

1. BACKGROUND

Land application of sewage biosolids is desirable because it contains nitrogen and phosphorus that are valuable plant nutrients. Sewage biosolids also contain organic matter that improves soil structure and water holding capacity. On the other hand, sewage biosolids can contain a variety of contaminants such as heavy metals and pathogens. Therefore, proper management of sewage biosolids application to land is very important to ensure this practice does not adversely affect human and animal health and the environment. Generally, the heavy metals and organic contaminants are derived largely from industrial wastewater, and the pathogens are derived from human waste.

Land application of sewage biosolids has been widely practiced in North America (including Ontario) and Europe for many decades. Over the years, regulations and procedures have been developed to manage biosolids land application to protect human and animal health and the environment. A considerable amount of scientific study has been undertaken to support the development of the regulations, and to confirm the effectiveness of the application procedures. In Ontario, sewage biosolids must be stabilized in the sewage treatment process to reduce organic matter, odour and pathogens before being applied to agricultural land. The public, however, still has concerns that land application may be unsafe because it involves human and industrial waste. Numerous questions have been raised privately and in the media about the potential health and environmental effects of land applying sewage biosolids.

Although diverse, public concern has tended to coalesce around the fate and effects of:

- pathogens;
- heavy metals; and
- organic contaminants

on human health, and on soil and surface and ground water qualities.

This study is part of an ongoing effort by the Water Environment Association of Ontario (WEAO) and government (federal, provincial and municipal) to:

- update current knowledge on the fate and significance of trace contaminants in sewage biosolids applied to agricultural lands;
- identify knowledge gaps; and
- recommend future research priorities.

Various stakeholders and experts were consulted, through questionnaires and a Stakeholder Workshop, to assist in identifying a list of "Contaminants of Concern".

2. INTRODUCTION

2.1 Project Partners

The Water Environment Association of Ontario (WEAO) undertook this project in conjunction with the following partners:

- Ontario Ministry of the Environment (MOE)
- Environment Canada
- Ontario Ministry of Agriculture, Food and Rural Affairs (OMAFRA)
- Regional Municipality of Waterloo
- City of Toronto
- Regional Municipality of Durham
- City of Windsor
- Regional Municipality of Niagara
- Regional Municipality of Halton
- City of Peterborough
- City of Ottawa
- City of Hamilton

The partners served on a Technical Steering Committee that oversaw the project and met periodically to review progress, project deliverables and to participate in decision making for recommendations as required in the project objectives.

2.2 Study Objectives and Phases

The objectives of the study were:

1. Review, assess, and summarize information assembled from literature and consultation with credible non-government organizations, farming associations, experts and regulatory agencies (Stakeholders) regarding the fate and significance of contaminants in sewage biosolids after they are applied to agricultural lands.
2. Use the information to divide specific contaminants in sewage biosolids into two groups:
 - Group I – Contaminants which have sufficient credible scientific information to assure the public that the current agricultural land application program/guidelines are adequate to protect the well being of soils, crops, animals, human health, ground and surface water qualities.
 - Group II – Contaminants which do not have sufficient credible scientific information to assure the public that the current agricultural land application program/guidelines are adequate to protect the well beings of soils, crops, animals, human health, ground and surface water qualities

2. INTRODUCTION - cont'd

3. Recommend a long-term study program that would allow sewage biosolids generators, the federal and provincial government agencies to complete the information gaps and identify actions that would be necessary to mitigate any adverse effects that may be caused by the presence of specific contaminants in sewage biosolids. The studies should be prioritized based on needs and consensus reached between Stakeholders.
4. Provide comprehensive Terms of Reference for the top three studies identified in the long-term study program.
5. Disseminate the study findings and recommendations to the Stakeholders and municipalities through report(s) and workshop(s). The information package should also include a well-organized bibliography (by topic/issue and where the reference materials can be obtained/ordered) and one copy of an appendix containing all the reference materials reviewed by this study.

The project consisted of a number of phases, as follows:

- Phase 1 – Collect Stakeholder (including Workshop) Information
- Phase 2 – Identify Contaminants of Concern
- Phase 3 – Conduct Literature Search
- Phase 4 – Draft Report on All Identified Contaminants of Concern from Literature Review
- Phase 5 – Draft Report on Recommended Study Program
- Phase 6 – Develop Terms of Reference for Top Three Studies
- Phase 7 – Prepare Final Report
- Phase 8 – Public Seminars

2.3 Project Consultant

The consulting team consisting of R.V. Anderson Associates Limited (RVA), M.D. Webber Environmental Consultant (Dr. Mel Webber), and Specialists in Energy, Nuclear and Environmental Sciences (SENES) Consultants Limited (Dr. Harriet Phillips) was retained in September 1999 to undertake the project, based on a proposal dated August 9, 1999.

2.4 Interim Report

In order to ensure that the methodology being followed for the study would satisfy the Technical Steering Committee, an Interim Report was prepared after Phase 2 was completed. This interim report included two selected contaminants (heavy metals, and endocrine disruptors – including hormones).

3. STAKEHOLDER INFORMATION

Information was solicited from a broad range of stakeholder and expert groups, concerning current and emerging issues and research/regulatory programs relating to the agricultural land application of sewage biosolids. Information gathering focused on the fate of metals, organic contaminants, pathogens, and other contaminants in land-applied sewage biosolids and on their significance for soil, crops, surface and groundwater quality, and human and animal health.

3.1 Stakeholder and Expert Groups

Information was solicited from the stakeholder and expert groups identified in Table 3.1.

