. EPA (2009)	Lindberg et al. (2005)		Harrison et al. (2006)	Spongberg and Witter (2008)	

[a] mean (median)

Concentrations of the three main identified fluoroquinolones in raw and digested sludge samples are summarized in Table 37. Many of the publications reviewed indicate that the concentrations of ciprofloxacin and norfloxacin in the sludge samples are similar in magnitude on the order of

2,000 to 6,000 ng/g TS. Lindberg et al. (2005) tracked the concentrations of ciprofloxacin and norfloxacin through the residual solids stream of a wastewater treatment plant in Umea, Sweden. The concentrations increased as the sludge was combined, digested and dewatered, but then dropped significantly after drying by pelletization. It appears that these compounds are relatively unaffected by the anaerobic digestion process, as concentrations are in many cases higher in the digested sludge than in the raw sludge.

Table 37. Concentrations of Three Fluoroquinones in Raw and Digested Sludge Samples

Sludge Type	Source	Concentration (ng/g TS)			Reference
		Ciprofloxacin	Norfloxacin	Ofloxacin	
Primary sludge		2,900	2,900		Lindberg et al. (2006)
Secondary sludge		2,500	1,500		
Raw sludge to digester		6,600	5,100		
Digested sludge		6,000	7,000		
Dewatered digested sludge		10,600	9,800		
Dried biosolids (Pellets)		2,600	3,400		
Raw sludge	WWTP1	1,400 ± 120 [a]	1,540 ± 30		Golet et al. (2002)
	WWTP2	2,030 ± 200	1,960 ± 150		
Digested sludge	WWTP3	2,420 ± 60	2,370 ± 70		
	WWTP4	2,720 ± 200	2,130 ± 190		
Waste activated sludge		2,500 ± 100	2,600 ± 100		Golet et al. (2003)
Raw sludge		2,200± 400	2,100 ± 200		
Anaerobic digested sludge		3,100 ± 400	2,900 ± 400		
Raw sludge		1,000-2,000	1,500–2,000		Jones-Lepp and Stevens (2007)
Digested sludge		2,300–2,400	2,100–2,400		
Digested sludge		3,100-5970	2,900-6970		Heidler and Halden (2008)
Waste activated sludge	Plant A		301± 89	227 ± 46	Xu et al. (2007)
	Plant B		40 ± 165	886 ± 222	
	Plant C		187 ± 38	165 ± 71	
	Plant D		372 ± 97	835 ± 186	
Primary Sludge				190±280	Radjenović et al. (2009)
Anaerobic Digested				80 ±30	

[a] mean ± standard deviation

Treatment

When aerobically digested sludge containing the ciprofloxacin was stored under different light and redox environments, there was no reduction in concentration from the initial starting level after 77 days of storage (Wu et al., 2008).

Macrolide Antibiotics

In Table 38, the data generated by the U.S. EPA's TNSSS suggest that azithromycin, tylosin and virginiamycin are present at the highest concentrations of the macrolide antibiotics, with mean values of 831 and 138 ng/g TS. Concentration data from an anaerobically digested sludge by Gobel et al. (2005) exhibited some of the highest concentrations of this class of antibiotics. Otherwise, concentration data for this class of antibiotics were sparse.

Table 38. Concentrations of Macrolide Antibiotics in Sludges and Biosolids

Sludge Type	Concentration (ng/g TS)						Reference
	Azithro-mycin	Clarithro-mycin	Erythro-mycin	Roxithro-mycin	Virginia-mycin	Tylosin	
Not specified (sludge survey)	831 (278)[a]	41.58 (13.4)	36 (19)	8.1 (4.7)	138 (73.3)	269 (128)	U.S. EPA (2009)
Activated and digested	1.3-158	0.3-63		nd-131			Jones-Lepp and Stevens (2007)
Anaerobic digestion	2,500± 1,000[b]	700 ± 400					Göbel et al. (2005)
Unknown sludge (urban WWTP)		<LOQ - 30.2					Spongberg and Wittmer (2008)
Unknown sludge (rural WWTP)		<LOQ					
Primary sludge			105±50				Radjenović et al. (2009)
Anaerobically digested			70 ±30				
Not specified (2 plants)				<LOQ – 1,800		300 – 4,000	Nieto et al. (2007)
Waste activated sludge (Plant A)			76 ± 25	40 ± 23			Xu et al. (2007)
Waste activated sludge (Plant B)			195 ± 56	64 ± 16			
Wasted activated sludge (Plant C)			38 ±14	32 ± 9			
Waste activated sludge (Plant D)			62 ± 24	44 ± 10			

[a] mean (median) [b] mean ± standard deviation
LOQ = limit of quantitation nd = not detected

Treatment

Only limited data were found on the removal of a macrolide antibiotic by biosolids treatment. Studies by Carballa et al. (2006, 2007a) indicated that roxithromycin was highly degradable in laboratory-scale anaerobic digesters. The combination of thermophilic operation and longer retention times appeared to contribute to higher removal efficiency (Table 39).

Table 39. Effect of Temperature and Retention Time on Removal of Roxithromycin in Anaerobic Digestion

Anaerobic Temperature	Retention Time (days)	Removal Efficiency (%)	Reference
mesophilic	20	(85 ± 15)[a]	Carballa et al. (2006)
mesophilic	30	99	Carballa et al. (2007a)
mesophilic	20	95	
mesophilic	10	no data	
thermophilic	10	(95 ± 5)	Carballa et al. (2006)
thermophilic	20	99	Carballa et al. (2007a)
thermophilic	10	98	
thermophilic	6	no data	

[a]mean ± standard deviation

When aerobically digested sludge containing the antibiotics clarithromycin and erythromycin was stored under different light and redox environments, both antibiotics declined from between 2,000 and 2,500 ng/g TS to non-detectable concentrations from between 7 and 35 days (Wu et al., 2008) (Table 40). Neither storage under anaerobic or aerobic conditions, nor the presence or absence of daylight appeared to affect the removal rates.

Table 40. Effect of Storage Treatment on Macrolides in Aerobically Digested Biosolids (Wu et al., 2008)

Storage Treatment	Storage Time (days)	Concentration (ng/g TS)	
		Clarithromycin	Erythromycin
Dark, Anaerobic	0	2,500	2,000
	7	300	1,200
	35	0	800
	77	0	250
Dark, Aerobic	0	2,500	2,200
	7	200	800
	35	0	400
	77	0	200
Daylight, Aerobic	0	2,500	2,200
	7	200	800
	35	0	300
	77	0	200

Beta-Lactam Antibiotics

This class of antibiotics contains the well-recognized penicillin and similar drugs. Few data were identified for these compounds, with only the U.S. EPA's TNSSS providing any information on occurrence on biosolids (Table 41). Penicillin V at 41 ng/g TS was detected at approximately twice the concentration of the other types of beta-lactams.

Table 41. Concentrations of Beta-Lactam Antibiotics in Sludges and Biosolids (U.S. EPA, 2009)

Beta-lactam	Concentration (ng/g TS)
Cloxacillin	26.4 (19.9)[a]
Oxacillin	20.8 (19.8)
Penicillin G	20.8 (19.8)
Penicillin V	41.4 (39.6)

[a] mean (median)

No data on removal efficiencies in biosolids treatment processes were found in this review.

Lincosamide Antibiotics

Only limited occurrence data in biosolids or sludges were found for this class of compounds (Table 42). In the EPA's TNSSS (U.S. EPA, 2009), lincomycin and clindamycin were found at median concentrations of 19.9 and 13.4 ng/g TS, respectively. In Ohio, clindamycin in sludges of three urban treatment plants ranged from 3.7 to 154 ng/g TS, while in sludge a rural treatment facility, the concentration was18.2 ng/g TS (Spongberg and Witter, 2008).

Table 42. Concentrations of Lincosamide Antibiotics in Sludges and Biosolids

Sludge Type	Lincomycin	Clindamycin	Reference
Not Specified (sludge survey)	30.2 (19.9) [a]	41.58 (13.4)	U.S. EPA (2009)
Unknown sludge (An urban WWTP)		3.7 - 154	Spongberg and Witter (2008)
Unknown sludge (A rural WWTP)		18.2	

[a] mean (median)

The effect of different storage conditions on the removal of clindamycin in aerobically digested solids was examined by Wu et al. (2008). The antibiotic declined rapidly in the first seven days of storage in both aerobic and anaerobic conditions and with daylight either present or absent in the aerobic samples (Table 43). Removal of the clindamycin was much slower in the remaining 70 days of the study. All test conditions produced approximately the same test results.

3.4.3 Nervous System

Anti-Epileptics (Anti-Convulsants)

Occurrence Data

Anti-epileptic drugs are used in the control of epilepsy. Occurrence data were primarily found for carbamazepine. The only reference to a second anti-epileptic drug, Primidone, stated the concentrations in sludge samples from three Ohio treatment plants were lower than the level of quantitation (Spongberg and Witter, 2008). Concentrations of carbamazepine in biosolids and sludge samples fell in a relatively narrow range (Table 44), from non-detectable to a maximum of 850 ng/g TS as reported by Jones-Lepp and Stevens (2007).

Table 43. Effect of Storage Treatment on Clindamycin in Aerobically Digested Biosolids (Wu et al., 2008)

Storage Treatment	Storage Time (days)	Clindamycin Concentration (ng/g TS)
Dark, Anaerobic	0	2,500
	7	1,000
	35	800
	77	800
Dark, Aerobic	0	2,500
	7	800
	35	700
	77	500
Daylight, Aerobic	0	2,500
	7	900
	35	700
	77	600

Table 44. Occurrence Data for Carbamazepine in Biosolids

Biosolids Source	Concentration (ng/g TS)	Reference
Raw sludge	69.6 ±2.2[a]	Miao et al. (2005)
Waste Activated Sludge	150 ng/g OC	Kinney et al. (2006)
Dewatered sludge	64 ng/g OC	
Not specified (Sewage treatment plant 1)	nd - 78	Nieto et al. (2007)
Not specified (Sewage treatment plant 2)	50 - 165	
Not specified (An urban WWTP)	4.8 - 12.9	Spongberg and Witter (2008)
Not specified (A rural WWTP)	21.1	
Not specified (sludge survey)	135 (55)[b]	U.S. EPA (2009)
Primary sludge	70 ±60[a]	Radjenović et al. (2009)
Biosolids class A + sludge	nd-850	Jones-Lepp and Stevens (2007)

nd = not detected [a] mean ± standard deviation [b] mean (median)

Concentrations of carbamazepine following biosolids treatment processes are presented in Table 45. Kinney et al. (2006) compared several treatment processes. Other than anaerobic digestion, which exhibited a very high carbamazepine concentration of 1200 ng/g TS, concentrations were less than 180 ng/g TS. Other samples of anaerobically digested sludge ranged up to 281 ng/g TS. Carbamazepine concentrations observed in the treated biosolids were similar in magnitude to those of the other biosolids samples presented in Table 45.

Metabolites of carbamazepine were monitored in the raw and digested solids of the Peterborough (ON) wastewater treatment plant (Miao et al., 2005) (Table 46). Concentrations of the metabolites included for analysis were either non-detected or present at very low concentrations

(e.g., 2 -4 ng/g TS), compared to the concentrations of the parent compound in raw sludge (70 ng/g TS, Table 44) or digested sludge (258 ng/g TS, Table 45).

Table 45. Concentrations of Carbamazepine in Treated Biosolids

Treatment Process	Concentration ng/g TS	Reference
compost	15-180 ng/g OC	Kinney et al. (2006)
heat dried	140 ng/g OC	
air dried	51 ng/g OC	
Anaerobic digested sludge	1,200 ng/g OC	
Anaerobic digested sludge	258.1 ±4.7[a]	Miao et al.(2005)
Digested sludge	281	Heidler and Halden (2008)
Anaerobic digested sludge	80 ±10	Radjenović et al. (2009)

[a] mean ± standard deviation OC = organic carbon

Table 46. Metabolites of Carbamazepine in Sludge (Miao et al., 2005)

Carbamazepine Metabolite	Concentration (ng/g TS)	
	Raw sludge	Anaerobic digested sludge
10,11-dihydro-10,11-epoxycarbamazepine	nd	nd
10,11-dihydro-10-hydroxycarbamazepine	nd	nd
2-hydroxycarbamazepine	1.9 ±1.1 [a]	3.4 ±0.9
3-hydroxycarbamazepine	1.6 ±0.8	4.3 ±0.9

[a] mean ± standard deviation nd = not detected

Removal of Carbamazepine in Biosolids Treatment Processes

Only a limited set of tests have examined the removal of carbamazepine by biosolids treatment processes. Trials with anaerobic digestion documented by Carballa et al. (2007a) at laboratory scale indicated that carbamazepine was not reduced by the treatment at time up to 30 days at mesophilic conditions, and up to 20 days at thermophilic conditions (Table 47). Ternes et al. (2005) also reported that batch anaerobic digestion tests resulted in no removal of carbamazepine.

Carballa et al. (2007b) also investigated the effect of pre-ozonation prior to anaerobic digestion under mesophilic and thermophilic conditions at laboratory scale (Table 48). The pre-ozonation treatment had a beneficial effect on the removal of carbamazepine, with up to a 60% reduction after 10 days at thermophilic temperatures. The authors considered the beneficial effect of the ozone was due to the carbamazepine existing primarily in the liquid phase, making it more susceptible to attack by the ozone (Carballa et al., 2007b).

Table 47. Effect of Anaerobic Digestion Treatments on Removal of Caramazepine (Carballa et al., 2007a)

Anaerobic Sludge Type	Solids Retention Time (d)	Removal Efficiency (%)
mesophilic	30	0
	20	0
	10	12
thermophilic	20	0
	10	0
	6	22
Average		0

Table 48. Effect of Pre-Ozonation on Anaerobic Digestion of Carbamazepine (Carballa et al., 2007b)

Sludge Type	Operating condition and SRT	Removal Efficiency, %
anaerobic digestion - mesophilic	Non-ozonated 20 d	0
anaerobic digestion - mesophilic	Ozonated 20 d	18± 5[a]
anaerobic digestion - thermophilic	Non-ozonated 10 d	0
anaerobic digestion - thermophilic	Ozonated 10 d	60 ± 3

[a] mean ± standard deviation

Wu et al. (2008) examined the effects of storage methods on the reduction of carbamazepine and other pharmaceuticals in aerobically digested sludge. Treatment methods consisted of aerobic and anaerobic treatment in dark containers, and aerobic treatment subjected to solar radiation. There was no reduction from the initial carbamazepine concentration of approximately 2,500 ng/g TS in any of the three treatments after 77 days of storage.

Mood-altering Pharmaceuticals

This section includes four different categories of pharmaceuticals including anti-anxiety, anti-depressants (Amitriptyline, Fluoxetine and Paroxetine), anti-psychotics (Chlorpromazine and Thioridazine), and psycho-stimulants (amphetamine, methamphetamine and caffeine). Data concerning these drugs in biosolids are generally scarce. The concentration data for fluoxetine indicate it is found typically in a range between 100 and 1,000 ng/g TS (Table 49). Concentrations of fluoxetine and paroxetine in a sample of primary sludge were of approximately the same magnitude (Radjenović et al., 2009).

Table 49. Concentration of Two Anti-Depressants in Sludge Samples

Sludge Source	Concentration (ng/g TS)		Reference
	Fluoxetine	Paroxetine	
Waste Activated Sludge	370 ng/g OC		Kinney et al. (2006)
Dewatered Sludge	830 ng/g OC		
Primary Sludge	100±50[a]	70±50	Radjenović et al. (2009)
Biosolids class A + sludge (literature review)	nd-59		Jones-Lepp and Stevens (2007)
Not specified (sludge survey)	245 (147)[b]		U.S. EPA (2009)

[a] mean ± standard deviation [b] mean (median) OC = organic carbon

Concentrations of two anti-depressants in the product of biosolids treatment processes are provided in Table 50. Although some of the treated samples exhibit concentrations similar to those in Table 49, the values reported by Kinney et al. (2006) for other drying and anaerobically digested sludge are an order of magnitude higher than for the other treated biosolids and sludge samples. The disparity emphasizes the variability that can be observed in the different sludge matrices and locations.

Table 50. Concentration of Two Anti-Depressants following Biosolids Treatment Processes

Treatment Process	Concentration (ng/g TS)		Reference
	Fluoxetine	Paroxetine	
Heat drying	480 ng/g OC		Kinney et al. (2006)
Composting	255-705 ng/g OC		
Air drying	2,800 ng/g OC		
Anaerobic digestion	4,700 ng/g OC		
Anaerobic digestion	150 ±60 [a]	50 ± 20	Radjenović et al. (2009)

[a] mean ± standard deviation

One study attempted to detect these compounds, as part of a suite of target compounds, using different analytical extraction procedures (Gielen, 2007). The data are useful to indicate that concentrations of the pharmaceuticals are lower than the limit of quantitation. Results of this testing is presented in Table 51.

Table 51. Concentrations of Mood-altering Pharmaceuticals in Sludges (Gielen, 2007)

Extraction Procedure	Biosolids Source	Concentration (ng/g TS)		
		Amitriptyline	Chlorpromazine	Thioridazine
Soxhlet extraction	compost	<1	<14	<70
	WAS	not detected	not detected	not detected
	primary sludge	<1	not detected	not detected
Supercritical fluid extraction	compost	<5	<5	<259
	WAS	<5	<5	<259
	primary sludge	<5	<5	<259

Only a few studies provided any data on concentrations of psycho-stimulants in sludges or biosolids. The occurrence data appear in Table 52. The data indicate that caffeine and its metabolite can be present in variable concentrations from 5 to 5,000 ng/g TS. Gielen demonstrated that different extraction procedures in the analysis of caffeine in sludges can have a significant effect on the concentration reported. The data for amphetamine and methamphetamine are limited, with amphetamine exhibiting a higher concentration range than methamphetamine.

Table 52. Concentrations of Psycho-Stimulants in Sludges

Constituent	Sludge Type	Concentration (ng/g TS)	Ref
Amphetamine	Biosolids class A + sludge (literature review)	5-300	Jones-Lepp and Stevens (2007)
Methamphetamine	Biosolids class A + sludge (literature review)	0-4	
Caffeine	Unknown sludge (WWTP 1)	57 - 69	Nieto et al. (2007)
	Unknown sludge (WWTP 2)	<LOQ - 65	
	Unknown sludge (An urban WWTP)	<LOQ - 5.2	Spongberg and Witter (2008)
	Unknown sludge (A rural WWTP)	4.8	
	compost	7.4/43 [a]	Gielen (2007)
	Waste activated sludge	238/1,888	
	primary sludge	4,530/1,585	
1,7-Dimethylxanthine	Not specified (sludge survey)	1,180 (987) [b]	U.S. EPA (2009)

LOQ = limit of quantitation
[a] Soxhlet extraction/Supercritical fluid extraction [b] mean (median)

3.4.4 Analgesics and Anti-Inflammatory Drugs

Occurrence

Analgesics are drugs that relieve pain (i.e., "pain-killers"). Non-steroidal-anti-inflammatory drugs (NSAIDs) may be used both as analgesics and for their anti-inflammatory purposes, in which they inhibit an enzyme (cyclooxygenase) contributing to the inflammation process.

The only analgesic compound identified in this review is acetaminophen (also called paracetamol in other countries). Several NSAIDs were identified in sludge and biosolids samples, including diclofenac, ibuprofen, naproxen, ketoprofen, indometacin and mefenamic acid. Occurrence data for these pharmaceuticals are found in Table 53. Compilation of data in this table reveals that the different analytical surveys have different suites of pharmaceuticals of interest. Only the NSAID Diclofenac appears on the target list of most publications examined, but was not an analyte of the recent EPA (2009) survey. Compounds that appear in the target lists of different research teams appear to be a function of pharmaceutical consumption (which in itself may be country-specific) and the analytical procedures used.

Removal of Analgesics and NSAIDs

There are few data reported in the literature on removal efficiencies of the analgesic and NSAID compounds. With laboratory-scale anaerobic digesters, Carballa et al. (2007a) observed that naproxen was readily removed under both mesophilic and thermophilic conditions, even at the shortest retention times tested (Table 54). Ibuprofen was more resistant to removal during anaerobic digestion, with slightly improved reductions at thermophilic over mesophilic operation

Table 53. Occurrence of Analgesics and Non-Steroidal Anti-Inflammatory Drugs (NSAIDs) in Sludges

Biosolids Source	Concentration (ng/g TS)								Reference
	Acetamin-ophen	Codeine	Diclofenac	Ibuprofen	Indomet-acin	Ketoprofen	Naproxen	Mefen-amic acid	
Not specified (Sewage treatment plant 1) (n=5)	nd - 34		nd - 65		70 - 99		nd - 242		Nieto et al. (2007)
Not specified (Sewage treatment plant 2) (n=5)	nd - 42		nd - 183		nd - 75		nd - 87		
Not specified (literature survey)	0.0006–4535								Harrison et al. (2006)
Not specified (An urban WWTP) (n=3)			<LOQ - 23.1						Spongberg and Witter (2008)
Not specified (A rural WWTP) (n=1)			28.5						
Not specified (sludge survey)	462 (396)[a]	30.6 (19.9)		653 (143)			86.2 (31.6)		EPA (2009)
Primary sludge			215±130[b]	535±193		220±110		10±5	Radjenović et al. (2009)
Anaerobic digested sludge			190±130	300 ±70		40±40		50±15	
Biosolids class A + sludge	nd-1400								Jones-Lepp and Stevens (2007)
Compost				<5					Gielen (2007)
Waste Activated Sludge				<1 - 41					
Primary sludge				153 - 299					

LOQ = Limit of Quantitation nd = not detected
[a] mean (median) [b] mean ± standard deviation

Table 54. Effect of Anaerobic Digestion Treatments on Removal of NSAIDs

Sludge Type	Condition	Removal Efficiency (%)			Reference
		Diclofenac	Ibuprofen	Naproxen	
anaerobic digestion - meso			40 ± 15 [a]	87± 5	Carballa et al. (2006)
anaerobic digestion - thermo			47 ± 10	91 ± 5	
anaerobic digestion - meso	SRT=30 d	5	30	80	Carballa et al. (2007a)
anaerobic digestion - meso	SRT=20 d	0	40	90	
anaerobic digestion - meso	SRT=10 d	80	60	90	
anaerobic digestion - thermo	SRT=20d	30	40	92	
anaerobic digestion - thermo	SRT=10 d	15	50	92	
anaerobic digestion - thermo	SRT=6 d	80	55	85	
anaerobic digestion		0	40	>80	Ternes et al. (2005)

[a] mean ± standard deviation

(Carballa et al., 2006, 2007a). Diclofenac was relatively more resistant to removal in anaerobic digestion than either naproxen or ibuprofen. The relative removal efficiencies of the three NSAIDs were confirmed in laboratory batch anaerobic digestion tests completed by Ternes et al. (2005).

The effect of pre-ozonation prior to anaerobic digestion under mesophilic and thermophilic conditions at laboratory scale was examined by Carballa et al. (2007b) (Table 55). The pre-ozonation treatment had no beneficial effect on the removal of the NSAIDs tested, with perhaps even a slightly negative impact on reduction by anaerobic digestion when preceded by ozonation.

Table 55. Effect of Pre-Ozonation on Anaerobic Digestion of NSAIDs (Carballa et al., 2007b)

Sludge Type	Condition	Removal Efficiency (%)	
		Diclofenac	Ibuprofen
anaerobic digestion - meso	Non-ozonated	80 ± 1 [a]	40 ± 10
anaerobic digestion - meso	Ozonated	75 ± 0	20 ±8
anaerobic digestion - thermo	Non-ozonated	75 ± 3	50 ± 10
anaerobic digestion - thermo	Ozonated	70 ± 1	45 ± 7

[a] mean ± standard deviation

3.4.5 Bacteroistat Antibiotics

Occurrence

Triclosan and triclocarban are compounds displaying antimicrobial activity against both gram-positive and gram-negative activity, resulting in their use in an array of consumer products such as soaps, detergents and cosmetics (Heidler and Halden, 2007). Hexachlorophene is used as a topical anti-bacterial agent in soaps and some toothpastes. Due to these uses, they are transferred to grey water as a result of bathing, laundry and other domestic activities. At a wastewater treatment plant, the compounds are likely to be either sorbed onto solids, biodegraded to some extent or discharged in the treated effluent. As reported in the CCME Review of State of

Knowledge of Municipal Effluent Science and Research (Hydromantis et al., 2005), removal efficiency of triclosan by treatment plants can be variable.

Lee and Peart (2002) assessed the concentrations of triclosan and hexachlorophene as two of a suite of micro-constituents in Canadian sludges, both raw and digested (Table 56).

Table 56. Occurrence of Triclosan and Hexachlorophene in Canadian Municipal Sludges (Lee and Peart, 2002)

Municipal Treatment Plant and Sludge Type	Triclosan (ng/g TS)	Hexachlorophene (ng/g TS)
Edmonton (Goldbar) Raw	14,800	102
Regina Raw	7,160	216
Adelaide Raw	3,430	308
Burlington Raw	8,900	797
Ottawa Raw	17,900	208
Toronto (Ashbridges Bay) Raw	10,600	138
Toronto (Highland Creek) Raw	17,100	502
Toronto (Humber) Raw	16,500	397
Toronto (North) Raw	12,500	181
Montreal (MUC-PSI) Raw	14,200	562
Quebec City Raw	6,100	113
Quebec City Raw	5,500	72.6
Vancouver Digested	8,410	477
Vancouver Digested	24,700	420
Calgary (Bonnybrook) Digested	12,800	371
Calgary (Fish Creek) Digested	19,500	218
Edmonton (Goldbar) Digested	22,000	285
Regina Digested	18,900	420
Saskatoon Digested	9,900	352
Saskatoon Digested	19,400	597
Burlington Digested	7,480	451
Galt Digested	28,200	421
Guelph Digested	16,200	727
Hamilton Digested	11,500	1,190
Ingersoll Digested	16,100	640
Kitchener Digested	18,600	254
Ottawa Digested	11,700	693
Waterloo Digested	8,840	311
Windsor Digested	20,300	548
Toronto (Ashbridges Bay) Digested	16,600	328
Toronto (Humber) Digested	5,400	572
Toronto (North) Digested	900	22.6
Granby Digested	1,920	68.7
Moncton Digested	7,530	701
Truro Digested	8,410	477
median raw	10,600	208
median digested	14,450	421

The median values of triclosan in raw and digested sludge samples were 10,600 and 14,450 ng/g TS, respectively, suggesting that there is no reduction of triclosan as a result of digestion. Similar results were observed for hexachlorophene, with median values in raw and digested sludge samples of 208 and 421 ng/g TS, respectively. The highest concentrations of triclosan in raw and digested sludges were found in samples from Ottawa and Galt (ON) treatment plants, at 17,900 and 28,200 ng/g TS, respectively. A sample of digested sludge from the Toronto North facility had the lowest concentration of triclosan at 900 ng/g TS. The highest concentration of hexachlorophene in raw sludge in this survey was in a sample from Burlington, ON, while the highest concentration in digested sludge was observed in a sample from Hamilton, ON. The lowest concentration of hexachlorophene was recorded in a digested sludge sample from the North Toronto plant.

Concentrations of triclosan in other sludge samples are provided in Table 57. A recent survey of Canadian sludges by XCG Consultants (2007) reported concentrations of triclosan in a range of 900 – 28,000 ng/g TS, with a median value of 13,000 ng/g TS. In four Ontario treatment plants, concentrations of Triclosan ranged between 680 and 11,550 ng/g TS. Concentrations reported in other publications were of a similar magnitude, between 3,200 and 42,000 ng/g TS.

Table 57. Concentration of Triclosan in Other Sludge Samples

Sludge Source	Concentration (ng/g TS)		Reference
	Range	mean (median)	
Canadian sludge (1995-1998)	900 - 28,000	13,000	XCG (2007)
Treated Biosolids (4 Ontario plants)	680 – 11,550		Chu and Metcalfe (2007)
Mid-Atlantic U.S. plant	20,000 - 55,000	30,000 ± 11,000[a]	Heidler and Halden (2007)
Not specified (literature review)	nd – 15,600		Harrison et al. (2006)
dewatered anaerobically digested or dewatered secondary sludge (Greece)	190 – 9,850	3,210 (2,710)	Stasinakis et al. (2008)
Not specified (France)		41,900 ± 37,000	Ruel et al. (2008)
Not specified (U.S. survey)		16,100 (3,860)	U.S. EPA (2009)

nd = not detected [a] mean ± standard deviation

Biosolids treatment processes had no real discernible effect on reducing concentrations of triclosan, as indicated in Table 58.

Two additional pharmaceuticals with bacteriostatic properties were identified, including trimethoprim often used for fighting urinary tract infections and chloramphenicol, mainly used in eye drops or ointment for bacterial conjunctivitis. Concentrations of trimethoprim in sludges are generally low (less than 100 ng/g TS) as indicated in Table 59. Chloramphenicol concentrations were beneath the limit of quantitation in waste activated sludges analysed by Xu et al. (2007).

Table 58. Concentration of Triclosan following Biosolids Treatment Processes

Sludge Source	Concentration (ng/g TS)	Reference
Compost-B	7,400	LaGuardia et al. (2004)
Lime-A	4,700	
Heat treated-A	6,900	
Anaerobic digestion -A	5,200	
Anaerobic digestion -B	5,500	
Anaerobic digestion -E	3,600	
Anaerobic Digested sludge	1,200 - 30,000	Heidler and Halden (2008)
Anaerobic Digested Sludge	20,000 ± 18,000 [a]	Halden (2007)

[a] mean ± standard deviation

Table 59. Concentrations of Trimethoprim in Sludges and Biosolids

Sludge source	Concentration	Reference
Not Specified (Sludge survey)	30.4 (10.8)[a]	U.S. EPA (2009)
Biosolids Class A &B and sludge	nd – 22	Jones-Lepp and Stevens (2007)
Primary sludge	40±15[b]	Radjenović et al. (2009)
Anaerobic digestion	20 ±5	
Digested sludge	<0.1	Heidler and Halden (2008)
Anaerobic digestion	<100	Göbel et al. (2005)
Not specified (2 plants)	<LOQ	Nieto et al. (2007)

nd = not detected LOQ = limit of quantitation
[a] mean (median) [b] mean ± standard deviation

3.4.6 Cardiovascular Pharmaceuticals

This class of pharmaceuticals are those which affect the cardiovascular system. Drugs in this class have generic actions including beta-blockers (atenolol, propanolol), calcium-channel blockers (diliatazem), thiazides (hydrochlorothiazide) and digoxin, These pharmaceuticals are used to control heart arrhythmia and hypertension (high blood pressure). Hydrochlorothiazide is prescribed as an anti-diuretic, which ultimately helps to reduce hypertension. Dehydronifedipine is a metabolite of the calcium-channel blocker nifedipine.

The compounds in this class that were identified in the literature review are provided in Table 60. The range of concentrations for pharmaceuticals in this class appears to be on the order of 10 to 400 ng/g TS. Based on the data provided by Radjenović et al., (2009), treatment of primary sludge by anaerobic digestion appears to result in a reduction of this class of compounds, with atenolol having a mean concentration notably lower in digested sludge compared to the level in primary non-digested sludge.

3.4.7 Alimentary Tract Pharmaceuticals

Occurrence
This class of pharmaceuticals includes anti-diabetic drugs, and anti-dyspeptics or acid reflux

inhibitors. The latter group have technical names including hydrogen receptor agonists or proton pump inhibitors. In Table 61, the anti-diabetic drug metformin is higher in concentration than its alternate glibenclamide. Cimetidine was identified at the highest concentration (1,330 ng/g TS) of gastric reflux inhibitors by a wide margin. Studies involving testing of sludges revealed non-detectable concentrations of cimetidine (Spongberg and Witter, 2008) and the proton pump inhibitor omeprazole (Nieto et al., 2007).

Table 60. Concentrations of Cardiovascular Pharmaceuticals in Sludges and Biosolids

Compound	Concentration (ng/g TS)				
	Not Specified (Sludge Survey)	Biosolids class A + sludge	Primary Sludge	Anaerobically Digested Sludge	Not specified (2 plants)
Atenolol			90±30[b]	10±2	
Digoxin	208 (99.4)[a]				
Diltiazem	40.2 (14.8)	nd-26			<LOQ - 12.8
Hydrochlorothiazide			40±20	15±10	
Propranolol			40±20	30±15	
Dehydronifedipine	5.03 (4.04)	8-390			
Reference	U.S. EPA (2009)	Jones-Lepp and Stevens (2007)	Radjenović et al. (2009)		Spongberg and Witter (2008)

nd = not detected
LOQ = limit of quantitation
[a] mean (median)
[b] mean ± standard deviation

Table 61. Concentrations of Alimentary Tract Pharmaceuticals in Sludges and Biosolids

Compound	Application	Sludge Source	Concentration (ng/g TS)	Reference
Glibenclamide	anti-diabetic	Primary sludge	90±100[a]	Radjenović et al. (2009)
		anaerobic digested sludge	160±30	
Metformin (hydrochloride)	anti-diabetic	Not Specified (sludge survey)	533 (546)[b]	U.S. EPA (2009)
Cimetidine	H_2-receptor antagonist (Anti-dyspeptic)	Not Specified (sludge survey)	1,330 (171)	U.S. EPA (2009)
		Biosolids class A + sludge	nd-71	Jones-Lepp and Stevens (2007)
Famotidine	H_2-receptor antagonists (Anti-dyspeptic)	Primary sludge	20±20	Radjenović et al. (2009)
		anaerobic digested sludge	60±30	
Ranitidine (hydrochloride)	H_2-receptor antagonists (Anti-dyspeptic)	Not Specified (sludge survey)	57.5 (12.5)	U.S. EPA (2009)

nd = not detected
[a] mean ± standard deviation
[b] mean (median)

Treatment

Data provided in Radjenović et al. (2009) in Table 61 are useful for comparing the effect of anaerobic digestion on concentrations of glibenclamide and famotidine. The concentrations of these two pharmaceuticals were higher following anaerobic digestion than in the raw primary sludge, indicating that the drugs are not amenable to reduction by anaerobic digestion.

3.4.8 Blood-Modifying Pharmaceuticals

This class of drugs includes anti-lipid (cholesterol lowering) (e.g., gemfibrozil, bezafibrate and clofibric acid) and anti-coagulants (e.g. Warfarin). Gemfibrozil was the compound reported most frequently in the literature, with concentrations ranging as high as 1,500 ng/g TS (Table 62). Maximum concentrations of the other pharmaceuticals in this class were substantially lower. The study completed by Radjenović et al., (2009) indicated that gemfibrozil was not likely to be removed by the anaerobic digestion process. No other data on removal efficiencies in biosolids treatment processes were found.

Table 62. Concentrations of Blood-Modifying Pharmaceuticals in Sludges and Biosolids

Sludge Source	Concentration (ng/g TS)				Reference
	Bezafibrate	Clofibric Acid	Gemfibrozil	Warfarin	
Not specified (sludge survey)			214 (101)[a]	10.5 (9.9)	U.S. EPA (2009)
Literature review			nd - 1500	nd - 92	Jones-Lepp and Stevens (2007)
Literature review			nd - 1190		Harrison et al. (2006)
Unknown sludge (An urban WWTP)		<LOQ - 8.1	<LOQ - 3.4		Spongberg and Witter (2008)
Unknown sludge (A rural WWTP)			18.3		
Two treatment plant sludges	<LOQ - 88	<LOQ - 64			Nieto et al. (2007)
Primary sludge			50±50[b]		Radjenović et al. (2009)
Anaerobic digested sludge			140 ±80		

nd = not detected LOQ = limit of quantitation
[a] mean (median) [b] mean ± standard deviation

3.4.9 Respiratory and Anti-Allergenic Pharmaceuticals

Antihistamine drugs are used to prevent the formation of histamine as a result of allergic reactions to triggers such as pollens and insect stings. Occurrence data in sludges and biosolids are limited (Table 63) and generally focus on the compound diphenylhydramine. The data provided by Jones-Lepp and Stevens (2007) suggest the concentrations can be quite variable, although collectively the literature would indicate concentrations in the range of 100 to 1,000

ng/g TS might be expected. The mean concentration of loratidine in primary sludge at 50 ng/g TS (Radjenović et al., 2009) was lower than those observed for diphenylhydramine

Although the anti-bronchospasm drug albuterol was found at relatively low concentrations of 5 ng/g TS in the EPA's TNSSS (Table 63), the literature review by Jones-Lepp and Stevens (2007) indicated much higher concentrations have been observed in sludges and biosolids.

Table 63. Concentrations of Anti-Allergenic Pharmaceuticals in Sludges and Biosolids

Compound	Sludge Source	Concentration (ng/g TS)	Reference
Diphenylhydramine	Not specified (sludge survey)	871 (541)[a]	U.S. EPA (2009)
	Biosolids class A + sludge	15-7,000	Jones-Lepp and Stevens (2007)
	Waste activated sludge	150 ng/g OC	Kinney et al. (2006)
	Dewatered sludge	170 ng/g OC	
Loratidine	Primary sludge	50±40[b]	Radjenović et al. (2009)
Albuterol	Not specified (sludge survey)	5.23 (5.29)[a]	U.S. EPA (2009)
	Literature review	nd – 1,400	Jones-Lepp and Stevens (2007)

[a] mean (median) [b] mean ± standard deviation
OC = organic carbon

Concentrations of the antiallergenic drugs in treated biosolids are provided in Table 64. Concentrations of diphenylhydramine in the data provided by Kinney et al. (2006) are highly variable. As noted above, the sludge and biosolids occurrence data, concentrations of this drug are quite variable, and so it is not possible to determine if the different concentrations in the processes result from the variability inherent in the sludge, or due to the process treatments. The data of Radjenović et al., (2009) for mean concentrations of loratidine in primary sludge (50 ng/g TS) and anaerobically digested sludge (160 ng/g TS) suggest that there is no reduction of the compound during anaerobic digestion.

Table 64. Concentrations of Two Anti-Allergenics following Biosolids Treatment Processes

Compound	Biosolids Process	Concentration (ng/g TS)	Reference
Diphenylhydramine	heat drying	2,900 ng/g OC	Kinney et al. (2006)
	compost	32 – 550 ng/g OC	
	air drying	810 ng/g OC	
	anaerobic digestion	22,000 ng/g OC	
Loratidine	anaerobic digestion	160 ± 40[a]	Radjenović et al. (2009)

[a] mean ± standard deviation OC = organic carbon

3.4.10 Anti-Parasitic and Anti-Fungal Pharmaceuticals

Anti-parasitic pharmaceuticals are used to control parasitic protozoa, helminths and other similar pathogens. The anti-fungal agent identified in the survey, miconazole, is used for treating athlete's foot, jock itch and vaginal yeast infections.

Occurrence data for these compounds were identified in two sources as indicated in Table 65. Of the three anti-parasitics identified in sludges in this review, carbadox was present at the highest concentration with a median value on 103 ng/g TS. The median concentration of miconazole was 207 ng/g TS. Considerable variability was associated with this substance in the survey as the mean value is approximately six times the median value.

Table 65. Concentrations of Anti-Parasitics and Anti-Fungals in Sludges and Biosolids

Pharmaceutical	Use	Sludge Source	Concentration (ng/g TS)	Reference
Carbadox	Anti-parasitic	Not Specified (Sludge Survey)	232 (103)[a]	U.S. EPA (2009)
Ormetoprim	Anti-parasitic	Not Specified (Sludge Survey)	4.16 (3.96)	
Thiabendazole	Anti-parasitic	Not Specified (Sludge Survey)	36.6 (16.5)	
		Biosolids class A + sludge	nd - 420	Jones-Lepp and Stevens (2007)
Miconazole	Anti-fungal	Not Specified (Sludge Survey)	1,240 (207)	U.S. EPA (2009)
		Biosolids class A + sludge	nd - 460	Jones-Lepp and Stevens (2007)

[a] mean (median) nd = not detected

3.4.11 Miscellaneous Pharmaceuticals

The drugs included here depart from the classification provided by Gielen (2007) due to the limited number of compounds for each use or presence in sludges. Concentrations of the compound salicyclic acid were present at a wide range in the literature on sludges reviewed by Harrison et al. (2006) (Table 66). Cotinine is a metabolite of nicotine and is a marker of human presence in wastewaters and sludges. Digoxigenin is used to induce an immune system response in humans. The median concentration of this marker compound, at 39.8 ng/g TS, was relatively low in the EPA TNSSS, as was the median concentration of 19.9 ng/g TS for the ovulation inhibitor norgestimate.

No biosolids treatment data were found for these pharmaceuticals

Table 66. Concentrations of Miscellaneous Pharmaceuticals in Sludges and Biosolids

Compound	Use/Presence	Sludge source	Concentration (ng/g TS)	Reference
Digoxigenin	steroid immunohisto-chemical marker	Not specified (sludge survey)	57.2 (39.8)	U.S. EPA (2009)
Norgestimate	ovulation inhibitor	Not specified (sludge survey)	27.5 (19.9)	
Salicylic Acid	skin-care	Literature review	0.002–13,740	Harrison et al., (2006)
		Unknown sludge (An urban WWTP)	<LOQ - 253	Spongberg and Witter (2008)
		Unknown sludge (A rural WWTP)	253	
Cotinine	nicotine metabolite	Not specified (sludge survey)	58.0 (13.2)	U.S. EPA (2009)

nd = not detected LOQ = limit of quantitation
[a] mean (median)

3.4.12 Section Summary

The pertinent points from the review of these pharmaceuticals in sludges and biosolids follow.

1. There is a wide range of data available for the different pharmaceuticals that may be present in sludges and biosolids. Some compounds like carbamazepine have been widely characterized, while others have only one or two references in the literature.
2. As a result of there being limited occurrence data for many pharmaceuticals, there are even fewer data available investigating the reduction of these drugs in biosolids treatment processes.
3. Anaerobic digestion is the treatment process for which most data on pharmaceutical concentrations prior to and after treatment have been recorded. Reduction of the compounds appears to be highly specific to each class of pharmaceutical.

3.5 HORMONES AND STEROLS

3.5.1 Hormones

Compounds in this category include both natural and synthetic estrogens, and androgens which clearly have an effect on the human endocrine system. The synthetic estrogens, used for birth control and hormone replacement therapies, and the natural estrogens and androgens are excreted on a daily basis to sewage. Phytosterols are naturally occurring alcohols of steroids, and are present in vegetable oils used in cooking and salads. These can be ingested and excreted, or end up in household grey water during dish washing. The presence of the animal sterols in receiving waters is typically viewed as a marker for sewage contamination. Environmental concerns arising from this group of compounds is mostly focused on the synthetic estrogens, which have potency orders of magnitude higher than the natural estrogens.

Natural and synthetic estrogens found most regularly in wastewater sludges are summarized in Table 67. Concentrations of these compounds in sludges are typically less than 50 ng/g TS,

Table 67. Concentrations of Common Estrogenic Compounds in Sludges and Biosolids

Sludge Source	Concentration (ng/g TS)				Reference
	17α-ethinylestradiol (EE2)	17β-estradiol (E2)	Estriol (E3)	Estrone (E1)	
Sludge Survey	24.9 (25)[a]	34.3 (21.5)	38.7 (24.8)	106 (51.2)	U.S. EPA (2009)
Return activated sludge (2 plants)		nd	nd	nd - 17.5	Tan et al. (2007)
Literature review	<1.5 – 17	4.9 – 49		16 – 27.8	Harrison et al. (2006)
Compost	<5				Gielen (2007)
Waste activated sludge	<5				
Primary sludge	185 ± 185[b]				
Waste activated sludge	3	1.7		7	Andersen et al. (2003)
Primary sludge	<1.5	30		30	
Return activated sludge	<1.5	2.2		5.6	

nd = not detected
[a] mean (median) [b] mean ± standard deviation

although in the U.S. EPA's Targeted National Sewage Sludge Survey (TNSSS, U.S. EPA 2009) the concentration of the natural hormone estrone (E1) had a mean concentration of 106 ng/g TS. The data suggest that estrone (E1) is higher than most of the other common estrogenic compounds. In an analysis of New Zealand Sludge, Gielen reported a mean value of 185 ng/g TS for the synthetic hormone 17α-ethinylestradiol (EE2), however the standard deviation of the mean was also very high indicating a wide spread of experimental values.

There are other natural and synthetic estrogenic compounds present in biosolids as well as those listed in Table 67. Additional estrogens detected in sludges and biosolids in the EPA's TNSSS (EPA, 2009) are provided in Table 68. The concentration of the natural hormone progesterone was the highest of the others observed, with a median concentration of 139 ng/g TS. The median concentrations of the other estrogens were less than 50 ng/g TS.

Table 68. Concentrations of Other Estrogenic Compounds in Sludges and Biosolids (U.S. EPA, 2009)

Natural and Synthetic estrogens	Concentration (ng/g TS)
17α-Dihydroequilin	20.6 (19.4)[a]
β-Estradiol 3-Benzoate	146.9 (23.2)
Equilenin	16 (10.9)
Equilin	34.8 (23)
Mestranol (MEE2)	22.5 (21.4)
Norethindrone	101 (22.3)
Norgestrel	66.5 (42)
Progesterone	323 (139)

[a] mean (median)

Removal of Estrogens

The effect of anaerobic digestion on the removal of the common estrogens in sludge has been investigated by Carballa et al. (2006, 2007a). Data are summarised in Table 69. Data from Carballa et al. (2006) comparing mesophilic and thermophilic digestion indicate that the processes were approximately the same in their ability to remove the estrogens, based on a comparison of the standard deviations of the mean values. Removal efficiencies up to 85% were recorded for both 17α-ethinylestradiol (EE2) and a mixture of estrone (E1) and 17β-estradiol (E2). A second study comparing different solids retention times at the two operating temperatures yielded the unexpected result that a longer retention time provided lower removal efficiencies (Carballa et al., 2007a).

Carballa et al. (2007b) investigated the effect of pre-ozonation of the sludge prior to either mesophilic or thermophilic digestion. At both temperature regimes, the non-ozonated sludge exhibited higher removals of the estrogens than did the pre-ozonated sludge (Table 69).

Table 69. Effect of Anaerobic Digestion Conditions on Removal Efficiency of Estrogenic Compounds

Anaerobic Temperature	Condition	Removal Efficiency (%)		Reference
		Estrone (E1) + 17β-estradiol (E2)	17α-ethinylestradiol (EE2)	
Mesophilic	SRT=20 d	(85 ± 10)%[a]	(85 ± 5)%	Carballa et al. (2006)
Mesophilic	SRT=30 d	70%	40%	Carballa et al. (2007a)
Mesophilic	SRT=20 d	no data	no data	
Mesophilic	SRT=10 d	95%	90%	
Thermophilic	SRT=10 d	(85 ± 5)%	(75 ± 15)%	Carballa et al. (2006)
Thermophilic	SRT=20d	80%	35%	Carballa et al. (2007a)
Thermophilic	SRT=10 d	no data	no data	
Thermophilic	SRT=6 d	90%	90%	
Mesophilic	Non-ozonated	(90 ± 10)%	(90 ±1)%	Carballa et al. (2007b)
Mesophilic	Ozonated	(80±1)%	(85 ±1)%	
Thermophilic	Non-ozonated	(90 ± 1)%	(90 ±1)%	
Thermophilic	Ozonated	(88±1)%	(85 ±1)%	

[a] mean ± standard deviation

Concentrations of androgens in sludges were reported less frequently than estrogens (Table 70). Results from the EPA TNSSS U.S. EPA, 2009) reported three androgens with mean values ranging from a low of 85 ng/g TS for androstereone to a high of 158 for androstenedione. Tan et al. (2007) reported that two target androgens androstereone and the metabolite etiocholanolone were both below detection limits in return activated sludge samples from two different plants.

Table 70. Concentrations of Androgenic Compounds in Sludges and Biosolids

Androgens	Concentration (ng/g TS)	
	Sludge Survey	Return Activated Sludge
Androstenedione	327 (158)[a]	
Androsterone	120 (84.9)	nd
Testosterone	163 (95.2)	
Etiocholanolone (androgen metabolite)		nd
Reference	U.S. EPA (2009)	Tan et al. (2007)

nd = not detected [a] mean (median)

3.5.2 Sterols

Concentrations of plant sterols in sludges and biosolids (Table 71) were among the highest observed in this literature review, with concentrations in the tens of thousands of ng/g TS, and the median concentration of 207,000 ng/g TS reported for β-sitosterol in the EPA's TNSSS (U.S. EPA, 2009). The concentrations for the plant sterols reported in the literature review by Harrison et al. (2006) were much lower than those found in the EPA's TNSSS (U.S. EPA, 2009), and lower even than values reported for return activated sludge and waste activated sludge by Tan et al. (2007).

Table 71. Concentrations of Plant Sterols in Sludges and Biosolids

Compound	Concentration (ng/g TS)		Concentration (ng/g OC)	
	Literature review	not specified Sludge Survey	WAS	Dewatered
Campestanol (5α+5β)	3,000 – 14,000			
Campesterol	6,300	100,900 (46,500)[a]		
Desmosterol		15,650 (10,800)		
Ergosterol		19,830 (12,600)		
Sitostanol (5α-β+5β-β)	14,100 – 93,900			
Sitosterol (β-)	29,600 – 31,100	291,400 (207,000)	138,000	112,000
Stigmastanol; β-Stigmastanol; Stigmastanol (5α+5β)	1,900 – 12,900	168,100 (62,500)	44,600	40,500
Stigmasterol	6,700	321,200 (41,500)		
Reference	Harrison et al. (2006)	EPA (2009)	Kinney et al. (2006)	

[a] mean (median) OC = organic carbon

The concentrations of plant sterols in treated biosolids were documented in Kinney et al. (2006). Composting and heat drying resulted in lower concentrations of the phytosterols in the biosolids than did alternate drying methods or anaerobic digestion (Table 72). For both the β-sitosterol and β-stigmastanol, anaerobic digestion treatment had the highest concentrations of the four treatment methods examined.

Table 72. Concentrations of Plant Sterols following Biosolids Treatment Processes (Kinney et al., 2006)

Biosolids treatment	Concentration (ng/g OC)	
	Sitosterol (β-)	Stigmastanol; Stigmastanol (5α+5β)
Heat drying	110,000	9,310
Compost	50,800-200,000	2,760-17,400
Air drying	257,000	113,000
Anaerobic digestion	554,000	243,000

OC = organic carbon

Concentrations of the animal sterols reported in sludges varied substantially from one reference or source to the next, but were in any case among the highest concentrations observed in this review, as shown in Table 73. The literature review of Harrison et al. (2006) reported the lowest concentrations of the compounds, while the highest values were documented in the EPA's TNSSS (U.S. EPA, 2009), with the fecal indicator 3β-coprostanol having a median concentration of 827,000 ng/g TS. Concentrations of two animal sterols by Kinney et al. (2006) in waste activated sludge and dewatered sludge were intermediate between the EPA data and the review data of Harrison et al. (2006).

Table 73. Concentrations of Animal Sterols in Sludges and Biosolids

Compound	Concentration (ng/g TS)		Concentration (ng/g OC)	
	Literature review	Not specified (Sludge Survey)	Waste Activated Sludge	Dewatered
Cholestanol (5α-); Cholestanol	22,700	680,000 (187,200) [a]		
Cholesterol	57,400		386,000	333,000
Coprostanol; 3β-Coprostanol	216,900	4,367,000 (827,100)	355,000	325,000
Epicoprostanol		1,703,000 (108,000)		
Reference	Harrison et al. (2006)	U.S. EPA (2009)	Kinney et al. (2006)	

[a] mean (median) OC = organic carbon

Concentrations of animal sterols in biosolids treated by different processes are limited to two compounds in the work by Kinney et al. (2006) (Table 74). The results are not consistent for the two compounds. The highest concentration of cholesterol was found in biosolids treated by heat drying, whereas the highest concentration of 3β-coprostanol was observed following anaerobic digestion. For both compounds, however, composting resulted in the lowest concentrations. There are too few data to determine whether composting would produce the lowest concentrations of these compounds.

Table 74. Concentrations of Animal Sterols following Biosolids Treatment Processes (Kinney et al., 2006)

Biosolids treatment	Concentration (ng/g TS)	
	Cholesterol	Coprostanol; 3β-Coprostanol
Heat Drying	402,000	221,000
Compost	19,100-157,000	8,100-72,800
Air Drying	236,000	126,000
Anaerobic Digestion	209,000	1,460,000

3.5.3 Section Summary

The main points of interest in this section follow.