TABLE 3.1
Stakeholder and Expert Groups

<p><u>Non-Government Organizations</u></p> <ul style="list-style-type: none"> • World Wildlife Fund¹ • Toronto Environmental Alliance² • Greenpeace^{1,2} • Ontario Environmental Network • Sierra Legal Defence Fund • Canadian Environmental Law Association 	<p><u>Regulatory Agencies</u></p> <ul style="list-style-type: none"> • British Columbia Ministry of the Environment^{1,2} • Alberta Environment • Canadian Food Inspection Agency^{1,2} • United States Environmental Protection Agency (USEPA) – Washington¹ and – Cincinnati¹ • Agriculture and Agri-Food Canada^{1,2} • Grand River Conservation Authority^{1,2}
<p><u>Farm Associations</u></p> <ul style="list-style-type: none"> • Ontario Soil and Crop Improvement Association^{1,2} • Ontario Farm Environmental Coalition¹ • Ontario Federation of Agriculture^{1,2} 	<p><u>Other Expert Groups</u></p> <ul style="list-style-type: none"> • Cornell University - Waste Management Institute • University of Minnesota¹ • University of Guelph¹ • United States Department of Agriculture • Danish Environmental Protection Agency • German Standards Institute • Water Research Centre – England¹
<p><u>Industry</u></p> <ul style="list-style-type: none"> • Ontario Association of Sewage Industry Services^{1,2} • Ontario Clean Water Agency 	<p><u>Municipalities</u></p> <ul style="list-style-type: none"> • Technical Steering Committee member municipalities^{1,2} • Association of Municipalities of Ontario (including the Rural Ontario Municipalities Association)^{1,2}

Note: Bold indicates the following participation -

¹ Completed questionnaire

² Attended Workshop

3. STAKEHOLDER INFORMATION – cont'd

3.2 Stakeholder and Expert Survey

Stakeholders within Ontario, and experts outside Ontario, were surveyed by questionnaire. Initial contact was made by telephone after which the questionnaire was forwarded. It solicited information about understanding and perceptions of land application issues and research/regulatory programs. Follow up of the questionnaires was by telephone and email.

Twenty-seven (27) survey questionnaires were sent out of which sixteen (16) were completed and returned. Completed questionnaires are included in Appendix A.

A summary of the survey questions and responses is presented below:

- 1) *Identify contaminants that are adequately regulated by the biosolids guidelines in your jurisdiction.*

Ontario: 8 respondents
Metals (6), nutrients (2), pathogens (1), organic contaminants (1)

Outside Ontario: 3 respondents
Metals (3), pathogens (2), organic contaminants (2), all contaminants (1), maybe dioxins and furans (1)

- 2) *Identify contaminants that are not adequately regulated by the biosolids guidelines in your jurisdiction.*

Ontario: 7 respondents
Metals (1), pathogens (5) [bacteria, viruses, parasites], organic contaminants (6) [endocrine disruptors, pharmaceuticals, dioxins, polychlorinated biphenyls]

Outside Ontario: 3 respondents
Maybe not in order of priority - pathogen sampling for retail products; nitrates; *p*-cresol; phenol; phenanthrene; naphthalene; heavy extractable petroleum hydrocarbons; dioxins and furans; endocrine disruptors.

Formal full-scale risk assessment of pathogens.

Potential synergistic effects of the many organic contaminants present at low levels in biosolids.

Potential ecological impacts of land application of biosolids.

3. STAKEHOLDER INFORMATION – cont'd

- 3) *Are your concerns the same for biosolids application to lands with limited public access and high public exposure lands?*

Ontario: 4 respondents
Concerns the same but level of processing and quality control should be more strict where public exposure is greatest.

Direct public exposure to agricultural land may be limited but it would be unwise to relax any regulations because it produces food for human consumption.

Outside Ontario: 3 respondents
Concern depends on application: e.g., it is

- Less for mine reclamation than agriculture
- Less for golf course than home use.

When practiced in accordance with U.S. Environmental Protection Agency guidelines and regulations, land application presents negligible risk to the user, general public, wildlife or the environment.

- 4) *What information/action do you feel is required to respond to your concerns?*

Ontario: 4 respondents
Scientific studies to quantify concerns.

Develop tests to provide assurance that there are no health risks associated with land application of biosolids.

- 5) *Are you aware of other land utilization programs that you think offer better or safer practices? Please provide details and any documentation.*

Ontario: 4 respondents
Program operated out of Oregon State University held in high regard.

Halton Region program addresses Grand River Conservation Authority monitoring concerns.

Better or safer practices depend on the knowledge, commitment, training and credibility of applicators.

- 6) *Is sewage biosolids utilization on agricultural land a desirable practice?*

Ontario: 4 respondents
Desirable if biosolids meet standards to protect human health and the environment.

3. STAKEHOLDER INFORMATION – cont'd

Biosolids can be beneficial to both the agricultural community and the generator. A long-term objective should be to refine regulations and improve application techniques.

- 7) *Please rank the following biosolids management options in order of preference and explain your ranking - (1) utilization on agricultural land, (2) utilization on other lands with limited public access (or vice versa), (3) utilization on lands with high public access (e.g., backyards, golf courses, parks) (4) landfilling, and (5) incineration.*

Ontario: 4 respondents
3,4 (or 4, 3), 5,2,1

Outside Ontario: 3 respondents
4, 3, 5, 2, 1

- 1 - utilization on agricultural land
- 2 - utilization on other lands with limited public access (or vice versa)
- 3 - utilization on lands with high public access (e.g., backyards, golf courses, parks)
- 4 - landfilling
- 5 - incineration

- 8) *Please indicate any other concerns about present land utilization of sewage biosolids practices in Ontario.*

Ontario: 5 respondents
No specific research issues.

Many regulatory issues, e.g.

- inadequate monitoring and regulation
- move to SAR (Standard Approval Regulation)
- cross contamination of biosolids with toxic waste
- unwieldy and costly handling of public concerns, etc.

- 9) *Do you have any comments or recommendations about the WEAO study?*

Ontario: 5 Respondents

Biosolids utilization on agricultural land represents a tremendous bargain for society. Investment in security and liability issues is needed to establish confidence for wider adoption of this practice.