1. The hormones 17α-ethinylestradiol (EE2), estrone (E1) and 17β-estradiol (E2) are among the most frequently characterized compounds of this category in sludges and biosolids. Estrone (E1) is higher in concentration than the other common estrogenic compounds.
2. The concentration of the natural hormone progesterone was the highest of the other hormones observed, with a median concentration of 139 ng/g TS.
3. Concentrations of androgens in sludges were reported less frequently than estrogens, with median concentration of three identified androgens from the EPA sludge survey ranging from 85 to 158 ng/g TS.
4. Concentrations of plant sterols in sludges and biosolids were among the highest observed in this literature review, with concentrations in the tens of thousands of ng/g TS. Composting and heat drying resulted in lower concentrations of the phytosterols in the biosolids; anaerobic digestion treatment had the highest concentrations of the four treatment methods.
5. Concentrations of the animal sterols reported in sludges varied substantially from one reference or source to the next, but were in any case among the highest concentrations observed in this review.
6. Composting resulted in the lowest observed concentrations of both cholesterol and 3β-coprostanol in different biosolids treatment processes.
7. Removal efficiencies up to 85% were recorded for both 17α-ethinylestradiol (EE2) and a mixture of estrone (E1) and 17β-estradiol (E2) in both thermophilic and mesophilic anaerobic digestion.
8. Removal efficiency data for hormones and sterols in other biosolids treatment processes are scarce.

3.6 PERSONAL CARE PRODUCTS

3.6.1 Introduction

In the Review of the State of Knowledge of Municipal Effluent Science and Research: Review of Effluent Substances (Hydromantis et al., 2005), a number of different types of personal care products (PCP) were identified including synthetic musk fragrances, parabens (anti-microbial preservatives), sunscreen agents and insect repellents. In the current literature review focusing on

occurrence and removal in biosolids, data were decidedly more limited. The information retrieved has centred almost exclusively on the presence of the synthetic musk compounds, with only limited data on fluorescent whitening agents. As a result, this Section is devoted primarily to a discussion of the fragrance compounds.

Two main classes of fragrance compounds are used in consumer and commercial products, namely the nitro musks and the polycyclic musks. Nitro musks were first used as synthetic replacements for the natural musk obtained from glands of the male musk deer (Lee et al., 2003). Polycyclic musks have now become the most commonly used synthetic musk due to health concerns and concerns over persistence of the nitro musks in the environment. Both classes of musks are used not only for their own unique smell that influences the odour characteristic, but also for enhancing the quality of a fragrance (OSPAR, 2000). The musk compounds are used in fragrances for detergents, fabric softeners, fabric conditioners, cleaning agents, air fresheners, and cosmetics such as soaps, shampoos and perfumes (OSPAR, 2000). Common fragrance compounds in use are found in Table 75.

Table 75. Identification and Formulations of Common Synthetic Fragrance Compounds

Class of Fragrance	Compound (Trade) Name	Chemical name	CAS No.	Molecular weight (Daltons)
Polycylic Musk	HHCB (Galaxolide)	(1,3,4,6,7,8-hexahydro-4,6,6,7,8-hexamethylcyclopenta-ç-2-benzopyran)	1222-05-5	258.4
	AHTN (Tonalide)	7-acetyl-1,1,3,4,4,6-hexamethyl-1,2,3,4-tetrahydronaphthalene	1506-02-1	258.4
	ATII (Traseolide)	5-acetyl-1,1,2,6-tetramethyl-3-isopropylindan	68140-48-7	258.4
	ADBI (Celestolide)	4-acetyl-1,1-dimethyl-6-*tert*-butylindan	113171-00-1	244.3
	AHMI (Phantolide)	6-acetyl-1,1,2,3,3,5-hexamethylindan	15323-35-0	244.3
	DPMI (Cashmeran)	6,7-dihydro-1,1,2,3,3-pentamethyl-4(5*H*)-indanone	33704-61-9	206.3
	OTNE (Iso E super)	Ethanone, 1-(1,2,3,4,5,6,7,8-octahydro-2,3,8,8-tetramethyl-2-naphthalenyl	54464-57-2	234.4
Nitro Musk	Musk Xylene	1-*tert*-butyl-3,5-dimethyl-2,4,6-trinitrobenzene	81-15-2	297.2
	Musk ketone	4-*tert*-butyl-3,5-dinitro-2,6-dimethylacetophenone	81-14-1	294.3

Many products formulated with fragrance compounds (shampoos, soaps, cleaning products, fabric softeners are contacted with water through bathing and laundry, with subsequent release to municipal sewers. Synthetic musks are generally refractive (non-biodegradable) and highly lipophilic (high octanol:water partition coefficient) (Daughton and Ternes, 1999). At a municipal

wastewater treatment plant, these properties cause the compounds either to be discharged in treated wastewater effluents, or to accumulate in wastewater residual solids. Biodegradation would play only a minor role, if at all, in elimination of the compounds in wastewater treatment.

Polycyclic musks have now become the most commonly used synthetic musk due to concerns over environmental persistence and health issues related to the nitro musks. Peck and Hornbuckle (2004) identified a number of health concerns related to nitro musks, including estrogenic activity and accumulation in human adipose tissue and breast milk.

3.6.2 Fragrance Occurrence Data

Concentrations of polycyclic and nitro musk fragrances from a survey of Canadian biosolids samples are presented in Table 76 (Lee et al., 2003). Galaxolide (HHCB) and Tonalide (AHTN) were the two predominant polycyclic musks, with median concentrations in raw sludge samples of 11,850 and 8,005 ng/g TS, respectively. Traesolide (ATII) was observed with a median value of 1,345 ng/g TS, approximately an order of magnitude lower than the HHCB or AHTN. The remaining two polycyclic musks investigated, Celestolide (ADBI) and Phantoloide (AHDI or AHMI) were substantially lower at 175 and 110 ng/g TS, respectively. The highest concentration of HHCB was in a digested sludge sample from the Toronto Humber Wastewater Treatment Plant, at 26,700 ng/g TS, while the highest for AHTN was 20,600 from the Calgary Bonnybrook facility. With respect to the nitro musks analyzed, musk xylene was found at the highest concentration, 422 ng/g TS, in a sample of raw sludge from Montreal, while musk ketone was consistently found at higher concentrations than musk xylene, particularly in the raw sludge samples (median value of 144.5 ng/g TS).

Concentrations of the polycyclic musks increased from raw sludge to anaerobically digested sludge, suggesting that no reduction was occurring to due anaerobic biodegradation. Anaerobic digestion had an apparent beneficial reduction of the nitro musk compounds with Musk ketone and Musk xylene having median concentrations of 4.5 and 3.3 ng/g TS, respectively in the digested sludge samples (Lee et al., 2003).

Additional concentration data are provided in Table 77 for polycyclic musks and Table 78 for nitro musk compounds. Data provided by Smyth et al. (2007) is a survey of five wastewater treatment plants in the Grand River watershed of Ontario, while data provided by Yang and Metcalfe (2005) is from the Peterborough, ON facility. Data compiled by Webber and Sidwha (2005) are a summary of the data of Lee et al. (2003) presented above in Table 76. The concentration profiles in Tables 77 and 78 follow those in Table 76. Of the polycyclic musks, HHCB and AHTN were present at the highest concentrations, followed by ATII. Concentrations of ADBI and AHMI were of similar magnitude but much lower than the other polycyclic musks identified. The musk DPMI was not detected in the five Ontario plants tested by Smyth et al. (2007). Concentrations of the polycyclic musks from the Peterborough facility (Yang and Metcalfe, 2005) were substantially lower than those identified by Smyth et al. (2007) and Lee at al. (2003). It is not clear whether there differences in magnitude were due to differences in site-specific inputs, differences in analytical procedures, or other unidentifiable factors.

Table 76. Fragrance Concentrations in Canadian Municipal Sludges (Lee et al., 2003)

Biosolids Source	Fragrance Concentration (ng/g TS)						
	Galaxolide (HHCB)	Tonalide (AHTN)	Celestolide (ADBI)	Phantolide (AHDI or AHMI)	Traesolide (ATII)	Musk Xylene (MX)	Musk Ketone (MK)
Raw sludge (Adelaide)	4850	6150	170	<30	1270	33.2	142
Raw sludge (Toronto (North))	21200	8710	220	730	2070	12.8	24.3
Raw sludge (Burlington)	15200	8650	230	120	1650	14.4	17.2
Raw sludge (Edmonton (Goldbar))	6390	8060	130	110	1260	7.2	158
Raw sludge (Montreal (MUC-PSI))	1340	5360	90	80	1000	422	191
Raw sludge (Ottawa)	13200	8650	180	90	1130	46.1	245
Raw sludge (Quebec City)	10500	7300	160	<30	1470	341	364
Raw sludge (Regina)	9070	7260	170	80	1190	144	347
Raw sludge (Saskatoon)	7480	7950	140	130	1360	4.2	94.6
Raw sludge (Toronto (Ashbridges Bay))	21800	11000	230	110	2200	12.6	106
Raw sludge (Toronto (Humber))	17400	7020	200	70	1330	20.3	147
Raw sludge (Toronto (Highland Creek))	20900	10000	390	340	2100	17.6	36.7
Anaerobic digested (Burlington)	12000	8010	190	80	1360	3.3	3.7
Anaerobic digested (Calgary (Bonnybrook))	20800	20600	570	130	4150	5.1	7.3
Anaerobic digested (Calgary (Fish Creek))	18100	18500	480	180	3250	3.9	4.1
Anaerobic digested (Edmonton (Goldbar))	17800	18600	350	150	3680	2.9	6.4
Anaerobic digested (Guelph)	14500	14900	350	130	2180	1.8	2.4
Anaerobic digested (Ingersoll)	4460	6270	160	60	1200	2.3	2.8
Anaerobic digested (Ottawa)	18800	16700	370	130	3080	2.7	4.9
Anaerobic digested (Regina)	12600	12000	320	120	1870	2	4.1
Anaerobic digested (Saskatoon)	8890	9440	180	110	1650	3.3	4.8
Anaerobic digested (Toronto (Ashbridges Bay))	24300	12400	300	120	2290	13	8.3
Anaerobic digested (Toronto (Humber))	26700	12300	310	110	1610	6.9	4.5
Digested sludge (Toronto (North))	24500	12100	220	90	2330	3.8	7.2
Anaerobic digested (Vancouver)	9580	9050	260	60	1240	1.4	1.4
Anaerobic digested (Waterloo)	7340	12700	490	90	1810	2.9	2.2
Anaerobic digested (Windsor)	7810	9510	370	150	1380	3.4	36.7
Median concentration raw	11850	8005	175	110	1345	18.95	144.5
Median concentration anaerobic digested	14500	12300	320	120	1870	3.3	4.5

Table 77. Polycyclic Musk Compounds in Canadian Biosolids Samples

Sludge Source	Fragrance Concentration, ng/g						Reference
	Galaxolide (HHCB)	Tonalide (AHTN)	Celestolide (ADBI)	Phantolide (AHDI or AHMI)	Traesolide (ATII)	Cashmeran (DPMI)	
Raw sludge #1	12000	3550	100	79.6	681	nd	Smyth et al. (2007)
Aerobic Digested sludge #1	9430	2110	67.3	57.4	465	nd	
Raw sludge #2	33700	5860	207	114	1420	nd	
Aerobic Digested sludge #2	40300	8490	255	162	1890	nd	
Raw sludge #3	28400	8020	209	182	1400	nd	
Anaerobic Digested sludge #3	42000	10400	280	201	1910	nd	
Raw sludge #4	30200	8210	208	209	1500	nd	
Anaerobic Digested sludge #4	55500	13800	424	432	2880	nd	
Raw sludge #5	42600	11500	456	191	1630	nd	
Anaerobic Digested sludge #5	46300	10500	510	441	1720	nd	
Raw sludge (Canadian survey)	1300 – 22000 (9800) [a]	5400 – 11000 (7600)	130 – 390 (190)	70 – 340 (110)	1000 – 2100 (1300)	na	Webber and Sidwha (2005)
Digested sludge (Canadian survey)	4500 - 25000 (15000)	6300 - 21000 (12000)	160 – 570 (320)	60 - 180 (120)	1200 – 4200 (1900)	na	
Raw sludge	3303	720	23.6	20.1	198.5	31.4	Yang and Metcalfe (2005)
Return activated sludge	3310	776	29.7	27.1	264	46.9	
Digested sludge	6788	1349	51.2	33.8	413	57.3	
median raw sludge (n=7)	29300	6940	208	148	1410		
median aerobic sludge (n=2)	24870	5300	161	110	1178		
median anaerobic sludge (n=4)	44150	10450	352	317	1815		

na = not analysed; nd = not detected

[a] median value in parentheses

Table 78. Nitro Musk Compounds in Canadian Biosolids Samples

Sludge Source	Fragrance Concentration, ng/g					Reference
	Musk Xylene (MX)	Musk Ketone (MK)	Musk Ambrette (MA)	Musk Moskene (MM)	Musk Tibetene (MT)	
Raw sludge #1	64.6	115	44	nd	nd	Smyth et al. (2007)
Aerobic Digested sludge #1	18.9	45.3	31.3	nd	nd	
Raw sludge #2	96.9	290	nd	151	19.5	
Aerobic Digested sludge #2	25.1	242	nd	6	67.2	
Raw sludge #3	78.2	116	nd	nd	nd	
Anaerobic Digested sludge #3	61	8.16	nd	nd	nd	
Raw sludge #4	59.8	148	11.5	nd	nd	
Anaerobic Digested sludge #4	81.5	11.2	7.6	nd	nd	
Raw sludge #5	25	226	93.2	nd	nd	
Anaerobic Digested sludge #5	3.4	27.6	nd	nd	nd	
Raw sludge (Canadian survey)	4 – 422 (16)[a]	17 – 364 (124)				Webber and Sidwha (2005)
Digested sludge (Canadian survey)	1 – 7 (3)	1 – 37 (5)				
Raw sludge	76.6	39.8	nd	nd	nd	Yang and Metcalfe (2005)
Return activated sludge	115	73.1	nd	nd	nd	
Digested sludge	95.1	53	nd	nd	nd	
median raw sludge (n=7)	70.6	132				
median aerobic sludge (n=2)	22	143.65				
median anaerobic sludge (n=4)	71.25	19.4				

nd = not detected
[a] median value in parentheses

As observed with the data of Lee et al. (2003), aerobic digestion resulted in a decrease in the concentrations of the polycyclic musks in Table 77, whereas increased concentrations of the polycyclic musks were observed following anaerobic digestion.

With respect to nitro musks, the data in Table 78 indicate that Musk ketone and Musk xylene were the dominant compounds, with the Musk ketone present at the highest concentrations. Musk ambrette, Musk moskene and Musk tibetene were detected sporadically in the survey by Smyth et al. (2007), and were not detected in the Peterborough sludge samples by Yang and Metcalf (2005). Data in Table 78 used to estimate median concentrations present a confusing picture, likely due to the limited sample sizes used for assessment. The evaluation indicated that Musk xylene concentrations in raw sludge would be reduced by aerobic digestion, but were essentially unchanged by anaerobic digestion. Conversely, it appeared that Musk ketone would be reduced in concentration by anaerobic digestion of the raw sludge (in agreement with the data from Lee et al. (2003)), whereas aerobic treatment would result in a slight increase in concentration.

Additional polycyclic musk concentration data from other biosolids samples are provided in Table 79. HHCB an AHTN are the most commonly characterized polycyclic musks in these samples. When other polycyclic musk data are presented, the concentrations are much lower than those reported for HHCB and AHTN.

Other Fragrance compounds

In addition to the polycyclic and nitro musks identified above, a number of other fragrance compounds have been identified. These are summarized in Table 80. Most of the additional compounds were identified in the survey of biosolids completed by Harrison et al. (2006).

Kinney et al. (2006) reported high levels of indole (maximum value of 7,000 ng/g TS) and d-limonene (maximum value of 744 ng/g TS) in samples of return activated sludge and dewatered sludge. Otherwise, with the exception of musk ketone, and its derivative, amino musk ketone, the maximum concentrations of the alternate fragrance compounds were less than 100 ng/g TS.

3.6.3 Other Personal Care Product Occurrence Data

Fluorescent whitening agents (FWAs) are chemicals used with textiles and papers to increase the appearance of whiteness by absorbing invisible ultraviolet light and re-emitting it in the blue region of the visible spectrum. Concentrations of FWAs in sludge samples were reported in the survey of Harrison et al. (2006). The compound DAS 1 was observed to have the highest levels in this class of compounds, with a maximum value of 112,000 ng/g TS (Table 81). The agent DSBP was present at approximately half the concentration of DAS 1, while the compound BLS was an approximate order of magnitude lower than the DSBP levels.

Table 79. Concentrations of Polycyclic Musk Compounds in Biosolids from Other Studies

Biosolids Source	Concentration (ng/g TS)						Reference
	Galaxolide (HHCB)	Tonalide (AHTN)	Celestolide (ADBI)	Phantolide (AHDI or AHMI)	Traesolide (ATII)	Cashmeran (DPMI)	
Digested	26,000	4,000					Jones-Lepp and Stevens (2007)
Biosolids Class A	5,000-18,000	2,000-4,000					
Biosolids Class B	10,000	3,000					
Biosolids class A	13-177,000	78-427,000					
Waste activated sludge	3,210 ng/g OC	15,900 ng/g OC					Kinney et al. (2006)
Dewatered sludge	3,150 ng/g OC	16,700 ng/g OC					
Digested sludge	3,068 - 6,788	1,525-1,349					Heidler and Halden (2008)
Not specified	ND – 8,100	ND – 5,100	10–1,100	32–1,800	44–1,100	ND – 332	Harrison et al. (2006)
Unknown sludge (Germany)	4,300 – 13,000 (8,900)	4,000 – 13,000 (8300)	120 – 290 (200)				Webber and Sidwha (2005)

Table 80. Concentrations of Other Fragrance Compounds in Biosolids

Fragrance Compound	Concentration (ng/g TS)		Concentration (ng/g OC)	
	Not specified	Unknown sludge (Germany)	Waste Activated	Dewatered
Musk Xylene (MX)	ND – 32.5	<5		
Musk Ketone (MK)	ND – 1300	<10 – 60		
Acetyl Cedrene	9.0 – 31.1			
Amino Musk Ketone	ND – 362			
Amino Musk Xylene (AMX)	ND –31.5			
Diphenyl Ether	ND – 99.6			
Galaxolide lactone	0.6 – 3.5			
Hexyl salicylate	Trace – 1.5 [52			
Hexylcinnamic Aldehyde (Alpha)	4.1			
Methyl ionone (gamma)	1.1 – 3.8			
OTNE	7.3 – 30.7			
D-Limonene			744	630
Indole			4,290	7,000
Reference	Harrison et al. (2006)	Webber and Sidwha (2005)	Kinney et al. (2006)	

Table 81. Concentration of Fluorescent Whitening Agents in Biosolids (Harrison et al., 2006)

Fluorescent Whitening Agent	Formulation	Concentration (ng/g TS)
BLS	(4,4'-bis(4-chloro-3-sulfostyryl)-biphenyl)	5,400 – 5,500
DSBP	(4,4'-bis(2-sulfostyryl)biphenyl)	31,000 – 50,000
DAS 1	(4,4'-bis[(4-anilino-6-morpholino-1,3,5-triazin-2-yl)-amino]stilbene-2,2'-disulfonate)	86,000 – 112,000

3.6.4 Effect of Biosolids Treatment Processes

Concentration data for fragrance compounds in four biosolids treatment processes, including composting, lime treatment, heat drying, and anaerobic digestion are provided in Table 82 (LaGuardia et al., 2004; Kinney et al., 2006).

Exceptionally high concentrations of HHCB and AHTN were observed in the anaerobically digested sample reported by Kinney et al. (2006). A heat-dried sludge sample reported by LaGuardia et al. (2004) had low concentrations of both HHCB and AHTN. In the study by Kinney et al. (2006), the composted and heat dried samples exhibited lower concentrations of d-limonene than did sludges produced by other drying procedures or by anaerobic digestion. Indole concentrations suggested that there was no significant effect between processes on compound reductions. There are insufficient data reported in the literature to determine the effectiveness of

the different treatment processes for reductions of the compounds.

Table 82. Comparison of Fragrance Compound Concentrations in Biosolids Treatment Processes

Treated Biosolids	Concentration (ng/g TS)				Reference
	Galaxolide (HHCB)	Tonalide (AHTN)	D-limonene	Indole	
Compost	47-12,300 (ng/g OC)	281-11,600 (ng/g OC)	255-705 (ng/g OC)	4,210-38,200 (ng/g OC)	Kinney et al. (2006)
Compost-B	7,000	5,600			LaGuardia et al. (2004)
Lime-A	12,400	7,400			
Heat dry	3,900 (ng/g OC)	11,000 (ng/g OC)	520 (ng/g OC)	20,700 (ng/g OC)	Kinney et al. (2006)
Heat-A	1,100	400			LaGuardia et al. (2004)
Air dry	21,900 (ng/g OC)	43,900 (ng/g OC)	2,120 (ng/g OC)	19,400 (ng/g OC)	Kinney et al. (2006)
Anaerobic digestion	554,000 (ng/g OC)	1,340,000 (ng/g OC)	3,340 (ng/g OC)	21,300 (ng/g OC)	
Anaerobic digestion -A	17,900	9,000			LaGuardia et al. (2004)
Anaerobic digestion –B	11,400	5,400			
Anaerobic digestion –E	10,200	6,600			

3.6.5 Removal of Personal Care Products during Biosolids Treatment Processes

There are few data available on calculated removal efficiencies of fragrances during biosolids treatment. Studies by Carballa et al. (2007a, 2007b) focusing on treatment using anaerobic digestion are summarised in Table 83.

Table 83. Removal Efficiencies of Two Polycyclic Musks by Anaerobic Digestion (Carballa et al., 2007a)

Anaerobic Sludge Type	Solids Retention Time (d)	Removal Efficiency	
		Galaxolide (HHCB)	Tonalide (AHTN)
mesophilic	30	60%	50%
	20	65%	60%
	10	70%	62%
thermophilic	20	70%	75%
	10	75%	85%
	6	80%	80%
Average		(69 ± 9)%	(63 ± 14)%

The data presented in Table 83 are not intuitive as they seem to indicate that the removal

efficiencies decline as the solids retention time (SRT) in the digester increases. Typically longer SRT values provide greater time for acclimation and biodegradation to occur in a reactor. In general, the data indicate that removal efficiencies of the two polycyclic fragrances are similar, with HHCB being slightly more degradable than AHTN under mesophilic conditions. Removal efficiencies were nearly identical under thermophilic treatment.

Table 84 summarizes the effect of pre-ozonating sludge prior to anaerobic digestion under mesophilic and thermophilic conditions (Carballa et al., 2007b). Under mesophilic conditions, pre-ozonation of the feed sludge had a positive influence on the removal efficiency of the two musk compounds, with improvement of approximately 20 percentage points. In thermophilic treatment, the data appear to indicate that pre-ozonation resulted in lower removal efficiencies for the two musks. The cause of lower removal following pre-ozonation is unclear.

Table 84. Effect of Pre-Ozonation on Anaerobic Digestion of Two Polycyclic Musks (Carballa et al., 2007b)

Sludge Type	Operating condition and SRT	Removal Efficiency	
		Galaxolide (HHCB)	Tonalide (AHTN)
anaerobic digestion - mesophilic	Non-ozonated 20 d	(65 ±5)%	(60 ± 5)%
anaerobic digestion - mesophilic	Ozonated 20 d	(85±0)%	(82±2)%
anaerobic digestion - thermophilic	Non-ozonated 10 d	(78 ± 10)%	(82 ± 1)%
anaerobic digestion - thermophilic	Ozonated 10 d	(69±2)%	(30±3)%

Studies conducted by Carballa et al. (2007a, 2007b) with spiked laboratory anaerobic reactors indicate digestion results in reduced concentrations of the HHCB and AHTN. The laboratory results disagree with the field data compiled by Lee and Peart (2002), Smyth et al, (2007, and Webber and Sidwha (2005) and Yang and Metcalfe (2005).

3.6.6 Section Summary

Important points regarding personal care products in sludges are as follows.

1. Polycyclic musks are present at higher concentrations than nitro musks.
2. HHCB and AHTN are the predominant polycyclic musks, followed by ATII.
3. The two main nitro musks identified in sludge samples were Musk ketone and Musk xylene.
4. Full-scale anaerobic digestion does not appear to reduce concentrations of polycyclic musks in sludges, with concentrations in the digested sludges higher than in the raw sludge. Laboratory spiked studies indicate reduction in AHTN and HHCB concentrations are possible.
5. Aerobic treatment conversely, appears to cause a reduction in concentrations of polycyclic musks.
6. There are insufficient data reported in the literature to determine the effectiveness of the different biosolids treatment processes for reductions of the compounds.
7. Data compiled in this review are focused on fragrance compounds, with negligible focus on other PCP compounds identified in MWWE Science and Research document.

3.7 METALS AND METALLOIDS

3.7.1 Introduction

Concentrations of metals in sludges and biosolids have been of concern for decades because of the use of biosolids as a soil amendment in agriculture and silviculture, as well as in recreational uses in parks and golf courses. Concentrations of metals have been reported for decades, and have been well documented in other surveys dating back to the early 1970s (e.g., Leeper, 1972; Page 1974) and later in the 1980s and 1990s (e.g., Monteith et al. 1987, Canviro Consultants Ltd. 1988; Webber and Nichols, 1995) and more recently (XCG, 2007). The intent of this review is not to duplicate these earlier efforts given the project schedule, but to retrieve and expand the body of literature in a limited review of more recent data.

The substances included in this review include a number of elements that include both metals and metalloids, such as arsenic and selenium, which will be referred to as "metals" for simplicity. Metals accumulate in sludges at wastewater treatment when they are discharged by domestic and industrial sources. Aluminum and selenium are present in personal care products. Copper, lead and zinc can enter wastewater from plumbing. Concerns with the metals in biosolids are related to their potential uptake by agricultural crops or foraging animals. Because of the diversity of the metals in this class of contaminants, concerns with respect to human health can be varied. Mercury, cadmium and lead are neurotoxins for example, while cadmium has also been linked to kidney disease.

3.7.2 Occurrence Data

A comprehensive survey of concentrations of metals in the U.S. was recently released by the EPA (2009) in the Targeted National Sewage Sludge Survey (TNSSS) report. Concentrations from this and other recent surveys are presented in Table 85. The EPA survey provides a list of both commonly reported metals and those that are less well documented. The data provided in Table ME1 for the rows of metals below selenium are not as reported in the source table from the EPA (2009) report (Table B-7). In comparison with that report's Table B-6, the values of Table B-7 have been shifted by one row (Stevens, 2009). The effect is that in Table B-7, the reported concentration for thallium is almost two orders of magnitude higher than that for sodium, and the concentration of zinc is less than those for the uncommon elements vanadium and yttrium. As a result, the values from Table B-7 following selenium by row have been shifted upwards by one row for compiling in Table ME1, and the value for zinc, which is then missing, was taken as the value reported as "Aggregated Data" in Table B-6 of the EPA (2009) report.