WEAO study is welcome and timely. Long-term biosolids effects should be studied following multiple applications to land over several years.

Request continuing stakeholder participation in the study.

Survey not prepared with biosolids generators in mind.

3. STAKEHOLDER INFORMATION – cont'd

10) *Are concerns in your jurisdiction different for US EPA Class A and Class B biosolids?*

Outside Ontario: 4 Respondents

Increasing preference for “advanced treatment process (Class A) biosolids” due to pathogen concerns.

11) *What is your opinion of the long-term future for biosolids utilization on agricultural land in your jurisdiction? What are the major potential barriers to long-term success?*

Outside Ontario: 4 Respondents

Anticipate that biosolids utilization on agricultural land will continue to grow in use and acceptance provided:

- Information is made readily available;
- Nuisance problems and regulatory issues are addressed at the local project level; and
- New issues raised by interested parties are thoroughly addressed.

Potential barriers to the long-term success biosolids utilization on agricultural land include:

- Growing public concern about food safety production methods and food safety – e.g., the future of biosolids utilization in the UK can no longer be guaranteed by compliance with all relevant legislation and guidelines.
- Increasing quantities of animal manure – e.g., the lower mainland of British Columbia has an over-supply of animal manure for land application.

12) *Please indicate how the regulations in your jurisdiction may be more or less effective than the Ontario Guidelines*

Outside Ontario: 3 Respondents

In general, Ontario guidelines are more restrictive than the U.S, EPA requirements. Prior to the development of risk-based Part 503 rule, EPA's guidelines (and most state requirements) also were more restrictive.

3. STAKEHOLDER INFORMATION – cont'd

13) *Other comments on the significance of contaminants in biosolids applied to land.*

Outside Ontario: 1 Respondent

Available data and past experience (more than 50 years) indicate that, when practiced in accordance with existing federal guidelines and regulations, land application presents negligible risk to the user, general public, crops, wildlife or the environment. However, we must strive to address areas of limited information and improve understanding of the fate and effect of pollutants.

Although the completed questionnaires offered no clear consensus of opinion, the following contaminants were often cited as concerns:

- Pathogens
- Endocrine disrupter compounds (EDCs)
- Organics (polychlorinated biphenyls – PCBs, dioxins, furans)
- Pharmaceuticals

In addition, a number of respondents expressed concern with the regulation, monitoring, inspection and control of the program for sewage biosolids application to agricultural land. This was related to all aspects of the program, including the quality of the biosolids being produced at the treatment plants, haulage, application, soil impact, subsequent farming practices, etc. These concerns fell outside the mandate of this study but are identified for future action.

4. STAKEHOLDER WORKSHOP

A one-day workshop involving mainly Ontario Stakeholders was held on December 1, 1999. The agenda and attendance list are included in Appendix B. The purpose of the workshop was to review the survey results and invite discussion of land utilization of sewage biosolids concerns. A summary of the workshop proceedings is included in Appendix B.

5. CONTAMINANTS OF CONCERN

Information obtained from the survey and workshop, in combination with input from the Technical Steering Committee and the consultants, was used to identify the following contaminants of concern:

- Heavy Metals including Regulated and Unregulated
- Pathogens
- Trace Organics, including Volatile Organic Contaminants (VOCs), Polychlorinated Biphenyls (PCBs), Polynuclear Aromatic Hydrocarbons (PAHs), Pesticides, etc.
- Linear Alkylbenzene Sulphonate (LAS) Surfactants
- Endocrine Disrupter Compounds (EDCs) including Alkylphenol Surfactants (APs), Estrogenic Hormones
- Dioxins and Furans (PCDD/Fs)
- Pharmaceuticals
- Radionuclides
- Nutrients including Nitrogen and Phosphorus

6. LITERATURE REVIEW - BACKGROUND

Detailed literature reviews of metals, pathogens, and organic contaminants in land-applied sewage biosolids prior to the mid 1990's were used as the starting point for this literature study.

Recent literature was searched electronically as indicated below.

6.1 Electronic Databases

The electronic database searches were conducted based on the following search criteria:

- Start date: January 1, 1995.
- Using the DIALOG databases selected for this topic, a basic search strategy was used with the terms "sludge or biosolid", combined with the terms "municipal wastewater or sewage". This created a file of results with over 80,000 articles. To narrow the search, only the articles that had these terms in either the title or as major descriptors (indexing terms used by DIALOG) were searched. This resulted in over 44,000 hits or articles.
- This basic set was combined with the following search terms, one at a time – contaminant*, pathogen, pharmaceutical, antibiotic, endocrine and disrupt*, EDC, alkyl and phenol and ethoxylate, and surfactant, radionuclides, polymer, and medic*, to obtain the final list of articles.

6.2 Information Sources

The following were searched for information:

a) Electronic Databases

- Agricola
- Commonwealth Agricultural Bureau
- Environmental Sciences & Pollution Management
- Biological Sciences
- BIOSIS Previews (1969 – present)
- NTIS – National Technical Information Service
- EI Compendex
- SciSearch – A Cited Reference Science Database - 1990
- Enviroline
- Pollution Abstracts
- Environmental Bibliography
- EMBASE (1974 – present)
- General Science Abstracts/Full text
- Wilson Applied Science & Technology Abstracts

6. LITERATURE REVIEW – BACKGROUND – cont'd

- Energy Science and Technology
- WasteInfo
- PASCAL
- TOXLINE
- CAB Abstracts
- Federal Research in Progress

b) Canada Centre for Inland Waters Library.

Abstracts from the search were input into an ACCESS™ database for ease of analysis. This database allows searching by author or title (e.g. endocrine disruptor) to find the relevant articles. The database can be easily expanded. Appendix C includes a hardcopy of all abstracts included in the database.

6.3 Literature Review - Sections 7 through 15

Sections 7 through 15 provide a literature review on the fate and significance of various contaminants in sewage biosolids after they are applied to agricultural land.

Based on the findings of this literature review, conclusions were drawn dividing specific contaminants into two groups as follows:

- Group I – Contaminants which have sufficient credible scientific information to assure the public that the current agricultural land application program/guidelines are adequate to protect the well being of soils, crops, animals, human health, ground and surface water qualities.
- Group II – Contaminants which do not have sufficient credible scientific information to assure the public that the current agricultural land application program/guidelines are adequate to protect the well beings of soils, crops, animals, human health, ground and surface water qualities

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND

7.1 Introduction

A long history of experimental effort in North America and several European countries has resulted in an enormous volume of published information on the fate and effects of heavy metals in sewage biosolids applied to land. This information has identified soil, plant and animal processes that affect the potential for heavy metal transfer through food chains and that have become the basis for regulatory controls in many nations. Comprehensive reviews of this information have been conducted at regular intervals and are available in the scientific literature. A further detailed review was, therefore, unnecessary but presentation of important background (i.e., cornerstone) information follows.

7.2 Background Information

7.2.1 Ontario

A Canada-Ontario Agreement established in the early 1970s provided funding for improved sewage treatment in the Lower Great Lakes basin. Although much of the funding was designated for infrastructure improvements, a portion was available for research and development including research on land application of sewage biosolids at the University of Guelph and the Environment Canada, Wastewater Technology Centre in Burlington. These experiments were maintained for more than five years and involved high biosolids application rates and four field crops - corn, wheat, orchard grass and brome grass. Results indicated no evidence for crop yield reductions from cumulative biosolids applications ranging up to 499 tonnes/ha dry weight and there were only minor increases of heavy metal concentrations in the plant materials. However, there was a potential for nitrate-N contamination of groundwater when biosolids application was in excess of that required to supply the nitrogen requirement of the growing crop (Soon et al. 1978a, 1978 b and 1980; Webber et al 1981). These authors concluded that biosolids application to land according to Ontario guidelines (MOEE & OMAFRA 1996) which limit nitrogen and heavy metal loadings to soil, would not reduce the yield or quality of the four test crops nor would it reduce groundwater quality (see discussion of Ontario guidelines presented below).

During the same period, a land application of sewage biosolids discussion paper (Webber 1984) and an Environment Canada "Manual for Land application of Municipal Wastewater and Sludge" (Black et al. 1984) were prepared based on literature review. These documents were prepared for use by the general public and received wide distribution within Canada. They contained information on wastewater treatment, sewage biosolids properties and the effects of land application on agriculture and the environment. They recommended careful control of land application, with limited heavy metal loadings to soil.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

7.2.1.1 Guideline Development

Concurrent with the Canada-Ontario Agreement activities, it was recognized that land application of sewage biosolids in Ontario would have to be regulated to protect the food chain and the environment. A committee including representatives from the Ontario Ministries of Agriculture and Food, Environment, and Health, Agriculture Canada, Guelph University, the Ontario Federation of Agriculture and invited experts was struck to develop appropriate guidelines. Because information about biosolids effects on soil and crop quality was limited, a conservative approach was adopted for developing the guidelines, based on the following premises:

- There should be efficient use of the fertilizer value of sewage biosolids applied to land, and
- Sewage biosolids should cause no net degradation of soil and environmental quality.

The Ontario guidelines for sewage biosolids use in agriculture (MOEE & OMAFRA 1996) were introduced in the mid-1970s and included the following nutrient and heavy metal criteria.

Nutrients

Ontario guidelines recommend applying liquid anaerobically stabilized biosolids at a rate not to exceed 135 kg of plant available nitrogen per ha per five years. This amount of nitrogen is sufficient to grow corn and is used by the crop in the year of application. However, phosphorus is added in excess of the corn crop requirement and a residue remains in the soil for crop use during subsequent years.

Heavy Metals

Ontario guidelines limit the maximum concentrations of eleven heavy metals in sewage biosolids and in agricultural soils (Table 7.1).

Background heavy metal data were available for Ontario soils (Frank et al. 1976 and 1979) and the maximum permissible concentrations in soils receiving biosolids were defined as follows for the eleven heavy metals. Some Ontario feeds are deficient in cobalt (Co), copper (Cu), selenium (Se) and zinc (Zn) and maximum soil concentrations for these metals were set at four times background to allow some buildup in soils. Similarly, maximum soil concentrations of chromium (Cr), mercury (Hg) and lead (Pb) were set at eight, five and four times background, respectively, reflecting low plant uptake of these elements and low Cr concentrations in food and fodder. By contrast, maximum soil concentrations for arsenic (As), nickel (Ni), cadmium (Cd) and

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

molybdenum (Mo) are only double the background because of known toxic effects and the potential for Mo leaching (see Section 7.4 for further discussion of these limits).

No maximum soil concentrations were defined for other elements such as aluminum (Al), boron (B), calcium (Ca), manganese (Mn), potassium (K), sodium (Na) and sulphur (S) in soils and biosolids because they were not expected to be problematic in Ontario soils.