Concentrations of metals common to the three surveys provided in Table 85 are relatively consistent, on an order of magnitude basis, between the three studies. In the TNSSS, (U.S. EPA, 2009), aluminum and iron are present at the highest concentrations because they are metals frequently used as flocculants and precipitants for phosphorus in wastewater treatment. Zinc and copper are among the next highest concentrations, at median concentrations of 764 and 446 mg/kg respectively. Data provided by Ruel et al. (2008) are in agreement with zinc and copper present at the highest concentrations of the metals analysed. Zinc and copper were not among the metals in sludges reported by XCG (2007), in which the metals with the highest concentrations

were chromium and lead. In the EPA (2009) report, the median concentration of mercury (0.83 mg/kg TS) is higher than for the less frequently reported beryllium and thallium 0.27 and 0.13 mg/kg TS, respectively.)

Table 85. Concentrations of Metals in Sewage Sludges and Biosolids.

Metal/Metalloid	Concentration (mg/kg)		
	sewage sludge survey	17 Canadian sludges (1995-1998)	anaerobic digestion, limed, drying
Aluminum (Al)	13,480 (11,200)[a]		
Antimony (Sb)	2.26 (1.42)		
Arsenic (As)	6.76 (4.95)	0.6 - 4 (2)[b]	not reported
Barium (Ba)	572 (452)		
Beryllium (Be)	0.38 (0.27)		
Cadmium (Cd)	2.48 (1.72)	0.2 – 13 (5)	3.5 ± 21[c]
Chromium (Cr)	78.2 (30.6)	15 – 1,800 (82)	43.2 ± 38
Copper (Cu)	559 (449)		335 ± 338
Iron (Fe)	24,740 (13,250)		
Lead (Pb)	74.0 (44.4)	4.6 – 186 (75)	71 ± 70
Mercury (Hg)	1.27 (0.83)	0.5 - 5 (2.6)	3 ± 2.3
Molybdenum (Mo)	15 (11)		
Nickel (Ni)	47.4 (22.8)	8 – 68 (36)	28.6 ± 38
Selenium (Se)	7.1 (6.2)		
Silver (Ag)	31 (22)		
Thallium (Th)	0.17 (0.13)		
Tin (Sn)	43.5 (36.2)		
Titanium (Ti)	221 (80.9)		
Vanadium (V)	33.9 (11.6)		
Yttrium (Y)	4.55 (3.54)		
Zinc (Zn)	970 (764)		875 ± 1005
Referece	EPA 2009)	XCG (2007)	Ruel et al. (2008)

[a] mean (median) [b] range (median) [c] mean ± standard deviation
for cells in grey shading see text above for explanatory note

3.7.3 Organotin compounds

Concentrations of organotin compounds in sludges have received some attention because of their toxic properties to aquatic organisms. Table 86 provides concentration data for organotin compounds from two publications. Concentrations of the organtins in the earlier literature provided by Fent (1996) are higher than those reported in Webber and Sidwha (2005) for data from the North Toronto plant. In the Toronto data, the monobutyltin and dibutyltin appear to be present at a slightly higher concentration than tributyltin. The data of Fent (1996) include the compound triphenyltin, present at concentrations similar to the butyltin compounds. The survey of sludges by Ruel et al. (2008) listed a concentration of tributyl tin as 0.09 ± 0.05 mg/kg TS, which is similar to the values presented by Webber and Sidwha (2005), but lower than those of Fent (1996).

The data of Webber and Sidwha (2005) comparing the butyltins in raw and digested sludges form the North Toronto plant indicate that the compounds are not reduced by anaerobic degradation. Conversely, Fent (1996) identified that some biodegradation of tributyltin was possible (up to a maximum of 30%) based on laboratory studies of aerobic and anaerobic digestion under both mesophilic and thermophilic conditions.

Table 86. Concentrations of Organotin Compounds in Sludges

Organotin compound	Concentration (mg/kg TS)			
	Survey 25 Swiss plants	Literature	Raw sludge (16 results)	Digested sludge (18 results)
Triphenyltin	0.50	0.04 - 3.4	not reported	not reported
Tributyltin	1.1	0.1 - 3.4	0.014 – 0.176 (0.095)[a]	0.072 – 0.502 (0.111)
Dibutyltin	1.5	0.04 - 4.8	0.076 – 0.336 (0.146)	0.098 – 0.424 (0.264)
Monobutyltin	0.5	0.02 – 6.9	0.113 – 0.205 (0.144)	0.211 – 0.357 (0.254)
Total butyltin	not reported	not reported	0.240 – 0.524 (0.388)	0.424 – 1.080 (0.616)
Reference	Fent (1996)		Webber and Sidwha (2005)	

[a] range (median)

3.7.4 Section Summary

The main points of interest from this survey follow.

1. The concentration database for metals and metalloids is limited because this review was focused on data from the year 2000 on, and much of the documented research on metals occurred previously.
2. After iron and aluminum, the metals of highest concentration are zinc and copper, two metals commonly used in household plumbing.
3. There are few data characterizing concentrations of elements such as selenium, thallium, antimony and molybdenum and others in biosolids.
4. Organotin compounds are present in sludges at low concentrations of less than 1 mg/kg TS (less than 1,000 ng/g TS).
5. Limited data suggest the organotin compounds are not reduced in concentration by anaerobic digestion.

3.8 OTHER SUBSTANCES

3.8.1 Introduction

This section brings together compounds which were not readily included in the previous sections. The major groupings include the polyaromatic hydrocarbons (PAHs) and

polychlorinated polyaromatic compounds (biphenyls (PCBs); dibenzofurans (PCDFs); and dibenzo-p-dioxins (PCDDs)). PAHs are a product of carbon combustion, and enter the environment from volcanoes, forest fires, residential wood burning, and exhaust from automobiles and trucks (NRCC, 1983). Atmospheric deposition, and road oils and exhaust particulates are thus major routes to wastewater treatment via combined sewers. Food cooked at high temperatures (e.g., grilling of barbecuing) may also produce PAHs, which may then be discharged with dishwater. PCBs were once widely used in a variety of products such as electrical transformer fluids, but their manufacture and use have been phased out. PCDDs and PCDFs result from combustion of products consisting of chlorinated organics (e.g. polyvinyl chloride plastics) and as a by-product of pentachlorophenol production. Atmospheric deposition of these chlorinated substances is likely a major contributor in wastewater treatment. Health concerns related to both these classes of compounds are their potential human carcinogenic properties.

As with certain other groups of contaminants reviewed herein, the PAHs and polychlorinated aromatic compounds have received considerable attention in past reviews (e.g. Canviro Consultants, 1988) and thus this section is intended to provide a more recent update on these compounds.

3.8.2 Occurrence of Polyaromatic Hydrocarbons

Concentrations of the PAHs in sludges are provided in Table 87. Based on the survey of Canadian sludges (XCG, 2007), these compounds have median concentrations typically lying in the range of 100 to 1500 ng/g TS. The simplest PAHs, naphthalene and phenanthrene, consisting of two and three fused benzene rings, respectively, have the highest median concentrations of the PAHs in the Canadian survey (XCG, 2007) at 2,700 (phenanthrene) and 1,500 (naphthalene) ng/g TS. The literature review of Harrison et al. (2006) demonstrated that the maximum concentrations of the PAHs could be higher than those summarized by XCG (2007), with the upper range of naphthalene, methylnaphthalene isomers and benzo(a)anthracene at or above 100,000 ng/g TS. The U.S. EPA's TNSSS (EPA, 2009) included only two PAHs on its list of target analytes. Median concentrations of benzo(a)pyrene and 2-methylnaphthlene were 302 and 200 ng/g TS, respectively. Concentrations of four PAHs reported by Kinney et al. (2006) in waste activated sludge and dewatered sludge were less than 200 ng/g organic carbon.

Concentrations of PAHs following biosolids treatment processes were provided by Kinney et al. (2006) and appear in Table 88. For the lower molecular weight PAHs anthracene and phenanthrene, composted and air dried biosolids have apparent lower concentrations than biosolids produced by heat drying or after anaerobic digestion. This trend did not follow through in the two higher molecular weight PAHs fluoranthene and pyrene, for which there was no discernible difference between composting air drying and heat drying. The anaerobic digested sludge had the highest concentrations of the four PAHs examined. There are too few data to determine whether anaerobic digestion is the least effective biosolids treatment for reduction of PAHs.

Table 87. Concentrations of Polyaromatic Hydrocarbons in Sludges

Compound	Concentration (ng/g TS)				
	19 Canadian sludges	Sludge survey	Literature review	WAS[c]	Dewatered sludge[c]
Acenaphthene	nd - 3,000 (400)[a]		nd - 6,600		
Acenapthylene	nd – 3,400 (100)		3.6 - 300		
Anthracene	3 – 3,300 (200)		nd - 44,000	92	74
Phenanthrene	900 - 14,000 (2,700)		<10 - 44,000	159	166
Benzo(a)anthracene			nd - 99,000		
Chrysene			nd - 32,400		
Benzo(a)anthracene + Chrysene	170 – 36,000 (1,100)				
Benzo(b)fluoranthene + Benzo(k)fluoranthene	130 – 39,000 (700)		6 - 34,200		
Benzofluorene congeners			nd - 8,100		
Benzo(g,h.i)perylene	30 - 15,000 (300)		nd - 12,900		
Benzo(a)pyrene	50 - 25,000 (300)	661 (320)[b]			
Benzopyrene congeners			nd - 24,700		
Dibenzo(ah)anthracene	nd - 5,100 (20)				
Dibenzoanthracene congeners			nd - 13,000		
Fluoranthene	250 - 27,000 (1,000)		nd - 60,000	166	166
Fluorene	nd - 3,300 (800)		<10 - 8,100		
Indenol(1,2,3-cd)pyrene	nd - 15,000 (200)		nd - 9,500		
Naphthalene	80 - 13,000 (1,500)		nd - 6,610,000		
Perylene			nd - 69,300		
Pyrene	260 - 24,000 (1,300)		10 - 37,100	186	169
2-methylnaphthylene		449 (200)			
Methylnaphthalene isomers			nd - 136,000		
Methylphenanthrene isomers			nd - 37,400		
Reference	XCG (2007)	EPA (2009)	Harrison et al. (2006)	Kinney et al. (2006)	

nd = not detected
[a] range (median) [b] mean (median) [c] ng/g organic carbon (OC)

Table 88. Concentrations of Polyaromatic Hydrocarbons following Biosolids Treatment Processes (Kinney et al., 2006)

Biosolids Process	Concentration (ng/g OC)			
	phenathrene	anthracene	fluoranthene	pyrene
Heat drying	1,090	324	1,090	1,310
Compost	176 - 376	56 - 253	744 – 2,470	43 – 1,420
Air drying	535	359	1,150	1,110
Anaerobic digestion	5,430	1,000	2,980	2,320

No removal efficiency data for PAHs were identified in the literature review.

3.8.3 Occurrence of Polychlorinated Polyaromatics

Concentrations of PCBs, PCDDs and PCDFs in sludges are summarized in Table 89 in two different units of expression. Some reports list the PCDDs and PCDFs in terms of toxic equivalents of the 2,3,7,8-tetrachlorodibenzo-p-dioxin, the most toxic congener of this class of compounds. For the literature surveyed, the range and means reported from different countries appear to be very similar, with mean values in the range of 0.020 ng TEQ/g TS. Concentrations of the PCDDs and PCDFs as total congener concentrations were documented by XCG (2007), with mean values of the total congeners at least an order of magnitude higher than the TEQ-based concentrations.

For the PCB data summarized by XCG (2007), the upper end of the concentration range, at 2,027 ng/g TS, is substantially higher than the maximum values reported in sludges from European countries. The mean concentration likewise is higher in the XCG (2007) report than for the sludges from Norway, Sweden and Germany as documented by Jaganyi (2007).

3.8.4 Section Summary

The main points of interest from this section follow.

1. The upper range of naphthalene, methylnaphthalene isomers and benzo(a)anthracene were at or above 100,000 ng/g TS in the literature review of Harrison et al. (2006), although a survey of Canadian sludges resulted in median concentrations typically lying in the range of 100 to 1,500 ng/g TS.
2. The simplest PAHs, naphthalene and phenanthrene, consisting of two and three fused benzene rings, respectively, have the highest median concentrations of the PAHs in the Canadian survey.
3. Data on the effect of biosolids treatment processes on reducing concentrations of PAHs are very limited. For the lower molecular weight PAHs anthracene and phenanthrene, composted and air dried biosolids have apparent lower concentrations than biosolids produced by heat drying or after anaerobic digestion. The anaerobic digested sludge had the highest concentrations of the four PAHs examined.
4. For the literature surveyed, the range and means of the PCDDs and PCFDs reported from different countries appear to be very similar, with mean values in the range of 0.020 ng TEQ/g TS.
5. Concentrations of total PCBs listed in Canadian sludge samples appeared to be higher than corresponding sludge samples from Europe.

Table 89. Concentrations of Polychlorinated Polyaromatics in Sludges

Sludge Source	Concentration (ng TEQ/g TS)			Concentration (ng/g TS)			
	Polychlorinated Dibenzo-p-Dioxins and Furans (PCDD/Fs)			Total Polychlorinated Dibenzo-p-Dioxins (PCDD)	Total Polychlorinated Dibenzofurans (PCDFs)	Total Polychlorinated Biphenyls (PCBs)	
Denmark sludge (not specified)	0.0007 - 0.055 (0.021)[a]	0.010 - 0.034 (no mean)[a]					
Germany sludge (anaerobic digested)	0.0007 - 1.21 (0.020 - 0.040)	no range (0.019)					
Germany sludge (not specified)							154 - 340
Spain sludge (anaerobic digested)	no range (0.064)						
UK sludge (not specified)	0.009 - 0.192 (no mean)						
Austria (not specified)	0.008 - 0.038 (0.015)						
Sweden sludge (not specified)	0.00002 - 0.115 (0.020)	0.0057 - 0.115 (no mean)					0.6 - 232 (113)
Norway sludge (not specified)							17 - 100 (42)[a]
Canadian sludge (1995-1998)			0.004 – 0.12 (0.022)[a]	1.1 - 22 (4.1)[a]	0.07 – 4.2 (0.7)[a]	nd – 2,027 (345)[a]	
Reference	Jaganyi (2007)	Langenkamp et al. (2001)	XCG (2007)	XCG (2007)			Jaganyi (2007)

[a] range (mean) nd = not detected

4. ANALYSIS OF LITERATURE REVIEW FINDINGS

There is a great disparity in the attention devoted to the occurrence of different micro-constituents in sludges and biosolids. Some compounds have been examined comprehensively, and there is a substantial database on occurrence of these substances. Examples of the well-documented substances include the plasticizer bis(2-ethylhexyl)phthalate, the surfactant nonylphenol and some of its ethoxylates, the synthetic musk fragrances HHCB and AHTN, the flame retardants polybrominated diphenyl ethers, and the bacteriostat triclosan. In other cases however, there is a significant lack of information on concentrations of other substances in sludges and biosolids, including many antibiotics and other pharmaceuticals. The report on the State of Knowledge of Municipal Effluent Science and Research identified many classes of personal care products in treated effluents, including parabens (anti-microbial preservatives), sunscreen agents and insect repellents, for which no occurrence data in sludges or biosolids were identified. Lack of adequate analytical protocols may hinder this effort.

In much of the data reviewed, including other literature reviews, the sludges or biosolids are not specified by type (raw or treated, primary or secondary, etc) which makes the effort of determining if some processes are more beneficial than others in minimizing the concentrations of these substances. Some reviews, which summarize concentration data, group different types of sludges together without regard to the nature of the sludges. There appeared in the review to be evidence that sludges from some countries have lower concentrations than others, which may be reflective of manufacturing or use restrictions.

The treatment process most characterized for ability to reduce contaminant concentrations is the anaerobic process. For example in the studies by Carballa et al. (2006, 2007a, 2007b), concentrations of a variety of contaminants are provided for both raw sludge and sludges digested under different temperature regimes and retention times. From the review it appears that certain micro-constituents can be reduced by anaerobic digestion, while others are recalcitrant, or perhaps even increased in concentration, by anaerobic biotransformation processes.

With respect to different biosolids treatment processes, only the final treated biosolids concentrations are typically documented. Without the accompanying raw sludge concentrations, an evaluation of the effectiveness of the processes becomes more tenuous. In the data provided by Kinney et al. (2006), there appears to be evidence of some reduction in certain biosolids treatment processes such as composting or drying, but without raw sludge data, no firm conclusions can be drawn.

5. RECOMMENDATIONS

Based on the literature review and above assessment, the following recommendations are offered:

1. There is a need to define criteria for what is an adequate database for characterization of contaminants in sludges and biosolids, then to apply the criteria to the compiled data.
2. Where there are insufficient data, the availability of adequate analytical protocols needs to be determined. If they not available, then method development should be set as a priority.
3. If acceptable analytical protocols are available, characterization of sludges and biosolids treatment processes should follow, such as the planned follow-up field investigation accompanying this literature review project. Assessment of biosolids treatment effectiveness requires both raw and treated biosolids samples.
4. Lastly, data produced by this and similar reviews, and by the forth-coming field investigation, need to be transferred out to appropriate agencies and researchers.

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Case for Caution Revisited: Health and Environmental Impacts of Application of Sewage Sludges to Agricultural Land

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Over the past 15 years since the 40CFRPart503 rules were promulgated, there have been many new scientific findings regarding the environmental and health implications of the application of sewage biosolids to agricultural soils. Many of these findings show increased risks, risks that were not assessed as part of the risk assessment that USEPA used as the basis for the standards promulgated in 1993. These new findings support the rational basis for U.S. EPA to revise the federal regulations and for states and municipalities to regulate the application of sewage biosolids in order to protect their citizens and the land-base.

Agricultural soils are a unique and valuable resource. Protecting agricultural soils requires anticipating and avoiding potential harms since once contaminated with persistent pollutants, the damage will remain for the foreseeable future. Once contaminated, stopping the application of pollutants such as metals and many organic chemicals that are in sewage biosolids will not correct the problem. The contamination will remain for decades or centuries. It is thus critical to prevent this essentially permanent degradation.

Current Rules are Based on Outdated and Inadequate Science

As pointed out the by the National Research Council, the risk assessment on which current rules are based was conducted nearly 20 years ago and is outdated. A tremendous amount of new knowledge about the presence and behavior of chemicals and pathogens has been developed in the last decades.

NRC Targets Pathogens in Sludge for Research. Rebecca Renner, 2002.Environmental Science and Technology: Science News - July 24. <http://pubs.acs.org/doi/pdf/10.1021/es022404s>

The U.S. EPA rules for using treated sewage sludge as fertilizer are based on outdated science, according to a report released in July from the National Academies, National Research Council (NRC). The report, which was produced after two years of study, recommends new research to update the rules. In particular, EPA needs to investigate the growing number of complaints about illnesses and even deaths from exposure to Class B sludge.

Under a 1993 Clean Water Act rule, treated sewage sludge, or biosolids, can be applied to land with certain limitations. Pathogen-containing Class B sludge, which makes up the bulk of sludge applied to land, may be used as fertilizer in situations in which public exposure is limited. Class A sludge can be applied on public sites. Of the 5.6 million tons of sewage sludge generated in the United States each year, 60% ends up being applied as fertilizer.

The agency needs to investigate the potential health effects from sludge exposure and find out more about the pathogens in sludge, according to committee chair Thomas Burke, a public health professor at Johns Hopkins University in Baltimore, Md. There is a serious lack of health-related information about populations exposed to treated sludge, adds Burke.

The NRC report also recommends a new national sludge survey to measure sludge contaminants, which would update the previous 1988 survey. This earlier study was unreliable and needs to include newly recognized chemicals of potential concern, including polybrominated biphenyl ether flame retardants, pharmaceuticals, and personal care products such as shampoos and soaps, says the NRC committee. EPA also needs to redo its assessment of the human health risks posed by metals in sludge. The revised risk assessments should reflect the potential for variations in climate, water flow, and sludge characteristics. The report also notes that more rigorous enforcement of the current standards is needed."

Targeted National Sewage Sludge Survey Report Released in 2009. USEPA, <http://earth1.epa.gov/waterscience/biosolids/tnsss-overview.html>

The last EPA survey of sewage sludges nationally occurred in 1988. The EPA 503 rule was based in large part on the levels of contaminants detected in that survey. Many contaminants have emerged since then as being potentially harmful in the environment. This new survey by

EPA provides much-needed information on chemicals likely to be found in sewage sludges across the country.

In 2006 and 2007, the USEPA collected samples of sewage sludge from 74 randomly-chosen wastewater treatment facilities in 35 states. The sampled facilities are considered to be representative of the nation's 3,337 largest treatment facilities. The samples were tested for 145 chemicals, including metals, PAHs, nitrogen, phosphorus, flame retardants (PDBEs), pharmaceuticals, hormones, and steroids.

It is notable that, while the median concentrations of toxic metals, trace elements, and organic chemicals were generally many times lower than the highest concentrations observed, quite high concentrations of one or more chemicals were measured in a substantial fraction of the 74 treatment plants. This survey, while quite informative, is not able to assess variability of sludge composition over time, as the sewage sludge was sampled at a single time point. The survey showed some very high concentrations of specific chemicals at one or more treatment plants, with peak concentration for the following elements being:

Barium	3,460 mg/kg		Mercury	8.26 mg/kg
Fluoride	234 mg/kg		Nickel	526 mg/kg
Molybdenum	132 mg/kg		Copper	2,580 mg/kg
Silver	856 mg/kg		Tin	522 mg/kg
Cobalt	290 mg/kg		Vanadium	617 mg/kg
Iron	299,000 mg/kg		Zinc	8,550 mg/kg
Lead	450 mg/kg			

This list is only a sampling of the inorganic contaminants reported in the survey.

In many cases, the highest contaminant concentrations were found in the smallest wastewater treatment plants included in the survey (1-10 MGD plant). The very high Fe sludge (reported in the list above) also had very high phosphorus, attributable to a tertiary treatment process using iron salts to remove P from wastewater. As tertiary treatment to lower P in treated water is likely to increase in the future, we can perhaps expect to see more sewage sludges with very high Fe content. Although ferric iron is not a toxic metal when mixed into soil, it has been known to be toxic to cattle where sludge was applied directly to pasture.

The high levels of several unregulated or inadequately regulated and potentially toxic metals (e.g., silver, molybdenum, tin) are a concern for land application. It should also be of great concern for land application that the measured concentrations of persistent organic pollutants (POPs), including the brominated fire retardants (PBDEs), and the antimicrobial chemicals (triclosan and triclocarban) are so high in some sludges. These POPs are likely to build up in soils with repeated application, and have the potential to bioconcentrate in foraging animals and therefore in meat and milk. One of the eleven PBDE congeners measured (BDE 209) reached a concentration of 17,000 µg/kg in one sludge, and the highly bioaccumulative BDEs 47 and 99 reached levels as high as 5,000 µg/kg. Triclocarban and triclosan had peak concentrations of 441,000 and 133,000 µg/kg in separate sludges. The impact of these persistent chemicals on soil organisms, the safety of food crops, and the environment is not known at this time because of very limited research on their behavior and toxicity in soil.

The prevalence of a wide array of pharmaceuticals, steroids and hormones, as summarized in the EPA report, is a clear indication that the sewage treatment process does not degrade these organic chemicals effectively, and sewage sludge therefore becomes the repository for a large fraction of the chemicals used commercially and domestically.

New information on the impacts of the regulated contaminants

Endocrine Disruption

New information indicates that some of the handful of metals that are regulated under Part 503 pose risks that were not evaluated in the risk assessment upon which the Part 503 USEPA rules are based. The whole subject of endocrine disruption due to exposure to chemicals in the environment (i.e. our knowledge regarding the disruption to human and animal hormones and reproductive systems posed by a number of chemicals) has developed since those rules were promulgated.

Examples of several of the regulated metals for which new risks have been identified are lead and cadmium. Recent work shows that lead has a number of effects on sperm and may play a role in the rising infertility that is being observed. Cadmium has been shown to mimic estrogen and may be related to increased breast cancer. These metals are contained in all sewage biosolids. The contaminant limits in Part 503 do not include any recognition of these endocrine-disrupting impacts.

Increased seminal plasma lead levels adversely affect the fertility potential of sperm in IVF. Susan Benoff, Grace M. Centola, Colleen Millan, Barbara Napolitano, Joel L. Marmar and Ian R. Hurley, 2003. Human Reproduction, V. 18, No. 2, 374-383

BACKGROUND: Lead remains in high levels in the environment and is known to reduce fertility in animal models, but a direct link between lead exposures and human infertility has not yet been established. METHODS: In a prospective, double-blind study of the metal ion levels and sperm function, semen was obtained from partners of 140 consecutive women undergoing their first IVF cycle. Lead in seminal plasma was determined by atomic absorption spectroscopy. Motile sperm populations were assessed for surface receptors for mannose binding, and the ability to undergo premature ('spontaneous'), and free mannose-induced acrosome reactions. Fertile donor (n = 9) sperm were exposed to exogenous lead during capacitating incubations and then assessed for mannose receptor expression and acrosome loss. RESULTS: Lead levels were negatively correlated with IVF rates. Lead levels were negatively correlated to two of the three sperm function biomarkers (mannose receptors, mannose-induced acrosome reactions). Lead levels positively correlated with the spontaneous acrosome reaction. These findings were mimicked by in-vitro exposure of fertile donor sperm to lead. CONCLUSIONS: Multiple sperm parameters are affected as lead levels rise. Increased lead levels may contribute to the production of unexplained male infertility.

Cadmium mimics the in vivo effects of estrogen in the uterus and mammary gland. Michael D Johnson, Nicholas Kenney, Adriana Stoica, Leena Hilakivi-Clarke, Baljit Singh, Gloria Chepko,

Robert Clarke, Peter F Sholler, Apolonio A Lirio, Colby Foss, Ronald Reiter, Bruce Trock, Soonmyoung Paik, and Mary Beth Martin, 2003. Nature Medicine, 9:1081-1084. Letter Published online: 13 July 2003.

Abstract: “It has been suggested that environmental contaminants that mimic the effects of estrogen contribute to disruption of the reproductive systems of animals in the wild, and to the high incidence of hormone-related cancers and diseases in Western populations. Previous studies have shown that functionally, cadmium acts like steroidal estrogens in breast cancer cells as a result of its ability to form a high-affinity complex with the hormone binding domain of the estrogen receptor1, 2. The results of the present study show that cadmium also has potent estrogen-like activity *in vivo*. Exposure to cadmium increased uterine wet weight, promoted growth and development of the mammary glands and induced hormone-regulated genes in ovariectomized animals. In the uterus, the increase in wet weight was accompanied by proliferation of the endometrium and induction of progesterone receptor (PgR) and complement component C3. In the mammary gland, cadmium promoted an increase in the formation of side branches and alveolar buds and the induction of casein, whey acidic protein, PgR and C3. *In utero* exposure to the metal also mimicked the effects of estrogens. Female offspring experienced an earlier onset of puberty and an increase in the epithelial area and the number of terminal end buds in the mammary gland.”