TABLE 7.1
Ontario Guidelines for Heavy Metals in Sewage Biosolids and Agricultural Soils
(after MOEE & OMAFRA 1996)

Heavy Metals	Anaerobic Biosolids	Aerobic, Dewatered and Dried Biosolids	Maximum Permissible Metal Concentrations in Soil	Maximum Permissible Metal Loadings to Soil
	Minimum (Ammonium + Nitrate) Nitrogen to Metal Ratios	mg/kg dry wt.	mg/kg dry wt.	kg/ha
Arsenic	100	170	14	14
Cadmium	500	34	1.6	1.6
Cobalt	50	340	20	30
Chromium	6	2800	120	210
Copper	10	1700	100	150
Mercury	1500	11	0.5	0.8
Molybdenum	180	94	4	4
Nickel	40	420	32	32
Lead	15	1100	60	90
Selenium	500	34	1.6	2.4
Zinc	4	4200	220	330

7.2.1.2 Land Application Experience

Land application has been practiced widely in Ontario with generally satisfactory results. Sewage biosolids is an effective fertilizer and soil conditioning material and survey results (OMAFRA 1995) indicated a statistically significant elevation of Cr, Cu and Hg concentrations in soils receiving up to three applications during a fifteen-year period. However, the resulting concentrations were well below the maximum permissible levels for soil. There was little or no change of other heavy metal concentrations in the soils probably because of small metal loadings and sampling and analytical variability. There was a general increase in plant-available phosphorus concentrations in the biosolids treated soils.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

7.2.2 USA

One of the first comprehensive reviews of the fate and effects of heavy metals in sewage biosolids applied to land resulted from a 1972 Conference on “Recycling Sludges and Effluents on Land” sponsored by the US Department of Agriculture, the US Food and Drug Administration, the US Environmental Protection Agency and the National Association of State Universities and Land Grant Colleges (USDA 1973). Subsequently, a Council for Agricultural Science and Technology task force considered the “Effects of Sewage Sludge on the Cadmium and Zinc Content of Crops” (CAST, 1980) and Workshops on “Utilization of Municipal Wastewater and Sludge on Land” (Page et al. 1983) and on the “Effects of Sewage Sludge Quality and Soil Properties on Plant uptake of Sludge Applied Trace Constituents” (Page et al. 1987) were held in the US in 1983 and 1985, respectively. The proceedings of these activities presented up-to-date reviews of heavy metal and other contaminant information and identified cadmium as the heavy metal of most immediate health concern. They also identified research requirements to develop management practices and regulatory controls to protect soil fertility, the food-chain and the environment from the contaminants in sewage biosolids and effluents.

7.2.2.1 Risk Assessment and EPA Regulation 503

In February 1993, The US Environmental Protection Agency (EPA) promulgated “Standards for the Use or Disposal of Sewage Sludge” (Code of Federal Regulation Title 40, Parts 257, 403 and 503, and hereafter referred to as “Reg. 503” (US EPA 1993)). It was developed using a risk-assessment approach and defines acceptable management practices and specific numerical limits for selected chemical pollutants and pathogens in sewage biosolids for land application. Numerical limits were defined for ten heavy metals as follows; As, Cd, Cr, Cu, Pb, Hg, Mo, Ni, Se and Zn (see Table 7.4 in Section 7.4 for actual limits).

Risk assessment involves a systematic mathematical modeling process(es) for calculating criteria based on biological, physical and chemical characteristics of contaminants. For a given site the heavy metals and pathways of exposure were identified, acceptable daily intakes of the metals for the receptor, usually human, were determined or estimated, single pathway limit values were calculated for all the pathways and metals, and the critical pathway(s) for each metal were selected.

In deriving the numerical limits for land application of sewage biosolids, EPA considered 14 transport pathways (Table 7.2) and, in all cases, selected the most stringent value as the limit for each pollutant. For the ten regulated heavy metals, the most stringent loading rates were derived from pathways involving a child directly ingesting sewage biosolids or from pathways involving effects on crops. Detailed information concerning development and application of the risk-assessment approach are presented in US EPA (1989 & 1992).

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 7.2
Pathways for Risk Assessment of Potential Transfer of Biosolids Applied Trace Contaminants to Humans, Livestock, or the Environment and the Highly Exposed Individual
 (after Chaney et al. 1996) ⁽¹⁾.

	Pathway	Highly Exposed Individual
1	Biosolids - Soil -Plant - Human	General food chain; 2.5% of all food for lifetime; 1000 tonnes/ha.
2	Biosolids - Soil -Plant - Human	Home garden 5 year after last biosolids incorporation; 1000 tonnes/ha; about 50% of garden foods for lifetime.
2D&M ⁽²⁾	Biosolids - Soil -Plant - Human	Home garden with annual biosolids application; 1000 tonnes/ha; 50% of garden foods for lifetime.
3	Biosolids - Soil -Human Child	Residential soil, 5 year after last biosolids incorporation; 1000 tonnes/ha; 200 mg soil/day.
3D&M ⁽²⁾	Biosolids - Human Child	Biosolids product on soil surface; 200 mg biosolids/day.
4	Biosolids -Soil - Plant - Animal ↓ Human	Rural farm families; 40% of meat from livestock produced using crops grown on soils with incorporated biosolids; 1000 tonnes/ha; lifetime exposure.
5-Surface	Biosolids - Animal - Human	Rural farm families; 40% of meat from livestock produced on pastures annually sprayed with biosolids, for lifetime.
5-Mixed	Biosolids - Soil - Animal - Human	Rural farm families; 40% of meat from livestock produced on pastures with 1000 tonnes/ha incorporated biosolids, for lifetime.
6	Biosolids - Soil - Plant - Animal	Livestock fed forages and grains, 100% of which are grown on biosolids amended land; 1000 tonnes/ha.
7-Surface	Biosolids - Animal	Grazing livestock on biosolids sprayed pastures; 1.5% biosolids in diet.
7-Mixed	Biosolids - Soil - Animal	Grazing livestock; 2.5% biosolids-soil mixture in diet; 1000 tonnes/ha.
8	Biosolids - Soil - Plant	"Crops"; vegetables in strongly acidic biosolids amended soil; 1000 tonnes/ha.
9	Biosolids - Soil - Soil Biota	Earthworms, slugs, bacteria, fungi in biosolids amended soil; 1000 tonnes/ha.
10	Biosolids - Soil - Soil Biota - Predator	Birds; 33% of bird diet earthworms affected by biosolids; 1000 tonnes/ha.
11	Biosolids - Soil - Airborne Dust - Human	Tractor operator; 1000 tonnes/ha.
12	Biosolids - Soil - Surface Water - Human	Water quality criteria; fish bioaccumulation, lifetime
13	Biosolids - Soil - Air - Human	Farm households.
14	Biosolids - Soil - Groundwater - Human	Farm wells supply 100% of water used for lifetime.

⁽¹⁾ Although a very high cumulative biosolids loading (1000 tonnes/ha) is assumed, the annual application is limited by the nitrogen requirement of the crop grown; it may take up to 100 years or more of annual applications to reach this cumulative loading.

⁽²⁾ Distribution and Marketing (D&M) biosolids are assumed to remain on the soil surface.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Reg. 503 (US EPA 1993) has many detractors who feel that it does not adequately protect human and animal health and the environment over the long term. The Cornell University, Waste Management Institute has been a focal point for criticism within the USA (Harrison et al. 1999), but criticism also has arisen from many European countries where guidelines/regulations are generally much more conservative. Examples of the Cornell University, Waste Management Institute criticisms included:

- Limits on metal additions in biosolids suffer from a lack of high quality data, particularly from long term experiments.
- Short term (1-10 years) effects may not be problematic but long term effects resulting from organic matter degradation following maximum heavy metal loading may greatly increase metal bioavailability and seriously reduce agricultural soil productivity (see the "time-bomb effect" below).
- Potential symbiotic effects of simultaneous loading with several heavy metals at EPA limits are untested.
- Organic chemicals no longer produced in the USA should not have been excluded from regulation because they are still found in biosolids.
- Alkylphenol ethoxylates should not have been excluded from regulation because they occur in biosolids in parts per hundred concentrations and one of the degradation products, 4-nonylphenol, is an endocrine disrupter.
- EPA should re-evaluate the safety for grazing animals of the 30-day waiting period following Class B biosolids application to pastures.
- Risk to soil productivity by decreasing valuable soil organisms is inadequately assessed.
- There is little assurance that home gardeners or landscapers will either be aware or able to follow requirements for safe use of biosolids.

The EPA and Water Environment Federation have responded by pointing out that Reg. 503 is based on a very large body of scientific information and is designed to protect the "highly exposed individuals" (human, animal, crop, etc.). Moreover, the Reg. 503 was peer reviewed and received approval from a committee of agricultural and environmental scientists with expertise in all aspects of biosolids use. Land application of biosolids continues in much of the USA according to Reg. 503 but research also is continued in an effort to obtain ever more reliable information and insure the safety of this practice. Some of the recently completed and on-going research studies regarding biosolids processing, management and the impacts of land application on human and the environment, funded by the Water Environment Research Foundation (WERF) are summarized in Appendix D. A brief statement of research objectives and status (as of 1999) are also included.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

7.2.2.2 Land Application Experience

Recently, the Water Environment Research Foundation published a review of long term experience of biosolids land application at ten sites in the US (WERF 1993). In general, the programs reported few technical problems which spoke well for their overall management and success. However, some complaints about heavy truck traffic and road damage, odours and soil compaction were reported.

Similarly, "Use of Reclaimed Water and Sludge in Food Crop Production" was reviewed by a National Research Council Committee (NRC, 1996). The review was comprehensive and concluded that "While no disposal or reuse option can guarantee complete safety, the use of these materials (i.e., treated municipal wastewater and sewage sludge) in the production of crops for human consumption, when practiced in accordance with existing federal guidelines and regulations, presents negligible risk to the consumer, to crop production, and to the environment. Current technology to remove pollutants from wastewater, coupled with existing regulations and guidelines governing the use of reclaimed sludge and wastewater in crop production, are adequate to protect human health and the environment".

7.2.3 European Community

Several European countries have long histories of recycling sewage effluent and biosolids to agricultural land. Originally, regulation of this practice was a local or national concern because the produce was consumed locally. However, formation of the European Community (EC) resulted in greatly increased exchange of produce and recognition of the need for reconciling land application Guidelines/regulations among member countries to ensure consistent product quality.

A European Community (EC) "land application of sewage sludge program" was initiated in the late 1970s and continued for approximately ten years. The major objective of the program was development of a Directive, promulgated in 1986, "on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture" (EC Council Directive 1986). In essence, the Directive was based on the "no net degradation of soil" approach and defined heavy metal guidelines for agricultural use of sewage biosolids which applied to all EC member countries (see Tables 7.4 and 7.5). The Directive is presently under revision and more restrictive heavy metal guidelines are being proposed (Draft EC Council Directive 2000).

An important aspect of the EC program was that it brought together wastewater treatment and agricultural experts from all EC and several invited countries. A large number of conferences and workshops were held on all aspects of sewage biosolids treatment and use and proceedings were published.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

In addition to the numerous EC publications mentioned above, Smith (1996) prepared an exhaustive review and assessment of biosolids recycling with special reference to the UK and "A Global Atlas of Wastewater Sludge and Biosolids Use and Disposal" has been prepared (IAWQ 1996). The atlas is the most complete collection available of world information on generation, treatment and management/disposal - including land application guidelines - of sewage biosolids.

7.3 Recent Information

A comprehensive review of the fate and effects of heavy metals and some inorganics in sewage biosolids applied to land was prepared recently by the University of Washington for the Northwest Biosolids Management Association (Harrison et al. 1997). Information for the 10 heavy metals currently regulated under the Part 503 (US EPA 1993) and 12 other metals/inorganics (aluminum, antimony, asbestos, barium, beryllium, boron, cyanide, fluoride, manganese, silver, thallium and tin) were included in the review. Moreover, the effects of biosolids application on plant yield, metal concentrations in soil, metal uptake by plants, and metal movement in soil were considered. It was concluded that biosolids is a safe soil amendment if proper management practices are observed during land application. This conclusion was based on the following information:

- Much research has been conducted with biosolids containing high levels of heavy metals, or with high biosolids application rates, or both. Even under such conditions, detrimental effects were observed only in peculiar situations;
- Metals generally do not move down through the soil profile, but persist in the zone of incorporation and build up with repeated applications;
- The availability of metals to plants generally decreases with time due to the formation of organic or oxide complexes.