Cadmium mimics effects of estrogen. NewScientist.com News Service, 13:44, July 14, 2003.

Cadmium is astonishingly good at mimicking the effects of the female sex hormone estrogen, new research on rats has revealed. The discovery raises concerns that the metal, and others like it, could increase the risk of illnesses like breast cancer in people.

Cadmium is widely used in batteries, and is present in cigarette smoke and sewage sludge spread on agricultural land. It is best known for obvious toxic effects on the liver and kidneys.

But new research by Mary Beth Martin's team at Georgetown University in Washington DC shows that, at much lower doses, cadmium can cause very similar effects as estrogen.

Martin gave cadmium to female rats whose ovaries had been removed, so they could not make estrogen themselves. The animals received doses comparable to the level set by the World Health Organization as a tolerable weekly intake for people. The results were unexpectedly striking, with the effects of the cadmium appearing almost identical to those of estrogen.

Denser tissue

Rats given cadmium rapidly developed heavier wombs, denser mammary glands and thicker womb linings - just as they did when given estrogen itself. They also began to make milk, and two genes usually activated by estrogen were switched on.

And when Martin's team gave cadmium to pregnant rats, their female offspring went through puberty sooner and developed denser mammary gland tissue, again matching the effects of estrogen.

Impacts on livestock

Livestock that graze on sludge-amended pastures ingest biosolids that adhere to the forage plants and also ingest soil directly. Particularly in arid conditions, soil can be up to 18% dry weight of a grazing animal's diet. Even where lesser amounts are ingested, recent research has shown impacts to grazing animals from biosolids additions to soils. These impacts include an accumulation of toxic metals in edible body organs, with implications for the human food chain. Additionally, endocrine disruption (reduced testis size) has been documented, with implications for livestock reproduction. There is now evidence that elements in sludge, particularly molybdenum and sulfur, are readily taken up by forages and can lead to Cu deficiency in livestock.

Accumulation of potentially toxic elements by sheep given diets containing soil and sewage sludge. 1. Effect of type of soil and level of sewage sludge in the diet. Hill, J. B. Stark, J. Wilkinson, M. Curran, I. Lean, J. Hall, C. Livesey, 1998. Animal Science, 67:73-86.

Live weight gain was depressed by the addition of sludge to the diet. Levels of cadmium and lead in liver and kidneys increased, with the lead levels approaching the UK statutory limit for human food.

The long-term effect of sludge application on Cu, Zn, and Mo behavior in soils and accumulation in soybean seeds. B.J. Kim, M.B. McBride, B.K. Richards, T.S. Steenhuis, 2007. Plant and Soil, 299:227-236.

Molybdenum and copper uptake by forage grasses and legumes grown on a metal-contaminated sludge site. M.B. McBride, 2005. Communications in Soil Science and Plant Analysis, 36: 2489-2501.

Molybdenum extractability in soils and uptake by alfalfa 20 years after sewage sludge application. M.B. McBride and B. Hale, 2004. Soil Science, 169:505-514.

Molybdenum, sulfur, and other trace elements in farm soils and-forages after sewage sludge application. M.B. McBride, 2004. Communications in Soil Science and Plant Analysis, 35:517-535.

The EPA 503 rule regulated the loading of only 8 heavy metals on agricultural soils. Molybdenum loading on soils is not limited by the 503 rule even though this trace metal presents a well-documented danger for ruminant animals due to its ready uptake into forage legumes, grasses, soybeans and other crops. The 4 research papers cited above demonstrates that molybdenum in land-applied sewage represents a sustained and long-term risk to livestock health from increased molybdenum in forages and soybeans.

Effects of pasture applied biosolids on performance and mineral status of grazing beef heifers. M.E. Tiffany, L.R. McDowell, G.A. O'Connor, F.G. Martin, N.S. Wilkinson, E.C. Cardoso, S.S. Percival and P.A. Rabiansky, 2000. J. Animal Science, 78:1331-1337.

Effects of residual and reapplied biosolids on performance and mineral status of grazing beef steers. M.E. Tiffany, L.R. McDowell, G.A. O'Connor, F.G. Martin, N.S. Wilkinson, S.S. Percival and P.A. Rabiansky, 2002. J. Animal Science, 80:260-269.

Molybdenum and sulfur in forage crops are known to reduce the availability of copper to ruminant animals, and can lead to severe copper deficiency in livestock.

Studies in Florida have revealed that, while molybdenum applied with sewage sludges on bahiagrass was not taken up by the grass to a significant degree, grazing beef cattle nevertheless developed signs of copper deficiency as confirmed by reductions in liver copper stores. This negative effect of sewage sludge on copper availability to the cattle was attributed to high sulfur concentrations in the sludge-amended pastures. The low uptake of molybdenum by grass in that study can be attributed to the low pH of the pasture soils.

Exposure to pastures fertilised with sewage sludge disrupts bone tissue homeostasis in sheep. P. Monica Lind, M. Gustafsson, S.A.B. Hermsen, S. Larsson, C.E. Kyle, J. Orberg and S.M. Rhind, 2009. Science of the Total Environment, 407:2200-2208.

A recent study has shown that male sheep exposed to low levels of pollutants by grazing on pastures fertilized with sewage sludge developed bone tissue abnormalities.

Cellular and hormonal disruption of fetal testis development in sheep reared on pasture treated with sewage sludge. Catriona Paul, Stewart M. Rhind, Carol E. Kyle, Hayley Scott, Chris McKinnell, and Richard M. Sharpe, 2005. Environmental Health Perspectives, 113(11):1580-1587

Fetuses of pregnant sheep reared on sludge-treated pasture had reduced body weight. Male fetus testis were significantly reduced. "These findings indicate that exposure of the developing male sheep fetus to real-world mixtures of environmental chemicals can result in major attenuation of testicular development and hormonal function, which may have consequences in adulthood." This has the potential for impact on fertility.

Movement to groundwater through facilitated transport

New understanding about the movement of contaminants (both chemicals and pathogenic organisms) through soils into groundwater has been developed in recent years. This includes information showing that contaminants may "piggy-back" on other chemicals that move in water (this is termed "facilitated transport"). Thus a chemical which by itself is relatively immobile in soils (such as many metals), can move rapidly through soils when other chemicals are present (such as organic matter in biosolids). In addition, another mechanism that provides for rapid movement of chemicals through soils is that water and the contaminants carried in it can move through soils along preferential flow paths (such as worm holes, root channels or wetting fingers).

Recent short feature articles on these topics prepared by Cornell include *Colloidal transport: the facilitated movement of contaminants into groundwater* (B.K. Richards, J.F. McCarthy, T.S. Steenhuis, A.G. Hay, Y. Zevi, A. Dathe. 2007. Journal of Soil & Water Conservation 62(3)55A-

56A) and *The unintentional secret*. (B.K. Richards, N. Peranginangin, T.S. Steenhuis and L.D. Geohring. 2003. Journal of Soil & Water Conservation, September-October 2003 59(5):104A-105A). By these mechanisms, contaminants can move through the soil and into groundwater much more quickly than predicted in the very limited risk assessment of groundwater transport potential performed to support the Part 503 rules. The rate of contaminant movement predicted by that risk assessment relied on data from a *single* paper based on *test tube* mobility tests from a *single soil type*. No actual field data were used. Furthermore, the transport models employed by that assessment assumed uniform homogenous soils. The risk assessment thus did not account for these common rapid flow phenomena.

Biosolid colloid-mediated transport of copper, zinc, and lead in waste-amended soils. A.D. Karathanasis, D.M.C. Johnson, and C.J. Matocha, 2005. Journal of Environmental Quality, 34(4):1153-1164

A significant increase in the leaching of metals (up to 10,000 times) was measured in a laboratory experiment as a result of the binding of metals to the organic colloids in sewage sludge. "The findings demonstrate the important role of biosolids colloids as contaminant carriers and the significant risk they pose."

Effect of Mineral Colloids in Virus Transport through Saturated Sand Columns. Yan Jin, Ellen Pratt, and Marylynn V. Yates, 2000. Journal of Environmental Quality, 29(2):532-539

The movement of viruses through soils was facilitated by adsorption on to colloidal particles.

Facilitated Transport of Napropamide by Dissolved Organic Matter in Sewage Sludge-Amended Soil. L. Nelson, W. Farmer, C.J. Williams, and M. Ben-Hur, 1998. Journal of Environmental Quality, 27:1194-1200.

Abstract: The application of sewage sludge to agricultural soils is practiced to minimize landfill disposal. Organic matter amendments to soil are generally thought to improve soil quality, but pesticide application to these soils may lead to groundwater contamination problems. The complexation of pesticides with a water-soluble carrier such as dissolved organic matter (DOM) may facilitate chemical movement through soil. Sewage sludge amendments may lead to greater downward movement of organic chemicals if associated with DOM. Napropamide [2-α-napthoxy)-*N*,*N*-diethylpropionamide] was applied to a silt loam soil with (SS) and without (NoSS) sewage sludge application. Laboratory batch equilibrium and soil column studies were performed to determine the potential for herbicide complexation with DOM. Over 98% of the herbicide in soil columns followed typical adsorption and transport behavior as the center of mass of the lower organic matter soil (NoSS) moved twice the depth as that of SS. However, napropamide was detected in the initial leachate eluted from repacked soil columns with steps taken to prevent preferential flow. Napropamide concentrations in the initial leachate of SS were twice that from NoSS with <1.5% of the total applied chemical mass eluting from the bottom of each column. A strong positive relationship was found between napropamide concentration and DOM content in soil leachates. Equilibrium dialysis methods were used to determine that napropamide moving

through the soil columns was complexed with DOM. The results show that DOM can facilitate herbicide movement through soil and that sewage sludge-derived DOM may lead to enhanced chemical transport in sludge-amended soils.

Enhanced Transport of Pesticides in a Field Trial with Treated Sewage Sludge. E. Grager, I. Dror, F. Bercovich, and M. Rosner, 2001. Chemosphere, 44: 805-811

Pesticide leaching in arid field soils was increased by the application of sewage sludge.

Aerosols and human health effects

Health effects from exposure to sewage sludge during land spreading have been reported frequently, but these reports have been considered anecdotal and not confirmatory evidence that illness can result from aerosols released during application. Few studies have actually addressed symptoms related to land application. A study of people living near application sites compared with a control population showed statistically elevated health-related symptoms in the exposed population. Another study of 48 people located near 10 land application sites indicated that chemical irritants and pathogens in sludge may interact to cause symptoms.

Several recent publications have tracked aerosol emissions from fields during sewage sludge (biosolids) application and tillage. DNA-based microbial tracking has proven that wind is a critical factor in the formation and off-site migration of aerosols. Biosolids aerosols of inhalable size (< 10 μm), containing bacteria such as coliforms and Health survey of residents living near farm fields permitted to receive biosolids.

Health Survey of Residents Living near Farm Fields Permitted to Receive Biosolids. Sadik Khuder, Sheryl A. Milz, Michael Bisesi, Robert Vincent, Wendy McNulty, and Kevin Czajkowski, 2007. Archives of Environmental and Occupational Health, 62(1):5-11.

Abstract: The authors studied the health status of residents living in Wood County, OH, near farm fields that were permitted to receive biosolids. They mailed a health survey to 607 households and received completed surveys from 437 people exposed to biosolids (living on or within 1 mile of the fields where application was permitted) and from 176 people not exposed to biosolids (living more than 1 mile from the fields where application was permitted). The authors allowed for up to 6 surveys per household. Results revealed that some reported health-related symptoms were statistically significantly elevated among the exposed residents, including excessive secretion of tears, abdominal bloating, jaundice, skin ulcer, dehydration, weight loss, and general weakness. The frequency of reported occurrence of bronchitis, upper respiratory infection, and giardiasis were also statistically significantly elevated. The findings suggest an increased risk for certain respiratory, gastrointestinal, and other diseases among residents living near farm fields on which the use of biosolids was permitted. However, further studies are needed to address the limitations cited in this study.

Interactions of pathogens and irritant chemicals in land-applied sewage sludges (biosolids). David L Lewis, David K Gattie, Marc E Novak, Susan Sanchez, and Charles Pumphrey , 2002.

BMC Public Health, 2:11

Background: Fertilisation of land with processed sewage sludges, which often contain low levels of pathogens, endotoxins, and trace amounts of industrial and household chemicals, has become common practice in Western Europe, the US, and Canada. Local governments, however, are increasingly restricting or banning the practice in response to residents reporting adverse health effects. These self-reported illnesses have not been studied and methods for assessing exposures of residential communities to contaminants from processed sewage sludges need to be developed.

Methods: To describe and document adverse effects reported by residents, 48 individuals at ten sites in the US and Canada were questioned about their environmental exposures and symptoms. Information was obtained on five additional cases where an outbreak of staphylococcal infections occurred near a land application site in Robesonia, PA. Medical records were reviewed in cases involving hospitalisation or other medical treatment. Since most complaints were associated with airborne contaminants, an air dispersion model was used as a means for potentially ruling out exposure to sludge as the cause of adverse effects.

Results: Affected residents lived within approximately 1 km of land application sites and generally complained of irritation (e.g., skin rashes and burning of the eyes, throat, and lungs) after exposure to winds blowing from treated fields. A prevalence of *Staphylococcus aureus* infections of the skin and respiratory tract was found. Approximately 1 in 4 of 54 individuals were infected, including 2 mortalities (septicaemia, pneumonia). This result was consistent with the prevalence of *S. aureus* infections accompanying diaper rashes in which the organism, which is commonly found in the lower human colon, tends to invade irritated or inflamed tissue.

Conclusions: When assessing public health risks from applying sewage sludges in residential areas, potential interactions of chemical contaminants with low levels of pathogens should be considered. An increased risk of infection may occur when allergic and non-allergic reactions to endotoxins and other chemical components irritate skin and mucus membranes and thereby compromise normal barriers to infection.

Particulate matter composition and emission rates from the disk incorporation of class B biosolids into soil. Tania Paez-Rubio, Xin Huab, James Anderson, Jordan Peccia, 2006. Atmospheric Environment, 40:7034-7045

Abstract: Biosolids contain metal, synthetic organic compound, endotoxin, and pathogen concentrations that are greater than concentrations in the agricultural soils to which they are applied. Once applied, biosolids are incorporated into soils by disking and the aerosols produced during this process may pose an airborne toxicological and infectious health hazard to biosolids workers and nearby residents. Field studies at a Central Arizona biosolids land application site were conducted to characterize the physical, chemical, and biological content of the aerosols produced during biosolids disking and the content of bulk biosolids and soils from which the aerosols emanate. Arrayed samplers were used to estimate the vertical source aerosol concentration profile to enable plume height and associated source emission rate calculations. Source aerosol

concentrations and calculated emission rates reveal that disking is a substantial source of biosolids-derived aerosols. The biosolids emission rate during disking ranged from 9.91 to 27.25 mg s^{-1} and was greater than previously measured emission rates produced during the spreading of dewatered biosolids or the spraying of liquid biosolids. Adding biosolids to dry soils increased the moisture content and reduced the total PM10 emissions produced during disking by at least three times. The combination of bulk biosolids and aerosol measurements along with PM10 concentrations provides a framework for estimating aerosol concentrations and emission rates by reconstruction. This framework serves to eliminate the difficulty and inherent limitations associated with monitoring low aerosol concentrations of toxic compounds and pathogens, and can promote an increased understanding of the associated biosolids aerosol health risks to workers and nearby residents.

Source Tracking Aerosols Released from Land-Applied Class B Biosolids during High-Wind Events. Carolina Baertsch, Tania Paez-Rubio, Emily Viau, and Jordan Peccia, 2007. Applied and Environmental Microbiology, 73:4522-4531

Abstract: DNA-based microbial source tracking (MST) methods were developed and used to specifically and sensitively track the unintended aerosolization of land-applied, anaerobically digested sewage sludge (biosolids) during high-wind events. Culture and phylogenetic analyses of bulk biosolids provided a basis for the development of three different MST methods. They included (i) culture- and 16S rRNA gene-based identification of Clostridium bifermentans, (ii) direct PCR amplification and sequencing of the 16S rRNA gene for an uncultured bacterium of the class Chloroflexi that is commonly present in anaerobically digested biosolids, and (iii) direct PCR amplification of a 16S rRNA gene of the phylum Euryarchaeota coupled with terminal restriction fragment length polymorphism to distinguish terminal fragments that are unique to biosolid-specific microorganisms. Each method was first validated with a broad group of bulk biosolids and soil samples to confirm the target's exclusive presence in biosolids and absence in soils. Positive responses were observed in 100% of bulk biosolid samples and in less than 11% of the bulk soils tested. Next, a sampling campaign was conducted in which all three methods were applied to aerosol samples taken upwind and downwind of fields that had recently been land applied with biosolids. When average wind speeds were greater than 5 m/s, source tracking results confirmed the presence of biosolids in 56% of the downwind samples versus 3% of the upwind samples. During these high-wind events, the biosolid concentration in downwind aerosols was between 0.1 and 2 µg/m 3. The application of DNA-based source tracking to aerosol samples has confirmed that wind is a possible mechanism for the aerosolization and off-site transport of land-applied biosolids.

Off-Site Exposure to Respirable Aerosols Produced during the Disk-Incorporation of Class B Biosolids. Swee Yang Low, Tania Paez-Rubio, Carolina Baertsch, Matthew Kucharski, and Jordan Peccia, 2007. Journal of Environmental Engineering, 133:987-994

Abstract: Field experiments were conducted at a Class B biosolids land application site in central Arizona to measure, model, and source-track the off-site transport of aerosols emitted when biosolids were disk-incorporated into soils. Real-time PM10 monitoring provided time-resolved

aerosol information sufficient for verifying both off-site concentration and off-site exposure time model results. Under the conditions considered and at a distance of 165 m from the aerosol source, biosolids disk-incorporation resulted in an intermittent exposure to biosolids-derived aerosol concentration between 15 and 40 µg/m3 and an inhalable biosolids dose between 2 and 8 µg. Transport modeling predicted that these doses will decrease with increasing wind speed. In addition, three DNA sequence-based biosolids source tracking methods were applied to aerosol samples and confirmed the presence of biosolids in aerosols at 5, 65, and 165 m from the aerosol source. Field measurements and modeling indicate that the nature of biosolids-derived aerosol exposure is a series of intermittent high concentration puffs, rather than a continuous low concentration.

Emission Rates and Characterization of Aerosols Produced During the Spreading of Dewatered Class B Biosolids. Tania Paez-Rubio, Abel Ramarui, Jeffrey Sommer, Hua Xin, Hua, James Anderson, and Jordan Peccia, 2008. Environmental Science and Technology, 41(10):3537-3544.

Abstract: This study measured aerosol emission rates produced during the spreading of dewatered class B biosolids onto agricultural land. Rates were determined in multiple independent experimental runs by characterizing both the source aerosol plume geometry and aerosol concentrations of PM10, total bacteria, heterotrophic plate count bacteria (HPC), two types of biosolids indicator bacteria, endotoxin, and airborne biosolids regulated metals. These components were also measured in the bulk biosolids to allow for correlating bulk biosolids concentrations with aerosol emission rates and to produce reconstructed aerosol concentrations. The average emission rates and associated standard deviation for biosolids PM10, total bacteria, HPC, total coliforms, sulfite-reducing Clostridia, endotoxin, and total biosolids regulated metals were 10.1 ± 8.0 (mg/s), $1.98 \pm 1.41 \times 10^9$ (no./s), $9.0 \pm 11.2 \times 10^7$ (CFU/s), $4.9 \pm 2.2 \times 10^3$ (CFU/ s), $6.8 \pm 3.8 \times 10^3$ (CFU/s), $2.1 \pm 1.8 \times 10^4$ (EU/s), and 36.9 ± 31.8 (µg/s) respectively. Based on the land application rates of spreaders used in this study, an estimated 7.6 ± 6.3 mg of biosolids were aerosolized for every 1 kg (dry weight) applied to land. Scanning electron microscopy particle size distribution analysis of the aerosols revealed that greater than 99% of the emitted particles were less than 10 µm and particle size distributions had geometric mean diameters and standard deviations near 1.1 ± 0.97 µm. The demonstrated correlations of bulk biosolids concentrations with aerosol emission rates, and the reconstruction of aerosol concentration based on PM10 and bulk biosolids concentration provide a more fundamental, bulk biosolids based approach for extending biosolids aerosol exposure assessment to different land application scenarios and a broader range of toxins and pathogens.

Non-regulated contaminants and POPs

Only 9 contaminants are regulated under the Part 503 rules. There are many unregulated contaminants present in sewage biosolids. Some were considered when the rules were being developed and EPA decided not to regulate them. Chemicals considered for regulation, but not included in the 503 rules, include both chemicals for which there were insufficient data to evaluate the risks as well as chemicals for which EPA determined the risk was not substantial. There are,

however, many other chemicals now in widespread usage that were not even considered when the 503 rules were promulgated. Among those are the brominated flame retardants, antibacterials, wastewater treatment flocculant polymers, organotins, surfactants, fragrance chemicals and pharmaceuticals.

Over 500 different synthetic organic chemicals have been reported in sewage sludges. Concentrations of many exceed Soil Screening levels set by EPA. None are regulated in sewage biosolids in the US. EPA eliminated organic chemicals from regulatory consideration based on insensitive analyses that had high detection limits for most organic chemicals, too high to measure levels that would be of environmental significance.

All sewage biosolids contain an array of synthetic organic chemicals. An array of pharmaceuticals was found in all of the biosolids tested, regardless of the type of treatment. All biosolids are "highly enriched" in organic wastewater contaminants. Some are present in high concentrations in sewage biosolids (up to 1% by dry weight). Some have demonstrated toxicity. Pharmaceuticals are designed to be biologically active at very low concentrations and thus even at trace levels they may impact plants and animals. There is new information showing that antibiotics and other pharmaceuticals have an impact on plants grown in soils containing these chemicals.

The fate of chemicals entering a wastewater treatment plant depends on the chemical and the treatment processes. They may pass through the treatment plant virtually undegraded and travel with the water effluent, they may be sorbed onto the sludge solids, they may volatilize or they may be transformed or degraded in the treatment process. Most organic chemicals tend to sorb onto and thus concentrate in sewage biosolids rather than volatilizing or traveling through the wastewater treatment plant for discharge with the water effluent.

While many organic chemicals are not degraded or transformed by treatment processes (including composting), some compounds are transformed through chemical and biological process, creating daughter products that may be more or less toxic than the original compound. For example, surfactants are a group of chemicals present in large quantities in biosolids. The degradation products of alkyl phenol ethoxylate (APE) surfactants are significantly more toxic than the original compounds and anaerobic digestion processing at wastewater treatment plants promote this transformation, resulting in high concentrations of the recalcitrant and toxic daughter product. This has led to the restriction in use of APEs in Europe. Even compounds that may degrade to less toxic products may be present in such high concentrations in sludges that despite degradation that may take place when the sludge is applied to land, the concentration of the original compound remains at levels of concern. The surfactant LAS is such a compound.

Determination of Anionic and Nonionic Surfactants, Their Degradation Products, and Endocrine-Disrupting Compounds in Sewage Sludge by Liquid Chromatography/Mass Spectrometry. M. Petrovic and D. Barcelo, 2000. Analytical Chemistry, 72: 4560-4567

Surfactants are present in sludges in high concentrations. Degradation may result in more toxic compounds. Aerobic conditions are necessary for more complete degradation of some surfactants to more benign products.

Organic Chemicals in Sewage Sludges. Ellen Z. Harrison, Summer Rayne Oakes, Matthew Hysell, and Anthony Hay, 2006. Science of the Total Environment 367(2-3):481-497.

Abstract: Sewage sludges are residues resulting from the treatment of wastewater released from various sources including homes, industries, medical facilities, street runoff and businesses. Sewage sludges contain nutrients and organic matter that can provide soil benefits and are widely used as soil amendments. They also, however, contain contaminants including metals, pathogens, and organic pollutants. Although current regulations require pathogen reduction and periodic monitoring for some metals prior to land application, there is no requirement to test sewage sludges for the presence of organic chemicals in the U. S. To help fill the gaps in knowledge regarding the presence and concentration of organic chemicals in sewage sludges, the peer-reviewed literature and official governmental reports were examined. Data were found for 516 organic compounds which were grouped into 15 classes. Concentrations were compared to EPA risk-based soil screening limits (SSLs) where available. For 6 of the 15 classes of chemicals identified, there were no SSLs. For the 79 reported chemicals which had SSLs, the maximum reported concentration of 86% exceeded at least one SSL. Eighty-three percent of the 516 chemicals were not on the EPA established list of priority pollutants and 80% were not on the EPA's list of target compounds. Thus analyses targeting these lists will detect only a small fraction of the organic chemicals in sludges. Analysis of the reported data shows that more data has been collected for certain chemical classes such as pesticides, PAHs and PCBs than for others that may pose greater risk such as itrosamines. The concentration in soil resulting from land application of sludge will be a function of initial concentration in the sludge and soil, the rate of application, management practices and losses. Even for chemicals that degrade readily, if present in high concentrations and applied repeatedly, the soil concentrations may be significantly elevated. The results of this work reinforce the need for a survey of organic chemical contaminants in sewage sludges and for further assessment of the risks they pose.

Survey of Organic Wastewater Contaminants in Biosolids Destined for Land Application. C.A. Kinney, E.T. Furlong, S.D. Zaugg, M.R. Burkhardt, S.L. Werner, J.D. Cahill, and G.R. Jorgensen, 2006. Environmental Science and Toxicology, 40(23):7207-7215.

Abstract: In this study, the presence, composition, and concentrations of organic wastewater contaminants (OWCs) were determined in solid materials produced during wastewater treatment. This study was undertaken to evaluate the potential of these solids, collectively referred to as biosolids, as a source of OWCs to soil and water in contact with soil. Nine different biosolid products, produced by municipal wastewater treatment plants in seven different states, were analyzed for 87 different OWCs. Fifty-five of the OWCs were detected in at least one biosolid product. The 87 different OWCs represent a diverse cross section of emerging organic contaminants that enter wastewater treatment plants and may be discharged without being

completely metabolized or degraded. A minimum of 30 and a maximum of 45 OWCs were detected in any one biosolid. The biosolids used in this study are produced by several production methods, and the plants they originate from have differing population demographics, yet the percent composition of total OWC content, and of the most common OWCs, typically did not vary greatly between the biosolids tested. The summed OWC content ranged from 64 to 1811 mg/kg dry weight. Six biosolids were collected twice, 3-18 months apart, and the total OWC content of each biosolid varied by less than a factor of 2. These results indicate that the biosolids investigated in this study have OWC compositions and concentrations that are more similar than different and that biosolids are highly enriched in OWCs (as mass-normalized concentrations) when compared to effluents or effluent-impacted water. These results demonstrate the need to better describe the composition and fate of OWCs in biosolids since about 50% of biosolids are land applied and thus become a potentially ubiquitous nonpoint source of OWCs into the environment.

Organic Contaminants in Canadian Municipal Sewage Sludge. Part II. Persistent Chlorinated Compounds and Polycyclic Aromatic Hydrocarbons. J. Kohli, H.B. Lee and T.E. Peart, 2006. Water Quality Research Journal of Canada, 41: 47-55

PAHs, PCBs, and other persistent organic pollutants are found in essentially all sludges, but at widely varying concentrations depending on the source of sludge.

Persistence of organic contaminants in sewage sludge-amended soil: A field experiment. S.C. Wilson, R. E. Alcock, A.P. Sewart, K.C. Jones, 1997. J. Environ. Qual., 26: 1467-1477.

POPs introduced into soils by sewage sludge incorporation, specifically dioxins and PCBs, persisted in the soil with concentrations unchanged up to 260 days.

Partitioning, persistence, and accumulation in digested sludge of the topical antiseptic triclocarban during wastewater treatment. J. Heidler, A. Sapkota, R.U. Halden, 2006. Environmental Science & Technology, 40, 3634-3639.

Antibacterial chemicals, including triclosan and triclocarban, are common additives in many antimicrobial household products, including soaps and other personal care products. Research now confirms that most of the triclocarban in wastewater sludge is not decomposed during anaerobic digestion in the wastewater treatment plant, with the result that it concentrates to a high degree in sewage sludge.

Bioaccumulation of pharmaceuticals and other anthropogenic waste indicators in earthworms from agricultural soil amended with biosolid or swine manure. C.A. Kinney, E.D. Furlong, D.W. Kolpin, M.R. Burkhardt, S.D. Zaugg, S.L. Werner, J.P. Bossio and M.J. Benotti, 2008. Environmental Science & Technology, 42:1863-1870.

Triclosan has been shown to bioaccumulate in earthworms sampled from an agricultural field amended with sewage sludge.

Fate of higher brominated PBDEs in lactating cows. A. Kierkegaard, L. Asplund , C.A. deWit , M.S. McLachlan , G.O. Thomas, A.J. Sweetman, K.C. Jones, 2007. Environ. Sci. Technol., 41:417-423

Brominated fire retardant chemicals in contaminated feed accumulated in the fat of cows, indicating that meat consumption may be an important human exposure route to higher brominated BDEs. This

observation has important implications for pasture and forage land contamination by these chemicals in sewage sludge.

EPA finds record PFOS, PFOA levels in Alabama grazing fields. R. Renner, 2009. Environmental Science & Technology, 43(5):1245-1246.

Scientists with the EPA, USDA and FDA are investigating whether the high levels of perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) measured in agricultural soils in Alabama could have entered the food chain through beef cattle grazing on the land. Sewage sludge had been applied to these pasture lands used for grazing over a 12 year period, and is the likely source of these stable perfluorinated chemicals which are possibly carcinogenic.

Removal of Organotins During Sewage Treatment: A Case Study. N. Voulvoulis, M.D. Scrimshaw, and J.N. Lester, 2004. Environmental Technology, 25(6):733-740.

Organotins are highly toxic compounds found in sludges. They do not degrade in the wastewater treatment process.

The potential impact of veterinary and human therapeutic agents in manure and biosolids on plants grown on arable land: a review. Patrick K. Jjemba, 2002. Agriculture, Ecosystems and Environment, 93(1-3):267-278

Substantial quantities of pharmaceuticals are applied to land in sludges and manures. Detrimental impacts of pharmaceuticals on crops is observed with some species of plants.

Bacterial regrowth/viable non-culturable (VNC)

Recent research has demonstrated that sewage biosolids believed to meet Class A or Class B standards were subject to regrowth and reactivation of bacteria. Thus materials have been land applied that contained bacterial levels far above those of Class A or Class B as defined by USEPA under Part 503. Coliform concentrations were found to increase by 100-1000-fold in biosolids and in soil/biosolid mixtures after centrifugation of anaerobically digested biosolids. Coliform concentrations up to 100,000 times those measured by conventional culture methods may be found in thermophilically digested sludges after centrifugation. This results from the presence of viable but non-culturable bacteria.

Increases in Fecal Coliform Bacteria Resulting From Centrifugal Dewatering of Digested Biosolids. Yinan Qi, Steven. K. Dentel, and Diane S. Herson, 2007. Water Research, 41(3):571-580.

Abstract: In many countries, the classification of biosolids for disposal purposes can be based, in part, on fecal coliform levels, with alternative criteria also available based on the stabilization process used, such as anaerobic digestion. The assumption that these alternative criteria provide equivalent protection may be flawed. This paper demonstrates that fecal coliform levels determined after digestion do not always indicate the bacterial levels after the same biosolids have been dewatered by centrifugation. In samples from mesophilic digestion, half had significant

increases in coliform numbers ($P<0.05$) with up to one order of magnitude increase during centrifugation, suggesting coliform regrowth. Thermophilically digested samples had significant increases of several orders of magnitude during dewatering, more likely from reactivation of viable but non-culturable coliforms than from regrowth. In other cases, centrifugation induced coliform regrowth or reactivation upon incubation and storage of dewatered samples, but not digested samples. These 2–3 order of magnitude increases occurred with both 25 and 37 °C incubations. Coliform increases continued for up to 5 days, then gradually declined. However, by day 20 coliform numbers were still 2 orders of magnitude greater than when originally sampled. The magnitude of the increases could be due either to regrowth or reactivation, but the nature of the longer-term increases—also seen in biosolids/soil mixtures—suggests regrowth. Differences in numbers between digested and dewatered samples could not be duplicated with high shear processing in lab-scale devices, with nitrogen purging to remove volatile or gaseous constituents, or with redilution using centrate. They could not be attributed to enumeration methods, to interference of Bacillus spp. on apparent coliform counts, or to temperature changes. The increases have practical implications in the use of fecal coliform or alternative criteria to define pathogen content in biosolids.

Reactivation and Growth of Non-Culturable Indicator Bacteria in Anaerobically Digested Biosolids After Centrifuge Dewatering. Matthew J. Higgins, Yen-Chih Chen, Sudhir N. Murthy, Donald Hendrickson, Joseph Farrel, Perry Schafer, 2007. Water Research, 41(3):665-673

Abstract: Recent literature has reported that high concentrations of indicator bacteria such as fecal coliforms (FCs) were measured in anaerobically digested sludges immediately after dewatering even though low concentrations were measured prior to dewatering. This research hypothesized that the indicator bacteria can enter a non-culturable state during digestion, and are reactivated during centrifuge dewatering. Reactivation is defined as restoration of culturability. To examine this hypothesis, a quantitative polymerase chain reaction (qPCR) method was developed to enumerate Escherichia coli, a member of the FC group, during different phases of digestion and dewatering. For thermophilic digestion, the density of E. coli measured by qPCR could be five orders of magnitude greater than the density measured by standard culturing methods (SCMs), which is indicative of non-culturable bacteria. For mesophilic digestion, qPCR enumerated up to about one order of magnitude more E. coli than the SCMs. After centrifuge dewatering, the non-culturable organisms could be reactivated such that they are enumerated by SCMs, and the conditions in the cake allowed rapid growth of FCs and E. coli during cake storage.

Antibiotic resistance in sludge bacteria

Recent studies have confirmed that the use of antimicrobials had created a large pool of antibiotic-resistance genes in bacteria that are detected in sewage sludge and effluent from sewage treatment plants. Antibiotic resistant bacteria were found in higher numbers downstream of sludge-treated farmland as compared to upstream.

Increased Frequency of Drug-resistant Bacteria and Fecal Coliforms in an Indiana Creek Adjacent to Farmland Amended with Treated Sludge. Shivi Selvaratnam and David J. Kunberger, 2004. Canadian Journal of Microbiology, 50(8):653-656

Abstract: Many studies indicate the presence of human pathogens and drug-resistant bacteria in treated sewage sludge. Since one of the main methods of treated sewage disposal is by application to agricultural land, the presence of these organisms is of concern to human health. The goal of this study was to determine whether the frequency of drug resistant and indicator bacteria in Sugar Creek, which is used for recreational purposes, was influenced by proximity to a farmland routinely amended with treated sludge (site E). Surface water from 3 sites along Sugar Creek (site E, 1 upstream site (site C) and 1 downstream site (site K)) were tested for the presence of ampicillin-resistant (AmpR) bacteria, fecal and total coliforms over a period of 40 d. Site E consistently had higher frequencies of AmpR bacteria and fecal coliforms compared with the other 2 sites. All of the tested AmpR isolates were resistant to at least 1 other antibiotic. However, no isolate was resistant to more than 4 classes of antimicrobials. These results suggest that surface runoff from the farmland is strongly correlated with higher incidence of AmpR and fecal coliforms at site E.

Potential ecological and human health impacts of antibiotics and antibiotic-resistant bacteria from wastewater treatment plants. S. Kim and D.S. Aga, 2007. Journal of Toxicology and Environmental Health-Part B-Critical Reviews, 10:559-573.

Abstract**:** The occurrence of antibiotics and other pharmaceuticals in the environment has become an increasing public concern as recent environmental monitoring activities reveal the presence of a broad range of persistent pharmaceuticals in soil and water. Studies show that municipal wastewater treatment plants (WWTPs) are important point sources of antibiotics and antibiotic-resistant bacteria in the environment. The fate of antibiotics and other pharmaceuticals in WWTPs is greatly influenced by the design and operation of treatment systems. Because knowledge on the fate of antibiotics and resistant bacteria in WWTPs is important in estimating their potential impacts on ecology and human health, investigations on occurrence, treatment, and observed effects are reviewed in this article. In addition, human health risk assessment protocols for antibiotic and resistant bacteria are described. Although data on other pharmaceutical compounds are also presented, discussion is focused on antibiotics in the environment because of the potential link to increased emergence of resistance among pathogenic bacteria. The applications of modern analytical methods that facilitate the identification of novel transformation products of pharmaceuticals in environmental matrices are also included to illustrate that the disappearance of the parent pharmaceuticals in WWTPs does not necessarily equate to their complete removal.

Effect of wastewater treatment on antibiotic resistance in Escherichia coli and Enterococcus sp. S. Garcia, B. Wade, C. Bauer, C. Craig, K. Nakaoka, and W. Lorowitz, 2007.Water Environment Research, 79:2387-2395

Abstract**:** The effects of wastewater treatment on the proportion of Escherichia coli and Enterococcus sp. resistant to specific antibiotics were investigated at two facilities in Davis

County, Utah, one of which received hospital waste. Samples were taken from the influent, effluent before disinfection, and secondary anaerobic sludge digester effluent. There was very little difference in antibiotic resistance among E. coli in the inflow waters of the plants but the plant receiving hospital waste had a significantly higher proportion of antibiotic resistant Enterococcus. The effect of wastewater treatment on antibiotic resistance was more pronounced on enterococci than E. coli. Although some increases in antibiotic resistance were observed, the general trend seemed to be a decrease in resistance, especially in the proportion of multidrug resistant Enterococcus sp.

Antimicrobial resistance in Enterococcus spp. isolated in inflow, effluent and sludge from municipal sewage water treatment plants. P.M. Da Costa, P. Vaz-Pires, and F. Bernardo, 2006. Water Research, 40:1735-1740

Abstract**:** Antimicrobial resistance of enterococci was investigated in 42 samples of crude inflow, treated effluent and sludge collected in 14 municipal sewage treatment plants of Portugal. A total of 983 enterococci were recovered and tested, using the diffusion agar method, regarding their sensitivity to 10 different antimicrobial drugs. Multidrug resistance was present in 49.4% of the isolates. Only 3.3% and 0.6% of the investigated strains were resistant to ampicillin and vancomycin, respectively. Resistances found against rifampicin (51.5%), tetracycline (34.6%), erythromycin (24.8%) and nitrofurantoin (22.5%), are causes for substantial concern. Almost 14% of isolates were resistant to ciprofloxacin. Wastewater treatment resulted in enterococci decrease between 0.5 and 4log; nevertheless, more than 4.4 x 10(5) CFU/100ml were present in the outflow of the plants. Our data indicate that the use of antimicrobials had created a large pool of resistance genes and that sewage treatment processes are unable to avoid the dissemination of resistant enterococci into the environment.

Prions

The potential for prions that might be present in wastewater to accumulate in sludges and to persist through treatment is a concern.

***Persistence of Pathogenic Prion Protein during Simulated Wastewater Treatment Processes*.** G.T. Hincklley, C.J. Johnson, K.H. Jacobson, C. Bartholomay, K.D. McMahon, D. McKenzie, J.M. Aiken, and J.A. Pederson, 2008. Environmental Science and Technology, 42(14):5254-5259.

Abstract: Transmissible spongiform encephalopathies (TSEs, prion diseases) are a class of fatal neurodegenerative diseases affecting a variety of mammalian species including humans. A misfolded form of the prion protein (PrPTSE) is the major, if not sole, component of the infectious agent. Prions are highly resistant to degradation and to many disinfection procedures suggesting that, if prions enter wastewater treatment systems through sewers and/or septic systems (e.g., from slaughterhouses, necropsy laboratories, rural meat processors, private game dressing) or through leachate from landfills that have received TSE-contaminated material, prions could survive conventional wastewater treatment. Here, we report the results of experiments examining the partitioning and persistence of PrPTSE during simulated wastewater treatment processes including

activated and mesophilic anaerobic sludge digestion. Incubation with activated sludge did not result in significant PrPTSE degradation. PrPTSE and prion infectivity partitioned strongly to activated sludge solids and are expected to enter biosolids treatment processes. A large fraction of PrPTSE survived simulated mesophilic anaerobic sludge digestion. The small reduction in recoverable PrPTSE after 20-d anaerobic sludge digestion appeared attributable to a combination of declining extractability with time and microbial degradation. Our results suggest that if prions were to enter municipal wastewater treatment systems, most would partition to activated sludge solids, survive mesophilic anaerobic digestion, and be present in treated biosolids.

Ecological impacts

Soil microorganisms play a critical role in the functions of soil as a source of plant nutrition and in the cycling of nutrients. Recent research shows that sludge application changes the soil microbial community and decreases its diversity. A number of human-use compounds (such as triclosan found in many personal care products such as antibacterial soaps) bioconcentrate in earthworms where soil has been amended with sewage sludges.

Computational Improvements Reveal Great Bacterial Diversity and High Metal Toxicity in Soil. Jason Gans, Murray Wolinsky, and John Dunbar, 2005. Science, 309:1387-1390.

Sewage sludge greatly reduced the diversity of bacterial species in soils.

Parallel Shifts in Plant and Soil Microbial Communities in Response to Biosolids in a Semi-Arid Grassland. Tarah S. Sullivan, Mary E. Stromberger, and Mark W. Paschke, 2006. Soil Biology and Biochemistry, 38 449-459.

Abstract: Approximately 70,150 dry Mg of biosolids from over 450 wastewater treatment facilities are applied to the semi-arid rangelands of Colorado every year. Research on semi-arid grassland responses to biosolids has become vital to better understand ecosystem dynamics and develop effective biosolids management strategies. The objectives of this study were to determine the long-term (~12 years) effects of a single biosolids application, and the short-term (~2 years) effects of a repeated application, on plant and microbial community structure in a semi-arid grassland soil. Specific attention was paid to arbuscular mycorrhizal fungi (AMF) and linkages between shifts in plant and soil microbial community structures. Biosolids were surface applied to experimental plots once in 1991 (long-term plots) and again to short-term plots in 2002 at rates of 0, 2.5, 5, 10, 21, or 30 Mg ha^{-1}. Vegetation (species richness and above-ground biomass), soil chemistry (pH, EC, total C, total N, and extractable P, NO_3-N, and NH_4-N), and soil microbial community structure [ester-linked fatty acid methyl esters (EL-FAMEs)], were characterized to assess impacts of biosolids on the ecosystem. Soil chemistry was significantly affected and shifts in both soil microbial and plant community structure were observed with treatment. In both years, the EL-FAME biomarker for AMF decreased with increasing application rate of biosolids; principal components analysis of EL-FAME data yielded shifts in the structure of the microbial communities with treatment primarily related to the relative abundance of the AMF specific biomarker. Significant (p%0.05) correlations existed among biomarkers for Gram-negative and

Gram-positive bacteria, AMF and specific soil chemical parameters and individual plant species' biomass. The AMF biomarker was positively correlated with biomass of the dominant native grass species blue grama (Bouteloua gracilis [Willd. ex Kunth] Lagasca ex Griffiths) and was negatively correlated with western wheatgrass (Agropyron smithii Rydb.) biomass. This study demonstrated that applications of biosolids at relatively low rates can have significant long-term effects on soil chemistry, soil microbial community structure, and plant community species richness and structure in the semi-arid grasslands of northern Colorado. Reduced AMF and parallel shifts in the soil microbial community structure and the plant community structure require further investigation to determine precisely the sequence of influence and resulting ecosystem dynamics.

Bioaccumulation of Pharmaceuticals and Other Anthropogenic Waste Indicators in Earthworms from Agricultural Soil Amended With Biosolid or Swine Manure. C.A. Kinney, E.T. Furlong, D.W. Kolpin, M.R. Burkhardt, S.D. Zaugg, S.L. Werner, J.P. Bossio and M.J. Benotti, 2008. Environmental Science and Technology, 42:1863-1870.

Abstract: Analysis of earthworms offers potential for assessing the transfer of organic anthropogenic waste indicators (AWIs) derived from land-applied biosolid or manure to biota. Earthworms and soil samples were collected from three Midwest agricultural fields to measure the presence and potential for transfer of 77 AWIs from land-applied biosolids and livestock manure to earthworms. The sites consisted of a soybean field with no amendments of human or livestock waste (Site 1), a soybean field amended with biosolids from a municipal wastewater treatment plant (Site 2), and a cornfield amended with swine manure (Site 3). The biosolid applied to Site 2 contained a diverse composition of 28 AWIs, reflecting the presence of human-use compounds. The swine manure contained 12 AWIs, and was dominated by biogenic sterols. Soil and earthworm samples were collected in the spring (about 30 days after soil amendment) and fall (140-155 days after soil amendment) at all field sites. Soils from Site 1 contained 21 AWIs and soil from Sites 2 and 3 contained 19 AWIs. The AWI profiles at Sites 2 and 3 generally reflected the relative composition of AWIs present in waste material applied. There were 20 AWIs detected in earthworms from Site 1 (three compounds exceeding concentrations of 1000 μg/kg), 25 AWIs in earthworms from Site 2 (seven compounds exceeding concentrations of 1000 μg/ kg), and 21 AWIs in earthworms from Site 3 (five compounds exceeding concentrations of 1000/μg/kg). A number of compounds that were present in the earthworm tissue were at concentrations less than reporting levels in the corresponding soil samples. The AWIs detected in earthworm tissue from the three field sites included pharmaceuticals, synthetic fragrances, detergent metabolites, polycyclic aromatic hydrocarbons (PAHs), biogenic sterols, disinfectants, and pesticides, reflecting a wide range of physicochemical properties. For those contaminants detected in earthworm tissue and soil, bioaccumulation factors (BAF) ranged from 0.05 (galaxolide) to 27 (triclosan).Thisstudydocuments that when AWIs are present in source materials that are land applied, such as biosolids and swine manure, AWIs can be transferred to earthworms.

International Standards for Heavy Metals

The USEPA standards for sewage biosolid contaminant concentrations (standards are set for 9 metals) are higher than those in other developed countries and higher than recommendations of scientists in the northeastern U.S. Switzerland has banned sludge application.

Since the 503 rule was promulgated by USEPA, there has been no reassessment of the heavy metal loading limits on agricultural soils set at that time. In fact, there has been no significant research effort in the US to test the assertion by EPA that the very high metal loading limits (by international standards) of the 503 rule have a high safety margin in protecting soil productivity and crop quality.

Two recent large multi-site field investigations measuring the long-term impacts of sludge metals on soil health and crop quality were undertaken independently in Australia and the UK. In the absence of a comparable study of this scale or longevity in the US, the results of the Australian and UK studies are highly useful in developing guidelines for heavy metals in the US.

The Australian study addressed the impact of Cd loading on food crop quality (levels of Cd in edible crops), and Cu and Zn impacts on crop production (phytotoxicity) and soil health (microbial processes). The recommended limits are much lower for most soils than the allowed soil concentrations of Cd, Zn and Cu based on metal loadings permitted by the USEPA 503 rule. However, the study revealed the high sensitivity of harmful metal effects in soils on soil properties such as pH, clay content and organic matter content. Therefore, the recommended limits for the heavy metals vary greatly by soil type, with acid sandy soils being the most sensitive soils to metal additions.

Ban on the Use of Sludge as a Fertiliser. Switzerland: Federal Office for the Environment, 2003. http://www.bafu.admin.ch/dokumentation/medieninformation/00962/index.html?lang=en&msg-id=1673.

Bern, 26.03.2003 – The use of sludge as a fertiliser is to be banned throughout Switzerland; in the future sludge will have to be incinerated using an environmentally friendly method. The Swiss Federal Council will modify the Ordinance on Materials accordingly on 1 May 2003. The ban will be introduced in stages: from May this year, sludge may no longer be used in the production of fodder crops and vegetables. A period of transition lasting until 2006 at the latest has been accorded for other types of cultivation which until now have been fertilised using sludge; in individual cases the cantonal authorities may extend this period until 2008. This decision is part of the Federal Council's implementation of precautionary provisions for the protection of soils and public health.

Although sludge contains plant nutrients such as phosphorus and nitrogen it also comprises a whole range of harmful substances and pathogenic organisms produced by industry and private households. For this reason, most farmers already avoid using sludge as a fertiliser since they are

aware of the risk of irreversible damage to the soil, the danger to public health and possible negative effects on the quality of the food they produce."

Australian recommendations on soil limits for cadmium, zinc and copper

Recommendations of the Australian National Biosolids Research Program on Biosolids Guidelines. Michael Warne, Mike McLaughlin, Diane Heemsbergen, Mike Bell, Kris Broos, Mark Whatmuff, Glenn Barry, David Nash, Deb Pritchard, Daryl Stevens, Grant Pu, and Craig Butler, 2007. Draft Position Paper.

Executive Summary: A set of soil specific maximum limits for copper and zinc in soils that have received biosolids were derived. These recommended limits state the amount of copper or zinc that can be added to a soil. In acidic, low carbon soils (pH 5, OC 1%) the recommended limit is 25 mg/kg added copper, which increases to 245 mg/kg added copper in alkaline soils (pH 8) irrespective of the organic carbon content. The recommended limits are, depending on the soil properties at a site, considerably smaller to considerably larger than the current limits of 100 – 200 mg/kg total copper. In acidic, low cation exchange capacity (CEC) soils (pH 5, CEC 3 cmolc/kg) the recommended limit for zinc in soils that have received biosolids is 20 mg/kg added zinc, which increases to 300 mg/kg added zinc when the soil pH is greater than or equal to 7.5 irrespective of the cation exchange capacity. Thus, the recommended limits can be considerably lower to marginally higher than the current limits of 200 – 250 mg/kg total zinc, depending on the properties of the soils at sites. Critical soil concentrations of cadmium that would lead to exceedance of the Food Standards Australian New Zealand (FSANZ) standard (0.1 mg/kg) for human consumption were determined across all NBRP sites. The critical values were affected by soil properties, principally soil pH and clay content. A set of recommended soil specific maximum cadmium concentrations in soils that have received biosolids were developed. The recommended limit for total cadmium at a soil pH of 5.5 is 0.6 mg/kg in sandy soils (5% clay or less). In alkaline (pH 7.5 or greater) and clayey soils (25% or greater) the recommended limit for total cadmium in soil is approximately 1 mg/kg or greater. Thus depending on the soil properties at a site the recommended cadmium soil concentration is considerably smaller to considerably greater than the value of 1 mg/kg previously recommended by the National Cadmium Management Committee. From the above recommended limits for cadmium, copper and zinc it is apparent that soils that are acidic combined with either low organic carbon, low clay content or low cation exchange capacity have low critical soil metal concentrations. The critical soil concentrations increased as the pH, organic carbon content, clay content or cation exchange capacity of soils increased. Based on the recommended soil limits, typical metal concentrations in biosolids and current land application practices example masses of biosolids that could be applied cumulatively to land were calculated. For high risk sites as little as 40 to 90 tonnes in total may be added, while at low risk sites between 280 and 970 tonnes in total may be applied. At typical current agronomic application rates of 10 t/ha this translates to 4 to 98 applications.

UK findings on the effect of sewage sludge metals on soil health

The UK study also addressed the impact of Cd loading on food crop quality (levels of Cd in edible crops), and Cu and Zn impacts on soil health (microbial biomass, rhizobium numbers, and microbial respiration). The results suggest that Zn is the metal responsible for the decrease in rhizobial population. It is important to stress that this study was designed to test the adequacy of existing UK limits for Cd, Zn and Cu in agricultural soils (e.g., 200-300 mg/kg for Zn). As some important detrimentral effects are being seen, at least in the early years of this long-term study, it is possible that UK limits for these metals will be adjusted lower. The present UK limits are well below those permitted in the US under the 503 rule.

Effects of Sewage Sludge Applications to Agricultural Soils on Soil Microbial Activity and the Implications for Agricultural Productivity and Long-Term Soil Fertility: Phase III, ADAS, Rothamsted Research, Water Research Centre (WRc), 2007.

Project synthesis: During the four years (2002-2006) of this project, significant ($P < 0.05$) responses in soil microbial properties (i.e. rhizobia numbers and microbial biomass size) and agricultural crop quality (i.e. grain Cd concentrations) were measured following the application of metal-rich sludge cakes and metal-amended liquid sludges during Phase I (1994-1997). The soil samples taken in spring 2003 and 2005 at all nine sites in Britain (and additionally in 1999 and 2001 during Phase II of the project) showed significant ($P < 0.05$) responses in rhizobia numbers on the Zn sludge cake treatments, and in soil microbial biomass size on the Zn and Cu sludge cake treatments. Further soil sampling and measurements during future years of this long term study will help to establish whether the effects measured so far are permanent and consistent over time.

Northeastern U.S. application guidelines

A review of published research by 9 scientists from 5 Northeastern states produced recommended limits for heavy metals that are substantially lower than those permitted under the USEPA 503 rule.