Despite relatively little research, the 12 other metals/inorganics not regulated under Part 503 were not expected to pose significant health or environmental risks for biosolids utilization. Limited information indicates that:

- Their concentrations in biosolids are low compared to the 10 regulated metals and to their concentrations in the earth's crust.
- Biosolids reduce their bioavailabilities.

The degree to which biosolids adversely affect surface and groundwater quality is dependent upon a wide range of factors including climate, topography, land use, soil characteristics, and the chemical composition and application rate of biosolids. All of these factors are considered in watershed studies.

The Water Environment Research Foundation (WERF) sponsored a project (Draeger et al. 1999) to:

- Collect, review and analyze information on which to base sound biosolids management,
- Identify information gaps;
- Highlight research needs.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

A long term watershed study using biosolids for crop production in Rosemount, Minnesota provided considerable information needed for the Draeger et al. (1999) project. This study was initiated in 1973 to evaluate liquid biosolids application on: surface and groundwater quality and nutrient concentration/transport; heavy metal buildup, mobility, and phytoavailability in soil; crop quality and yields; and nutrient buildup in soil.

A 16 ha site, with slopes ranging from 1% to 12% was monitored. Parallel, graded terraces were constructed to control the surface movement of soil and water. Municipal sewage biosolids were applied at an average rate of 473 kg N/ha per year for 20 years (1973 to 1993) for corn production and at an average rate of 836 kg N/ha per year for 12 years for grass. Two areas within the research site received only commercial fertilizers to serve as control areas.

Monitoring results showed that:

- While slightly higher concentrations of Cu, Zn and B were measured in the leaf of the corn grown on biosolids amended than on control soil, and slightly higher concentrations of Cr, Cu and Zn were measured in the reed canary grass grown on biosolids amended than on control soil; all concentrations of these and other metals (Ni, Pb, Mn and Cd) in the plant materials were below toxic levels.
- After 20 years of high loading rates, Cr and Cu moved to a depth of 0.45 m and 0.6 m, respectively, in the soil; the other metals moved to lesser depths.
- Biosolids application did not increase Cd, Ni, Pb and Cr concentrations in snowmelt runoff. However, surface application of biosolids in winter increased the runoff of almost all of the metals (as well as P, N and K).

The study concluded that with Best Management Practices (BMPs) (ie. use of agronomic rates, correct timing of biosolids application and appropriate soil erosion control) biosolids application benefits crop yields and soil fertility, and has no negative effect on surface and ground water qualities.

7.4 Heavy Metals That Are Regulated In Ontario

Eleven heavy metals (Pb, Cd, Cr, Cu, Ni, Hg, Zn, As, Co, Mo, and Se) in sewage biosolids and soils are regulated by the Ontario guidelines (MOEE & OMAFRA 1996). These constituents received priority treatment because they were known to occur at significant concentrations in sewage biosolids and there was concern about detrimental effects on crop production, human and animal health, and environmental quality. Several (e.g., Cu, Zn, Co, Mo, Se) are micronutrients that are required by plants and animals but others (e.g., Pb, Cd, Hg, As) serve no biological purpose. With the exception of Co, these constituents were considered high priority by the US EPA and were evaluated in their risk assessment.

7.4.1 Biosolids

Recent surveys of Canadian (Webber and Nichols 1995) and (Webber and Bedford 1996) and US biosolids (Lue-Hing et al. 1999) indicate that:

- The considerable range of heavy metal concentrations occurring in sewage biosolids, and
- Significant reductions in concentration with time (Table 7.3) resulting from sewer use by-law implementation. A recent report (Sajjad et al. 1999) indicates that this trend of decreasing metals concentrations in biosolids continued in the USA.

**7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS
IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd**

TABLE 7.3
Mean Heavy Metal Concentrations (mg/kg dry wt.) in US and Canadian Sewage Biosolids
(after Webber and Nichols 1995 and Lue-Hing et. Al. 1999)

Metal	Canadian Surveys		US Surveys		
	1981	1995	1979	1988	1996
Arsenic		2.3	6.7	9.9	11.5
Cadmium	35	6.3	69	6.9	6.4
Chromium	1040	319	429	119	103
Copper	870	638	602	741	506
Lead	545	124	369	134	111
Mercury		3.5	2.8	5.2	2.1
Molybdenum		22	17.7	9.2	15
Nickel	160	38	135	43	57
Selenium		3.3	7.3	5.2	5.7
Zinc	1390	823	1594	1202	830

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Whereas most jurisdictions define six or seven heavy metal limits for land applied biosolids, Ontario defines eleven (Table 7.4). Even though the Ontario concentration limits are higher than for most European countries and the proposed European Union (EU) limits, Tables 7.5 and 7.6 indicate that Ontario soil concentration limits and maximum annual loading limits are similar to those for most European countries and to the proposed EU limits, given comparable soil pH conditions. Biosolids generators in Ontario are required to comply with all three conditions:

- (1) Maximum metal concentration limits in biosolids.
- (2) Annual plant available nitrogen loading limits for anaerobically stabilized biosolids or annual solids loading limits for aerobically stabilized, dewatered and dried biosolids; and
- (3) Maximum soil concentration limits.

Conditions (1) and (2) together effectively limit the annual metal loadings, and conditions (1), (2) and (3) together limit the life-time cumulative biosolids and metals loadings that can be applied to agricultural land.

No rationale was found for the more stringent soil concentration limits adopted by some European countries and the EU.