Guidelines for Application of Sewage Biosolids to Agricultural Lands in the Northeastern U.S., Ellen Z. Harrison and Uta Krogmann (Eds.), 2007. New Jersey Agricultural Experiment Station, Rutgers Cooperative Extension Bulletin, 36 pp.

Maximum recommended cumulative soil trace element concentration limits for sites to which sewage biosolids are applied are intended to address and protect the agricultural productivity under Northeast soil conditions and for Northeast farming practices and demographics some of which are unique to this region (Table 3).

Table 3. Recommended Maximum Soil Trace Element Concentrations for the Northeast US

Metal	Recommended Maximum Soil Concentration (mg/kg)		
	Sand to loamy sand	Sandy loam to silt loam	Silt to clay
cadmium	1.2	2	3
copper	50	75	120
nickel	30	40	60
lead	120	120	120
zinc	90	150	230

New Technologies as Alternative Beneficial Uses

Application of sewage biosolids is not the only option for recycling this material. New energy recovery technologies make use of the energy embedded in the sludge. Other technologies are in place to make construction material out of sludges.

***Emerging Technologies for Biosolids Management*, US EPA, 2006.**
http://www.epa.gov/OW-OWM.html/mtb/epa-biosolids.pdf

Preface: The U.S. Environmental Protection Agency (U.S. EPA) is charged by Congress with protecting the nation's land, air, and water resources. Under a mandate of environmental laws, the Agency strives to formulate and implement actions leading to a balance between human activities and the ability of natural systems to support and sustain life. To meet this mandate, the Office of Wastewater Management (OWM) provides information and technical support to solve environmental problems today and to build a knowledge base necessary to protect public health and the environment well into the future.

This publication has been produced under contract to the U.S. EPA by Parsons Corporation and provides information on the current state of development as of the publication date. It is expected that this document will be revised periodically to reflect advances in this rapidly evolving area. Except as noted, information, interviews and data development were conducted by the contractor. It should be noted that neither Parsons nor U.S. EPA has conducted engineering or operations evaluations of the technologies included. Some of the information, especially related to embryonic technologies, was provided by the manufacturer or vendor of the equipment or technology and could not be verified or supported by full-scale case study. In some cases, cost data were based on estimated savings without actual field data. When evaluating technologies, estimated costs, and stated performance, efforts should be made to obtain current information.

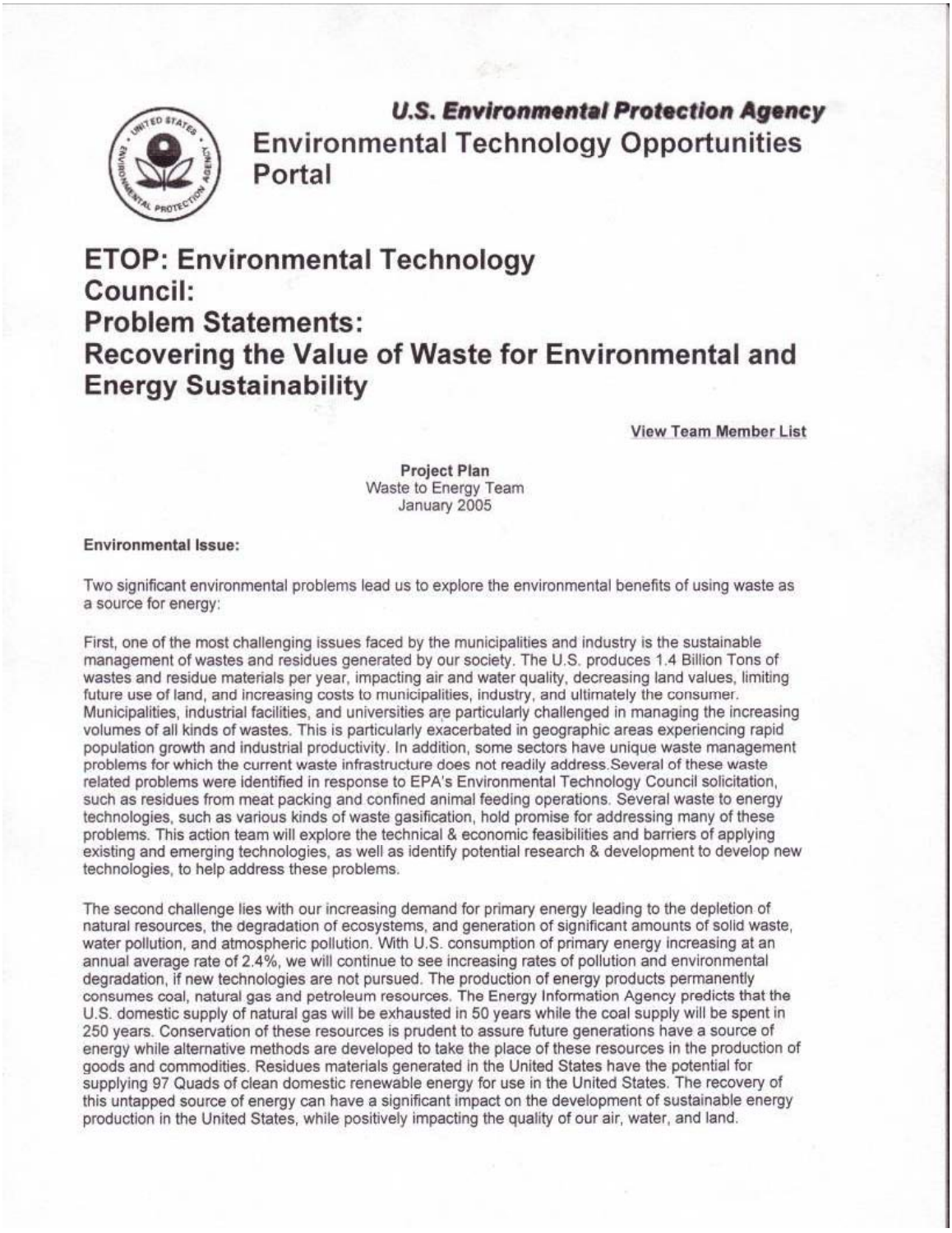
The mention of trade names, specific vendors, or products does not represent an actual or presumed endorsement, preference, or acceptance by the U.S. EPA or the Federal government. Stated results, conclusions, usage, or practices do not necessarily represent the views or policies of the U.S. EPA.

Energy alternatives

Combustion and Land Application Can Both be Beneficial? Roger Tim Haug, Deputy City Engineer City of Los Angeles, F. Michael Lewis, PE, Peter Brady, BE MIEI

Abstract: Both combustion and land application have played important roles in biosolids management practices for many decades. Land application in almost all of its forms has been proclaimed as beneficial use. By contrast, many have viewed combustion as a "disposal only" option, even if energy is recovered in the process and the resulting ash reused. These views and opinions are often proclaimed with no basis or criteria to support the conclusion. Five criteria are presented in this paper for judging whether a management practice is beneficial or not. When judged by these criteria, one can conclude that many combustion installations are beneficial. One can also conclude that land application is beneficial in most, but perhaps not all, installations."

Gasification presents an opportunity that EPA is promoting.



U.S. Environmental Protection Agency

Environmental Technology Opportunities Portal

ETOP: Environmental Technology Council: Problem Statements: Recovering the Value of Waste for Environmental and Energy Sustainability

View Team Member List

Project Plan
Waste to Energy Team
January 2005

Environmental Issue:

Two significant environmental problems lead us to explore the environmental benefits of using waste as a source for energy:

First, one of the most challenging issues faced by the municipalities and industry is the sustainable management of wastes and residues generated by our society. The U.S. produces 1.4 Billion Tons of wastes and residue materials per year, impacting air and water quality, decreasing land values, limiting future use of land, and increasing costs to municipalities, industry, and ultimately the consumer. Municipalities, industrial facilities, and universities are particularly challenged in managing the increasing volumes of all kinds of wastes. This is particularly exacerbated in geographic areas experiencing rapid population growth and industrial productivity. In addition, some sectors have unique waste management problems for which the current waste infrastructure does not readily address.Several of these waste related problems were identified in response to EPA's Environmental Technology Council solicitation, such as residues from meat packing and confined animal feeding operations. Several waste to energy technologies, such as various kinds of waste gasification, hold promise for addressing many of these problems. This action team will explore the technical & economic feasibilities and barriers of applying existing and emerging technologies, as well as identify potential research & development to develop new technologies, to help address these problems.

The second challenge lies with our increasing demand for primary energy leading to the depletion of natural resources, the degradation of ecosystems, and generation of significant amounts of solid waste, water pollution, and atmospheric pollution. With U.S. consumption of primary energy increasing at an annual average rate of 2.4%, we will continue to see increasing rates of pollution and environmental degradation, if new technologies are not pursued. The production of energy products permanently consumes coal, natural gas and petroleum resources. The Energy Information Agency predicts that the U.S. domestic supply of natural gas will be exhausted in 50 years while the coal supply will be spent in 250 years. Conservation of these resources is prudent to assure future generations have a source of energy while alternative methods are developed to take the place of these resources in the production of goods and commodities. Residues materials generated in the United States have the potential for supplying 97 Quads of clean domestic renewable energy for use in the United States. The recovery of this untapped source of energy can have a significant impact on the development of sustainable energy production in the United States, while positively impacting the quality of our air, water, and land.

Converting Biosolids to a Renewable Fuel. Michael Moore, Layne Baroldi, Deirdre Bingman, Ray Kearney, 2006. BioCycle, 47(10):32-35.

Orange County CA is working with EnerTech Environmental Inc on a facility to convert 1/3 of their biosolids to energy. The E-fuel is certified as a renewable fuel by CA Energy Commission.

Turning trash into energy in St. Lucie County. TCPalm newspaper editorial, December 1, 2006.

St Lucie County, FL is proceeding with plans to have Geoplasma INC build a plasma arc facility to deal with trash and sludge.

Green Production of Hydrogen from Excess Biosolids Originating from Municipal Waste Water Treatment. B. Bagchi, J. Rawlston, R.M. Counce, J.M. Holmes, and P.R. Bienkowski, 2006. Separation Science and Technology, 41:2613-2628

Rialto, CA OKs Energy Plant at Landfill Site. National Biosolids Partnership. 3/1/06 Weekly Biosolids Update. http://www.biosolids.org/news_weekly.asp?id=1911

Sewage turned into hydrogen fuel. NewScientist.com News Service, April 29, 2002.

RENEWABLE ENERGY: They hope to turn an array of biomass material into fuels by early 2008. John Welsh. The Press-Enterprise, Sept 14, 2006.

Bricks and glass

Sludge can be used to make construction materials including brick and aggregate.

Lightweight aggregate made from sewage sludge and incinerated ash. Ing-Jia Chiou, Kuen-Sheng Wang, Ching-Ho Chen, and Ta-Ting Lin, 2006. Waste Management, 26:1453-1461

Sewage sludge bulks up house bricks. Andy Cohlan, August 31, 2002. New Scientist

Advances in Envir Research. Chih-Huang Wend, I-Shou U in Kachsiung Co Taiwan.

Sewage vitrification. The Illinois North Shore Sanitary District has a new sludge recycling facility that is the first in the world to convert municipal biosolids into a reusable glass aggregate. Each day, up to 200 tons of municipal biosolids are transformed into 7.5 tons of glass.

Biosolids Reuse as Clear as Glass, 2006. Water Environment Federation, 18(11). http://www.wef.org/ScienceTechnologyResources/Publications/WET/06/06Nov/06NovemberProblemSolvers.htm

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Features

A CRITICAL REVIEW OF THE U.S. EPA'S RISK ASSESSMENT FOR THE LAND APPLICATION OF SEWAGE SLUDGE

JENNIFER M. J. MATHNEY

ABSTRACT

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.

Sewage sludge is defined by the U.S. Environmental Protection Agency (EPA) as the "solid, semi-solid, or liquid residue generated during the treatment of domestic sewage in a treatment works" [1]. Sewage from homes, industries, medical facilities, agriculture, street runoff, and businesses is collected at wastewater treatment facilities where it undergoes treatment processes to remove contaminants. Sewage sludge is the byproduct generated by the processes that remove contaminants from the wastewater so that the treated wastewater can be

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discharged back into waterways. Sludge is generated mainly during primary treatment, where solids settle out, and also during secondary treatment, where microorganisms are added to degrade the biological content of the sewage and the solids settle out. Further treatment can also generate sludge [1].

Many of the contaminants that were in the wastewater concentrate in the sludge, resulting in a mixture with an unknown composition of inorganic and organic materials and human pathogens [1]. Sludge itself can be treated by a variety of processes including aerobic digestion, anaerobic digestion, composting, heat drying, air drying, lime stabilization, and chemical fixation. Sewage sludge that has undergone treatment and meets federal and state standards for land application is called biosolids by EPA. Treated sludge can be applied to land—such as agricultural land, forests, parks and gardens, and home gardens and lawns. Sludge that is untreated or not treated enough to meet land application standards can be disposed of in landfills or incinerated [1-3].

The EPA and other agencies have widely promoted the use of sewage sludge for land application as a safe, beneficial, and economical way to recycle the massive amounts of sludge generated. They claim it is a fertilizer that contains beneficial plant nutrients and has other soil-conditioning properties [2, 3]. Approximately 5.6 million tons of dry sewage sludge are used or disposed of annually in the United States, of which 60 percent is used for land application or public distribution [1]. There are federal standards governing the use and disposal of all sewage sludge in Section 40 of the Code of Federal Regulations Part 503. The land application of sewage sludge has been a hotly debated topic since its inception. The EPA maintains that the standards for land application of sewage sludge are protective of human health and the environment [3]. Numerous reviews of the risk assessment used to establish the standards, however, have found serious flaws with the way EPA conducted the risk assessment. These reports critically assessed the methods used in the risk assessment, the data used, and current scientific data on sewage sludge to determine if the standards were adequate. The review presented here examined these papers and other current literature to determine if there was significant evidence to support the concern over the land application of sewage sludge and found that the literature clearly demonstrates that the current policies and regulations do not adequately protect human health and the environment. Based on the available data, the application of sewage sludge to land must be stopped because the current standards are based on inaccurate and outdated science. If the practice of land application is not stopped, the consequences to humans and the environment will be severe and long-lasting.

HISTORY AND CURRENT STANDARDS

Human excreta have been applied as fertilizer for hundreds of years, and this practice was generally safe because the excreta did not contain industrial waste. As populations grew, the old methods used to remove waste became inadequate

and resulted in numerous disease outbreaks. Sewers were invented to deal with the problem by removing the wastewater from the city and town centers. Domestic and industrial sewage was dumped into waterways until they became so polluted that a new method was needed to deal with waste.

Wastewater treatment became the new technique to deal with the problem and with wastewater treatment came sewage sludge. The passage of the Clean Water Act in 1972 more than doubled the amount of sludge generated as the treatment processes that create it became mandatory and all water had to be treated. The use of sludge for land application became widespread with the 1988 Ocean Dumping Ban, which eliminated dumping of sludge in the ocean and forced EPA to invest in land application. In 1990 the term "biosolids" was coined for sewage sludge that was treated and acceptable for land application in order to increase its appeal. Biosolids were classified as a fertilizer, and EPA pushed this use [4, 5].

In 1993 the Part 503 standards established pollution limits, operational standards, and management practices to "protect public health and the environment from any reasonably anticipated adverse effects from chemical pollutants and pathogenic organisms" in sewage sludge [1]. Minimum standards regarding ceiling concentration (mg/kg), pollutant concentration (mg/kg), cumulative pollutant loading rate limits (kg/ha), and annual pollutant loading rate (kg/ha/yr) for contaminants in sludge were established that had to be met for the sludge to be approved for land application. Originally 10 inorganic chemicals were regulated: arsenic, cadmium, chromium, copper, lead, mercury, molybdenum, nickel, selenium, and zinc. Chromium was dropped in 1995 and molybdenum has only a ceiling concentration [1]. Since sewage sludge can contain bacteria, viruses, protozoa, parasites, and other microorganisms, Part 503 mandates that sewage sludge undergo specific treatment processes to reduce pathogens before it can be applied to land. Based on the treatment processes and the amount of pathogens still present, treated sludge can be classified as Class A or B biosolids. Class A biosolids are treated to reduce pathogens to below detectable levels and can be used without any application restrictions. Class B biosolids are also treated to reduce pathogens, but pathogens remain at measurable levels, so there are restrictions regarding the application of Class B biosolids and the use of the land receiving the biosolids to minimize human contact until natural processes can further reduce pathogen content [3].

Using available data on chemicals and data from the 1988 National Sewage Sludge Survey (NSSS), EPA conducted an extensive risk assessment to establish the Part 503 standards. To support the safety of land application of sludge, proponents often quote a 1996 National Research Council (NRC) report that reviewed the use of wastewater and biosolids for agricultural purposes: the use of biosolids "presents negligible risks to the consumer, to crop production, and to the environment . . . existing regulation and guidelines governing the use of reclaimed wastewater and sludge in crop production are adequate to protect human health and the environment" [1]. What proponents fail to mention

is that the report also highlighted limitations and inconsistencies in the risk assessment approach and NSSS data used by EPA and made recommendations for further research [1, 6].

In fact, EPA did not follow through on any of the recommendations and made no changes to the standards. A 2002 National Research Council (NRC) report re-evaluated the standards and again focused on the inconsistencies and problems identified earlier, as well as on EPA's failure to make any adjustments [1]. The 2002 report found "no substantial reassessment has been done to determine whether the chemical or pathogen standards promulgated in 1993 are supported by current scientific data and risk-assessment methods" [1]. It is because of the inconsistencies, flawed methods, and outdated data used to create the Part 503 standards documented in the NRC reports and other reviews that strongly support the end to the land application of sewage sludge. There are fundamental errors in the science on which the standards are based because of inaccurate and outdated data, outdated methods, and questionable assumptions. Part 503 cannot be counted on to be truly protective of human health and the environment.

Inaccurate Data

A major problem with Part 503 is the way in which EPA determined which chemicals to regulate. Two rounds of hazard assessment and chemical selection were conducted. Round 1 identified an initial set of pollutants using hazard screening and risk assessment. Using information from studies from 1984, 200 potential chemicals of concern were initially identified, of which 50 were chosen for evaluation. These were further screened by data on toxicity, occurrence, fate, and pathway-specific hazards, and 22 chemicals were selected for potential regulation. Based on available data, a hazard index was calculated for each chemical via each of the 14 exposure pathways decided on by EPA to determine if a full risk assessment was needed for the chemical via the most limiting exposure pathway. Background exposure was eliminated from the assessment, and if the hazard index was greater than 1.0, a full risk assessment was done for the specific pathway [1]. Not including background levels is questionable because there are chemicals, like metals, for which background exposure in soil is high due to geologic properties of the area, so an additional source of exposure to the chemical could potentially elevate one's risk. Including all relevant sources of exposure would have been a better way to generate the hazard index to ensure all possible sources of exposure were assessed and included [1].

In 1988, the NSSS was conducted; it collected information on 400 pollutants from 180 sewage plants throughout the country. The EPA used this information to further screen out chemicals not at concentrations deemed to pose a risk. Chemicals were eliminated if they were banned from use, had restricted use, were no longer manufactured in the United States, had a detection frequency of less than 5 percent in the NSSS, and/or the concentrations reported in the NSSS

were so low that the estimated annual amount applied to cropland would fall below the standard annual pollution loading rate [1]. For example, even if the chemical was detected in more than 5 percent of the samples, it was not considered for further evaluation if it was no longer being manufactured.

The result of this first round of selection was regulation of 10 inorganic contaminants, and because of the criteria, all organic chemicals under consideration for regulation were eliminated. These criteria do not adequately address the adverse health effects of organic chemicals. Ignoring them does not make them or their toxic effects go away. As an example of the impact of these criteria, the selection process eliminated polychlorinated biphenyls (PCBs) because they were no longer used or manufactured, even though they were detected in more than 5 percent of samples and the concentrations would have resulted in an annual pollutant loading rate over allowable risk-based levels [1]. PCBs have not been manufactured in the United States since the 1970s but they continue to contaminate the environment and are found in sludge. Slow to degrade, they are persistent organic pollutants found all over the world and are classified as "probably carcinogenic." PCBs can bioaccumulate in animal fat, making ingestion of animal meat and milk of animals that grazed on sludge-covered land a significant concern [7]. Thus, PCB contamination is still a problem even though they have not been manufactured in almost 30 years. It is a matter of great concern that out of 200 chemicals from one study and 400 found in the NSSS, only 10 were deemed problematic, and all were metals. This is a very limited number of contaminants, and the fact that no organic chemicals were chosen raises serious questions about the validity of the methods EPA used.

A second round of evaluations was done using the 411 pollutants analyzed in the NSSS. This time chemicals were eliminated if they were not detected (254) or were detected in less than 10 percent of samples (69). Chemicals for which there was insufficient data to adequately complete the risk assessment (15) were also dropped from consideration. Of the 31 chemicals left, only dioxins, furans, and coplanar PCBs were evaluated in a risk assessment [1]. In 2003, EPA decided not to regulate these chemicals, believing they posed little risk. Yet dioxins are highly toxic and known to cause cancer and neurologic and immunologic problems. Since approximately 90 percent of dioxins in wastewater are likely to end up in sludge—and according to David Carpenter, director of the Institute for Health and the Environment at the State University of New York at Albany, "sewage sludge is the second greatest source" of exposure to dioxins for the general U.S. population—it is unclear how EPA arrived at this decision [7, 8].

The criteria that were used to eliminate chemicals in the second round of evaluations potentially missed many chemicals of concern. The 2002 NRC report found "no adequate justification for EPA's decision to eliminate from regulation all chemicals detected at less than 5% frequency in the NSSS" [1]. The NSSS reported data on a national level, which may not be representative of sludge in different locations. The contents of sludge are likely to be site-specific,

reflecting the homes and industries in the area that are discharging to local wastewater plants. Thus, for a particular type of industry that releases large amounts of certain chemicals, nationwide concentrations and frequencies appear low, but high concentrations in sludge from a specific site would be of a concern for the people receiving the sewage sludge from that treatment plant. Thus eliminating a chemical because it was detected at a low frequency in a national survey could be putting an area that does have high concentrations at risk [7]. Furthermore, eliminating a chemical because there is not enough data to do a risk assessment is irresponsible and not good science. Lack of data is a serious limitation, but "ignorance is not a solution to uncertainty" [7]. The EPA disregarded the chemicals on which there was not a lot of information as though this indicated there was not a problem with these chemicals. Lack of data is not equal to lack of risk. It means there are data gaps that need to be addressed by additional research. There might not have been enough information at the time, but these chemicals should not have been disregarded completely.

The EPA also relied on concentration data in the NSSS in the selection of chemicals to potentially regulate. The accuracy and reliability of the NSSS data have been called into question by two NRC reports [1]. Accurate concentration data is essential in assessing whether a chemical poses a risk. Errors in measurements can lead to over- or underestimation of concentrations, which in turn affect the risk estimates. The methods used by the NSSS were flawed and led to chemicals of concern being eliminated erroneously. Analytical problems and high detection limits prevented accurate measurements of chemicals. Some of the detection limits exceeded several hundred parts per million [1].

Many chemicals in the NSSS had levels of detection that were greater than EPA soil screening levels (SSLs) [1]. SSLs are soil concentrations used to determine if a risk assessment is required at a Superfund site, and they are risk-based conservative assumptions. The 2002 NRC report re-assessed eight organic chemicals and found that five of them had limits of detection higher than their respective SSLs [1, 9]. Thus the NSSS results were "not sensitive enough to detect pollutant concentrations that, if present in soil at a Superfund site, would have triggered a risk assessment" [9]. Hexachlorobenzene, a persistent organic pollutant considered a probable human carcinogen, is an example of a chemical that was eliminated because it was not detected in any of the samples. However, the limits of detection ranged from 5 to 100 mg/kg, while the SSL is 0.1 to 2 mg/kg, depending on the route of exposure [9]. Analysis of recent data on chemicals in sludge showed that the majority of reported hexachlorobenzene levels exceeded the lowest SSL [9]. Thus, the NSSS failed to achieve low enough detection levels to adequately determine if the concentrations present required further action.

The NSSS concentrations were used to calculate the hazard indexes to determine if a full risk assessment for a specific chemical via the most limiting exposure pathway should be done. Even if the hazard index for a chemical was

greater than 1, if the chemical was detected infrequently, it was eliminated [1]. Given that the detection limits were so high, it is unclear how many of these chemicals were incorrectly identified as having low frequencies and/or concentrations. If more sensitive detection limits had been used, many more chemicals of concern would have been selected to be evaluated further and possibly regulated. The NSSS data lack credibility, given that the limits of detection were so high that chemicals were missed but would have warranted assessment under different conditions. Every analytical method has a limit of detection, but the goal is to have consistent and low detection limits. One wants to be able to detect the lowest concentration present with the greatest accuracy possible. The fact that NSSS had unreliable data undermines all the standards in Part 503 [1, 7, 9]. How can these standards adequately protect human health and the environment given that chemicals were erroneously eliminated and never assessed because of poor science?

Outdated Exposure Assessment Methods and Flawed Assumptions

After choosing the chemicals to be included in the risk assessment, human exposure to sewage sludge by various exposure routes was assessed to calculate risks. For 14 exposure pathways, the risk associated with each pathway for each contaminant was assessed separately; risks from multiple pathways or from exposure to multiple chemicals were not examined. Current practice is to perform a risk assessment after aggregating all the pathways to which a single individual is likely to be exposed to in order to have the most complete exposure assessment. Part 503 assessed exposure assuming one would be exposed via only one pathway, which is not realistic. This method severely underestimates risks because it is highly unlikely one will be exposed to a chemical in the soil via only one route. It is much more likely that a child playing in the soil will have incidental ingestion of the soil, ingestion of plants that grew in the soil, ingestion of animals that grazed on grass that grew in the soil, and dermal contact with the soil, all contributing to the child's exposure to the chemicals. Exposure to a single pathway might not pose a significant risk but once all the pathways are combined, there could be a very different outcome [1, 7].

The EPA also used limited exposure pathways, assessing inhalation only for sludge applicators, not residents. The EPA also assessed only chronic exposure, but there is a risk of short-term exposure to volatile compounds. Volatile organic compounds were eliminated because EPA believed release occurred during the wastewater processing that produced the sludge. However, when sludge is applied, it can release volatile organic compounds (VOCs) such as sulfur- or nitrogen-containing compounds, acids, aldehydes, and ketones [1, 7]. There was also inadequate assessment of pathogen risk. Movement of pathogens to groundwater was not addressed completely, nor was exposure to pathogens in dust and

aerosols after land application of sludge. Exposure to radioactive chemicals was not addressed at all [7].

In generating risk calculations, EPA had to make many assumptions. A number of "untenable assumptions" were made and probably led to underestimation of risk [7]. A very limited risk assessment for groundwater contamination was conducted in Part 503, and contamination of waterways was not adequately assessed. The EPA assumed metals cannot leach into groundwater, but recent data has shown that metals exhibit facilitated transport, by which they attach to organic chemicals and travel to groundwater; metals can also move through flow paths created by worm holes or root channels [7, 10]. Also, the rate of contaminant movement in soil that was calculated was much slower than what actually occurs. The rate was not based on actual field data but on data from a single paper based on test tube motility tests from a single soil type [10]. Contamination of surface and groundwater is an area of great concern. Runoff or leachates from land that received biosolids is a significant source of exposure, and it is likely that important water resources could become contaminated, exposing people to the chemicals in drinking water that originated in sludge [10]. Not considering this exposure severely underestimates risk.

When determining cancer risk resulting from sludge application, EPA decided to use the less restrictive value of 1 in 10,000 as an acceptable level of cancer risk compared to what is used in most other regulations to determine cancer risk and influence regulations, including the drinking water standards, of between 1 in 10,000 and 1 in 1,000,000 [7]. When questioned on why this value was used, the EPA acknowledged it was a less restrictive number and was chosen as a policy decision because the agency considered the overall risk from sewage sludge was "especially low" and the more restrictive value would have an economic impact, and it was "difficult to justify such an expense for little or no actual difference in risk" [11]. For soil ingestion, only ingestion as a child was calculated even though incidental ingestion can occur throughout adulthood, especially for home gardeners [1]. Dietary intake of sewage sludge is a critical pathway, and EPA based its recommendations on dietary intakes from the late 1970s. American diets are very different now with regard to vegetable and fruit consumption, meat intake, and water consumption. Comparing the dietary assumptions EPA used with the current food pyramid guidelines shows that the current dietary recommendations specify 16 times the amount of fruits and vegetables that was assumed in developing the Part 503 standards. This is significant: for example, for cadmium, changing only the dietary assumptions, the standard drops from 39 ppm to 15 ppm [1, 7].

The EPA also assumed that the degradation products of organic chemicals were less toxic than the original chemical, but this is not always the case. Surfactants are a group of chemicals found in sludge, and the degradation products of the surfactant alkyl phenol ethoxylate are significantly more toxic than the original compound. The anaerobic digestion process at treatment plants

actually promotes this transformation, resulting in a much more toxic compound in the sludge [10]. Uptake by plants and animals is critical to assessing exposure to and risk from sewage sludge, and EPA used very low plant uptake coefficients and low ingestion rates for grazing animals. Many of the soil uptake coefficients are based on plants grown in greenhouses, but these conditions have been shown not to reflect how metals behave in biosolids [1, 7]. The EPA assumed its uptake coefficients would be applicable to all plants under all soil conditions, but uptake differs greatly across plants and soil conditions, so the numbers used were not highly protective [7]. The EPA also assumed that metals would be bound to the sludge, limiting the uptake by plants, but they did not assess if this was reversible due to soil changes or if continual application of sludge changed these parameters [1].

When doing a standard risk assessment, one accounts for the assumptions made and the uncertainties still present by incorporating safety or uncertainty factors. This was not done by EPA for the Part 503 standards [7]. Taken together, the incomplete exposure assessments and flawed assumptions probably lead to an underestimation of exposure to sewage sludge, indicating that the standards are not adequately protective.

PROBLEMS WITH REGULATED CHEMICALS

There are also problems with the chemicals for which there are standards. Arsenic is regulated in Part 503 as a noncarcinogen. However, arsenic is an established cause of skin cancer via ingestion of drinking water, and there is evidence that it also causes lung and urinary bladder cancer. There are no data to suggest that arsenic ingested in soil behaves differently from arsenic ingested in drinking water [1]. With cadmium, ingestion is a significant route of exposure. The EPA looked at ingestion of soil only for a child even though the reference dose is based on ingestion over a lifetime. Exposure as a child and as an adult should have been assessed. Furthermore, cadmium is well taken up by plants so exposure via multiple pathways of ingestion should have been analyzed to better assess risk. Recent studies also suggest that cadmium is an endocrine disruptor, an endpoint not assessed in Part 503 [1, 10]. The mercury assumed to be in the sludge was considered to be similar in toxicity to the inorganic form mercuric chloride. However, mercury can appear in many forms and the speciation is critical to its fate and transport. The organic form methylmercury has been found in sludge. This is of great concern because it can bioaccumulate in fish. Inhalation exposure to nickel is the most toxic pathway, but this was not thoroughly assessed. Molybdenum has no standard, just a ceiling concentration, but it is well known that molybdenum is toxic to ruminant animals, which are exposed by ingesting legumes, grasses, soybeans, and other crops [1, 10].

NEW CHEMICALS AND PATHOGENS

Another significant problem with Part 503 repeatedly discussed in the literature is that thousands of new chemicals have been produced, used, and released since 1990, and there are new pathogens of concern that have not been considered since the initial standards went into place. The Toxics Release Inventory tracks releases of over 600 toxic chemicals, of which only nine are currently being regulated in sludge; thus very few of these 600 chemicals have been assessed. Brominated flame retardants, antibacterials, pharmaceuticals, fragrance chemicals, surfactants, personal care products, and organotins are just a few of the new chemicals of growing concern. Kinney et al. (2006) analyzed organic wastewater contaminants in nine different sewage sludge products [12]. The most commonly detected chemicals were pharmaceuticals, detergent metabolites, steroids, fragrances, polycyclic aromatic hydrocarbons (PAHs), fire retardants, plasticizers, and disinfectants. Nonylphenol and octylphenol detergent metabolites, known or suspected endocrine disruptors, were detected in greater concentrations than most of the other chemicals measured. Polar compounds were also found at concentrations higher than previously thought possible. Harrison et al. (2006) examined peer-reviewed literature and official government reports to assess the presence and concentrations of organic chemicals in sewage sludge [9]. Data were found for 516 chemicals. There were SSLs for 15 percent of the chemicals, and for 86 percent of these, the reported maximum concentration exceeded the SSL. In 2006–2007, EPA conducted a new analysis of 145 chemicals in sewage sludge, including anions, metals, polycyclic aromatic hydrocarbons, semi-volatiles, flame retardants, pharmaceuticals, and steroids/hormones [13]. Twenty-seven metals were found in virtually every sample; four VOCs were in 72 samples; three pharmaceuticals were in all samples, and nine were in at least 80 samples; three steroids were in all samples, and six were in at least 80 samples; and all flame retardants except one were in every sample. The EPA states that it plans to evaluate the pollutants identified in the survey, first focusing on the nine they had previously determined to be of concern, but if EPA conducts the risk assessment in the same manner as was done for Part 503, the results will again have little credibility.

HEALTH EFFECTS

Occupational exposure to Class B biosolids is considered a concern by the U.S. Centers for Disease Control and Prevention (CDC) and the National Institute for Occupational Safety and Health (NIOSH) due to the pathogens still present in biosolids. Health effects after occupational exposure have been reported in numerous studies [1, 14]. There is little data regarding health effects in the general population exposed to sewage sludge. Two recent studies assessing health effects from exposure to aerosols after sewage sludge application to nearby lands suggest increased risk for certain respiratory, gastrointestinal, and other

diseases as well as irritation of the eyes, throat, and lungs and prevalence of *Staphylococcus aureus* infections [10]. The highly publicized case of Andy McElmurray and his dairy farm ruined by the application of sewage sludge further highlights the fact that there are health concerns associated with the application of sewage sludge. One of the chemicals found in Andy McElmurray's sludge was thallium, a metal not regulated under Part 503 [15]. The bacterium *Listeria monocytogenes* has been detected frequently in treated sewage sludge, and crop contamination has been observed when sludge containing this pathogen has been applied [1]. Even these few cases raise significant doubt regarding how protective the standards in Part 503 really are.

TOO COMPLEX TO REGULATE PROPERLY

The recent studies on the composition of chemicals in biosolids show the fundamental problem with sewage sludge: it is a complex, always-changing mixture. Even if major changes were made to the standards, there are too many unknowns regarding the amounts, behaviors, and toxicity of thousands of chemicals that are found in sewage sludge to regularly ensure the protection of human health. Sewage sludge is too complex to properly monitor and regulate. In a 2006 study examining reported organic compounds in sludge, of the 516 organic chemicals that had available data, 83 percent of the chemicals were not on the priority pollutant list and 80 percent were not on the target compound list of chemicals that must be detected and quantified in analyses of soil from Superfund sites, leading the authors to conclude that even if monitoring were expanded to include chemicals on these lists, it "will not capture the vast majority of chemicals that may be present" [9]. It is significant that this study found data on only 516 chemicals even though thousands are in use.

There are too many variables and too many unknowns to properly regulate the land application of sewage sludge in a way that adequately protects human health and the environment. The EPA assumes "that models approximating the reality of a ranch in west Texas are also appropriate for a vegetable farm in New York" [7]. This could not be further from the truth. The components of the wastewater, type of treatment process, application rates, climate, and soil characteristics vary greatly from location to location, and these are just a few of the numerous factors that impact the fate, transport, bioavailability, and toxicity of the chemicals in sewage sludge. People are not exposed to just one chemical. It is difficult enough to assess risk for one chemical, and adding multiple chemicals makes the assessment infinitely more difficult. Evaluating risk posed by individual chemicals requires multiple assumptions; adding in mixtures means more assumptions have to be made and this can lead to unacceptably high levels of uncertainty [1, 5, 7].

The 2002 NRC report concluded that it was "not possible to conduct a risk assessment for biosolids at this time (or perhaps ever) that will lead to risk

management strategies that will provide adequate health protection without some form of ongoing monitoring and surveillance," because sewage sludge is a complex mixture that can change unexpectedly over time and place [1]. It is impractical and financially impossible to continually monitor sewage sludge for every type of chemical that could be in it. For many of the chemicals, much is unknown: how they interact with other chemicals, the form that is found in sludge, how bioavailable they are, and how toxic. How can sewage sludge be properly regulated if there is not complete information on all the chemicals present in it and the variables that govern their fate in the environment? Ignoring the unknowns is not the answer.

Inadequate enforcement of rules and practice adds to the problem. The EPA itself says the Part 503 regulations are "self-implementing" [7]. Periodic reporting is required, but no permits are needed for land application and no record-keeping regarding application rates is required. After application of Class B biosolids, there are waiting periods from 30 days to one year. However, the rules for enforcement are vague, and there is no testing required after the time limit to ensure that natural processes have reduced the pathogens to safe levels [7]. A recent example exhibits the consequences of inadequate enforcement. In a county in Alabama, the blood of 200 residents is being tested for the presence of perfluorinated chemicals in drinking water. The chemicals were released from nearby industries and concentrated in sewage sludge, which was distributed as free fertilizer for 12 years. The EPA knew the chemicals were in the sludge but did not know the sludge was being applied to agricultural land until finding out by accident in 2008 [16].

The federal Clean Water Act defines sewage sludge as a pollutant, and it needs to be treated as one. It is not a fertilizer with soil-conditioning properties. Sludge is a complex mixture that contains organic, inorganic, and biological pollutants from wastewater coming from a variety of sources [5, 6]. Basically, anything flushed down the drain or toilet can make its way into sludge. The point of a wastewater treatment plant is to make the effluent as clean as possible. In doing so, the sludge becomes more toxic as it concentrates the pollutants that were in the liquid sewage [5]. Although EPA believes the standards in Part 503 are keeping the public safe, the "data gaps and non-protective policy choices result in regulations that are not adequately protective of human health and the environment" [7]. There are other methods to manage sludge that are more environmentally friendly and safer that need to be investigated [6]. Until the Part 503 standards are reevaluated using more current and reliable data and methods, the practice of land application must be discontinued because that is the only way to protect human health and the environment. The data strongly support that applying sewage sludge to land is not safe, and if things continue as they are, the long-term consequences to human health and the environment have yet to be felt.

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FREQUENTLY ASKED QUESTIONS (FAQ'S):

QUESTIONS, ANSWERS & FACTS ABOUT BIOSOLIDS IN NOVA SCOTIA

By NSEN Biosolids & Waste Water Caucus

(1) What are Biosolids? Biosolids are organic stabilized material produced during treatment of domestic, commercial, hospital, industry, and street run-off sources of sewage and septic sludges. They include the residue removed by waste water treatment processes but do not include screenings and grit. They are removed during the preliminary treatment stages. Biosolids differ from sewage and septage sludges in that they have been treated to remove pathogens. Biosolids are used as a soil amendment or fertilizer for use on agricultural land in Nova Scotia. N-Viro Systems Canada is one of the largest generators of biosolids in the province.

(2) How are Biosolids made? The treatment process includes the reduction of volatile solids by 38% or if sufficient alkaline material is added to sludge, such as cement kiln dust or fly ash, a highly alkaline material is produced. The alkaline content produces a 'corrosive' environment sufficient to reduce the numbers of microorganisms and can help decrease acidic soil conditions common in parts of the province. The alkaline mix also reduces soil availability of some metals and chemicals but has no effect on some and increases the availability of others.

(3) Are there different classes of Biosolids? N-Viro produces Class A biosolids which meet a higher standard for heavy metal concentration and pathogens than Class B biosolids. Class B biosolids, generated at other facilities, are not treated or "stabilized" to the same extent as Class A biosolids and are no longer permitted for use on agricultural lands. Class B biosolids have recently been approved to be placed on public areas such as golf courses, recreational lands, land reclamation sites, trails, forests, and in commercial sod.

(4) What nutrients are found in Biosolids? Biosolids are rich in nutrients such as nitrogen and phosphorous, and contain trace elements such as sulfur, magnesium, calcium, copper, and zinc. However, it is also important to note that many of the earth's major bodies of water are now developing large 'blooms' of algae overgrowth, resulting from excessive pollution of watercourses by fertilizers and human waste nutrient sources and, because they consume vast quantities of available oxygen, are responsible for killing off substantial amounts of marine life forms.

(5) What contaminants are found in most urban sludges? The Environment Protection Agency has completed, in 2009, a sludge survey which determined that most urban sludges contain the following:

- 30 metals (calcium, lead, zinc, arsenic, mercury, etc.)
- 6 polycyclic aromatic hydrocarbons (pyrene, furanthene, etc.)
- 6 semi-volatile organics (chloraniline, fluoride, etc.)
- 2 inorganic anions (nitrate, nitrite)

- 6 polybrominated diphenyl ethers (flame retardants)
- 40+ Antibiotics (penicillin, cephalosporins, fluroquinolones, etc.)
- 25 Steroids and Hormones (campesterol, estriol, desmosterol, etc.)
- 40+ Disinfectants, Antimicrobials, and Other Drugs (digoxin, codeine, soaps, perfumes, cleaners, etc.)

In addition to the above, sludges may have a potential to contain, yet not be tested for, illicit drugs, chemotherapy and radiation residues, chlorinated pesticides, and numerous bacteria, viruses, parasites, fungi, protozoa, and prions.

(6) How and what is N-Viro testing for in sewage sludge? The Nova Scotia Department of Environment's guidelines recommends that testing be done for 11 heavy metals, two bacteria, dioxins, and furans on every 1,000 tonnes of treated sludge. The generators of the product are conducting their own post-treatment sampling and sending to a laboratory of their choosing. Only one sample from 10,000 tonnes of treated sludge is required to test for: industrial chemicals; flame retardants, alkyphenols, ehtoylates, pharmaceuticals, hormones and steroids, personal care products, and others. No indication is given as to what specific contaminants are being sought, or at what concentrations they will be detected, or at what levels these contaminants are to be considered 'toxic'. It is noteworthy, that most Canadian labs do not have adequate equipment that can test for pollutants properly that would give any meaningful results. Additionally, some tests, such as a highly sensitive test for flame retardants, can cost upwards of $1,500 US each, making such testing a burden on taxpayers.

(7) Who is permitted to use Biosolids? According to the guidelines, Class A biosolids may be used by a farmer on any crop or forage field. Farmers are recommended to respect separations distances, slope of land, risk of flood areas, waiting periods and having a nutrient management plan in place for the farm. Class A biosolids users need no approval from the Department of Environment. Generators of biosolids must hold an approval, however. The department's Approval # 2005-0455546-A02 states that, "The Approval Holder shall communicate to the general public the use of Exceptional Quality/Class A biosolids generated at the Facility. The communication plan shall include, but not be limited to, local and, provincial media, contact with municipalities where product will be utilized, and a contact phone number where enquiries may be directed." This is not being done in communities receiving N-Viro or any other Class A biosolids.

(8) When did the practice of using treated sewage sludge as a soil amendment start in Nova Scotia? Although some municipalities allow sewage sludge to be put on farmland over past years, many others do not. N-Viro began processing biosolids in 2008, and immediately began land applying it once the Department of the Environment granted its Approval.

(9) Did the Province first consult with farming communities and engage in discussions with rural residents before approving the use of Biosolids on their agricultural land and in and around their communities? No. If asked, farming communities would have questioned the merit of

having urban waste products being transferred from HRM to rural landscapes. Broad public opinion considers food "tainted" if grown or raised using biosolids. The farming community would, undoubtedly, have expressed concerns that the use of biosolids by some farmers could negatively impact the buy local initiatives fostered by the Department of Agriculture.

(10) Is there problem with foul odors or particulate matter entering the air from land spreading of Biosolids? Yes. Cement kiln dust is very particulate and irritating to airways because of its caustic properties. Cement kiln dust may also contain numerous heavy metals such as the carcinogen, thallium. Dust particles may contain dead pathogens and fungi and can cause allergic alveolitis. Heavy metals such as beryllium are damaging to the lung tissue when inhaled. Problems with foul odors are a problem with neighbors and, although temporary, can negatively impact property values and the quality of living in rural communities.

(11) Is land application of bio-solids safe? No. The following adverse effects have been noted on lands following treated biosolids application:

- Loss of soil fertility (excessive loading of soils with heavy metals - copper, zinc, molybdenum - destroy good soil microorganisms)
- Contamination of ground water sources with pollutants (copper, lead, zinc, and pesticides via facilitated transport onto organic molecules)
- Regrowth of pathogens in biosolids after mixing with soil (potential for food-borne illnesses – salmonella, coliform bacteria, viruses, prions)
- Contamination of soils with persistant and bio-accumulative toxins, such as heavy metals (lead, cadmium - breast cancer), persistent and volatile organic pollutants (such as flame retardants - carcinogenic), hormones/steroids (affect human and wildlife reproductive function), any thousands of other chemicals
- Uptake and storage of contaminants by some plants/forages
- Livestock illness and death (nutrient imbalances, direct ingestion of biosolid contaminants/pathogens in forages)
- Risk of liability to end users (farmers) regarding complaints of human or animal illness, environmental contamination , and potential loss of property value.

(12) Is it safe to eat foods grown or raised on soils where Biosolids are used? No. It has been well documented that livestock ingesting biosolid treated fields can store contaminants in body fat and milk glands (thallium and flame retardants are both lipophillic (fat loving) and carcinogenic. Some plants, such as green leafy vegetables take up and store heavy metals (lead, molybdenum, etc). Incidences of cancer, respiratory diseases and food –borne illnesses are on the rise and we must consider that land application of toxic sludge could be a major contributor.

(13) How will I know if Biosolids are being used in my community? Unfortunately, you won't know. Regulations as set out in the Approval process and recommendations put forth in the provincial guidelines are already being ignored (see question 7). Asking whether a particular farmer is using the product will most likely result in denial. Asking the generator of biosolids to disclose

who is using their product and where it is being used will not be answered either. Trucking companies hauling the product to agricultural destinations are not permitted to disclose who the end users are. The Department of Environment has facilitated and promoted the biosolids industry to be non-transparent and non-liable for negative impacts. Citizens remain uninformed and unengaged.

(14) Can Nova Scotians expect to see products resulting from Biosolids use be labeled as a means to promote informed consent on consumer goods? Not likely. There are no plans to label foods grown using treated sewage sludge as fertilizer. The reason may be simple if you consider that, by adopting the practice of non-labeling, any complaints of ill health or environmental contamination resulting from product use would be virtually untraceable back to the users or generators of the product.
The Organic Council of Nova Scotia prohibits the use of any form of human waste/sewage products to be used in growing/raising of food stuffs. Organic farmers consider biosolids being used as fertilizer as an unethical and unsustainable land stewardship practice and warn that users of biosolids may not be allowed to convert to organic status in the future.

(15) Who is bearing the burden of costs of process and to transport this product to rural agricultural districts? Taxpayers paid for N-Viro's $12.5 million plant and the facility claims that it costs $175.00 to process one tonne of biosolids. No mention is made whether this cost includes transportation from the waste water plant to the facility or if taxpayers are subsidizing the transportation from the facility to rural communities. Selling price to farmers is $19.00 per tonne.

(16) Are farmers being advised of the potential risks of contamination to soil and/or groundwater sources? Of the potential for reduced fertility of soils? Of the potential for human and/or livestock illness? Of the potential to be legally bound to foreclose whether or not they have used bio-solids on their crop or forage fields at the time of sale or transfer of property?
Farmers may not be fully aware of the potential risks associated with using biosolids in the context described above. Some farmers may not be overly concerned about risks because chemical fertilizer costs are soaring. Even though many farmers have nutrient management plans or land application plans for fertilizer use and would receive instruction about how much needs to be applied at any given location for maximum benefits, some may not be aware that nutrient management planners may also pose as consultants or promoters for the biosolids industry.

(17) Who will be liable if agricultural soils and/or groundwater sources are contaminated, if soil fertility is lost, or property salability decreases as a result of bio-solids being used? The Department of Environment stated that it will be the end user who will be responsible. Without labeling or public knowledge about where product was used and by whom, perhaps no one will be ultimately liable for any resultant negative impacts.

(18) What are the risks to our agricultural industry if public perception about food safety of locally-produced goods is questioned? It is believed that the ramifications to the buy local initiative will be hugely negative as the practice of using a potentially toxic fertilizer on food crops or forage crops will undermine consumer confidence and eliminate any local advantage in selling local foods. This, in turn, will favor commercial enterprises which import from low wage labor communities and where their farming practices are unknown as concerning food safety and environmental issues.

(19) Are there other disposal alternatives for Biosolids? Yes. Landfilling is no longer an option in Nova Scotia for wastes considered "organic". Lagooning, wetlanding, and compost toilets are all natural options whereby a variety of organisms degrade human waste rendering clean water. Another alternative is fluid bed incineration where the sludge is reduced to a negligible mass and pollutants are destroyed without entering the atmosphere by high heat application. Plasma assisted oxidation of sludge (PASO) is a newer technology which creates energy from the UV irradiation of sludge which destroys contaminants and reduces the mass of sludge substantially. BioGas generation is another option whereby methane gas is produced by anaerobic digestion of waste material.

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April 26, 2011

To the CRD Environment Committee and the Saanich Peninsula Waste Commission,

Sierra Club BC would like to re-assert its strong support for the CRD ban on the land application of bio-solids passed by the Core Area Liquid Waste Committee last year, and to express our concern and opposition to the production and distribution of PenGrow by the Saanich Peninsula Waste Commission.

It has recently come to our attention that the Saanich Peninsula Waste Commission is planning to expand the PenGrow program to farmland on the Saanich Peninsula. At a time when the public is more and more supportive of, and aware of, the importance of local food production, the land application of bio-solids seems to directly contradict the responsibility of the CRD to protect the region's farmland, natural environment and public health.

As such, we support the motion before the CRD Environment Committee to prohibit the sale, storage and production of PenGrow on CRD facilities or Parks, and urge the Saanich Peninsula Waste Commission to re-consider the land application of bio-solids on local farmland.

Best regards,

George Heyman

Executive Director
Sierra Club BC

Islands Organic Producers Association (IOPA)
http://www.certifiedorganic.bc.ca/cb/iopa.php
April 24, 2011

Phillipe Lucas
plucas@victoria.ca
Victoria Councilor
CRD Board of Directors

Re: Use of bio-solids/sewage sludge on farmland.

This letter is to show our concern and opposition to the use of sewage sludge/bio-solids on farmland. The Canadian Organic Standards, which are followed by the bioregional certification body IOPA, prohibit the uses of sewage sludge on organic farms. http://www.cog.ca/index.php?page=organic-regulations.

Heavy metals found in sewage sludge adversely affect soil microbes that play a significant part in plant growth in organic soils. An article in the Journal of Industrial Microbiology confirms that long-term effects are unknown.

Article found in Journal of Industrial Microbiology. Vol 14. 94-104

Long-term effects of metals in sewage sludge on soils, microorganisms and plants

Steve P. McGrath, Amar M. Chaudri and Ken E. Giller

Abstract
This paper reviews the evidence for impacts of metals on the growth of selected plants and on the effects of metals on soil microbial activity and soil fertility in the long-term. Less is known about. This is not surprising few long-term experiments exist.

It is commendable to search for the opportunity to wisely recycle. We know that Class A bio-solids are treated to reduce most pathogens but what of the cleaners, hormones, medications that find their way into the system when flushed or poured down the drain? The EPA in a study by the National Research Council also has concerns about excess nitrate-nitrogen in regards to long-term impacts on water systems. http://www.epa.gov/waterscience/biosolids/nas/complete.pdf.

We are concerned that the long- term impacts of bio-solid application to farmland on water and soil systems are not conclusive. In using bio-solids we many be overlooking potential problems for the farmland foods that come to our tables.

Yours respectively,
Tina Baynes
Director of IOPA

DOGWOOD *initiative*
Solutions for Lands and People

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To the CRD Environment Committee and the Saanich Peninsula Waste Commission,

The Dogwood Initiative would like to re-assert its strong support for the CRD ban on the land application of bio-solids passed by the Core Area Liquid Waste Committee last year, and to express our concern and opposition to the production and distribution of PenGrow by the Saanich Peninsula Waste Commission. (SPWC)

It has recently come to our attention that the SPWC is planning to expand the PenGrow programme to include a pilot project on the land application of bio-solids on farmland on the Saanich Peninsula. Although we have great sympathy for the current economic plight of local farmers, we feel that this strategy will inevitably degrade the value and safety of local food and have inevitable negative effects on the local environment. At a time when the public is more and more supportive of and aware of local food production, land application of bio-solids (even on grazing land) seems to contradict the responsibility of the CRD to protect our farmland, natural environment and public health, and flies in the face of food security goals outlined in the Regional Sustainability Strategy.

As such, we support the motion before the CRD Environment Committee to prohibit the sale, storage and production of PenGrow on CRD facilities or Parks, and urge the Saanich Peninsula Waste Commission to re-consider the land application of bio-solids on local farmland.

Sincerely,

Gordon O'Connor
Vancouver Island Campaigner
Dogwood Initiative