**7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS
IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd**

TABLE 7.4
Maximum Permissible Heavy Metal Concentrations in Sewage Biosolids (mg/kg dry wt.)
For Selected Countries and Ontario Where Land Application is Practiced
(after Matthews and Lindner 1996)

Country	Pb	Cd	Cr	Cu	Ni	Hg	Zn	As	Co	F	Mo	Se
Austria	400	5	400	400	80	7	1600					
Belgium	600	12	500	750	100	10	2500					
	500	10	500	600	100	10	2500					
Denmark ⁽¹⁾	120	0.8	100	1000	30	0.8	4000					
EU (present) ⁽²⁾	750- 1200	20-40		1000- 1750	300- 400	16-25	2500- 4000					
EU (proposed) ⁽³⁾	750	10		1000	300	10	2500					
	500	5		800	200	5	2000					
	200	2		600	100	2	1500					
Finland	150	3	300	500	30	2	500					
France ⁽¹⁾	800	20	1000	1000	200	10	3000					100
	1600	40	2000	2000	400	20	6000					200
Germany	900	5	900	800	200	8	2000					
	900	10	900	800	200	8	2500					

**7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS
IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd**

TABLE 7.4 - cont'd
**Maximum Permissible Heavy Metal Concentrations in Sewage Biosolids (mg/kg dry wt.)
For Selected Countries and Ontario Where Land Application is Practiced**
(after Matthews and Lindner 1996)

Country	Pb	Cd	Cr	Cu	Ni	Hg	Zn	As	Co	F	Mo	Se
Ireland	750	20		1000	300	16	2500					
Italy	750	20		1000	300	10	2500					
Netherlands	100	1.25	75	75	30	4.75	300					
Norway	100	4	125	1000	80	5	1500					
Spain	750	20	1000	1000	300	16	2500					
	pH < 7											
	pH > 7											
Sweden	100	2	100	600	50	2.5	800					
Switzerland	500	5	500	600	80	5	2000					
UK	1000									1200		
US EPA ⁽⁴⁾	300	39		1500	420	17	2800	41				100
	Exceptional Quality											
	Ceiling Concentrations	85		4300	420	57	7500	75			75	100
Ontario	Present	34	2800	1700	420	11	4200	170	340		94	34
	Long Term	4	530	380	80	1.4	840	35	77		1.2	6

(1) Recently revised, with lower limits for heavy metals and limits for a number of organic contaminants

(2) EC Council Directive (1986)

(3) Draft EC Council Directive (2000)

(4) Biosolids meeting the Exceptional Quality limits for all 10 metals can be applied without any life-time cumulative loading restriction. Biosolids exceeding the Exceptional Quality limits for any one of the 10 metals but meeting the Ceiling Concentration Limits for the metals can be land applied but have to meet the life-time cumulative loading limits (see Table 7.6). Biosolids exceeding the Ceiling Concentration limits for any one of the 10 metals cannot be land applied.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

7.4.2 Soils

7.4.2.1 Heavy Metal Concentrations

Maximum permissible heavy metal concentrations for Ontario and European soils receiving sewage biosolids were developed (Matthews and Lindner, 1996) on the basis that there should be "no net degradation of soil" for agricultural production (Table 7.5). UK permissible concentrations are based on extensive research indicating the safety of generally larger values than are defined for other European countries. Moreover, metal bioavailability and degradation potential are greater in acid than in neutral soils. Several jurisdictions, including the UK, account for this effect by reducing permissible metal concentrations as soil pH decreases. Ontario requires that soils be limed to pH 6 prior to biosolids application. Considering the number of independent jurisdictions represented, the data in Table 7.5 exhibit remarkable agreement.

7.4.2.2 Heavy Metal Loadings

A comparison of annual and life-time cumulative heavy metal loading limits to soils for selected jurisdictions where biosolids are used in agriculture is presented in Table 7.6. Ontario annual loadings were calculated as one-fifth of the product of the metal concentration limits (mg/kg/1000 dry wt. = kg/tonne dry wt.) for sewage biosolids (from Table 7.4) multiplied by the maximum solids loading rate of 8 tonnes/ha per 5 years (e.g., the annual Zn loading is: 4.2kg/tonne x 8/5 tonnes/ha/year = 6.7 kg/ha).

Table 7.6 shows that permissible annual heavy metal loading rates to soil in Ontario are similar to those for European countries and the proposed EU medium term loading rate. With few exceptions, the differences between Ontario, European and EU annual loading rates are within a factor of two. Ontario annual loading rates are generally one-tenth of those permissible in the UK and USA. The soil limit essentially put a cumulative life-time loading limits, same as in Ontario. Actual life-time loading limit depends on the background concentration of metals in the particular field. The life-time cumulative loading limits for Ontario are from 1.3 to 50 times less than those for the USA. There are no life-time cumulative heavy metal loading limits for exceptional quality USA biosolids.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 7.5
Maximum Permissible Heavy Metal Concentrations in Soil (mg/kg dry wt.) for Selected Countries and Ontario Where Biosolids are Used in Agriculture¹
 (after Matthews and Lindner 1996)

Country		Pb	Cd	Cr	Cu	Ni	Hg	Zn	As
Austria	Upper	100	11	100	100	60	1	300	
Belgium	Flanders sand	50	1	100	50	30	1	150	
	Flanders silt	300	3	150	140	75	1.5	300	
Denmark		40	0.5	30	40	15	0.5	100	
EU (present) ⁽²⁾	pH 6-7	50-300	1-3		50-140	30-75	1-1.5	150-300	
EU (proposed) ⁽³⁾	pH 5-6		0.5		20	15	0.1	60	
	pH 6-7		1		50	50	0.5	150	
	pH >7		1.5		100	70	1	200	
Finland		60	0.5	200	100	60	0.2	150	
France	pH >6	100	2	150	100	52	1	300	
Germany	pH 5-6	100	1	100	60	50	11	150	
	pH >6	100	1.5	100	60	50	11	200	
Ireland		50	1		50	30	1	150	
Italy		100	1.5		100	75	1	300	
Netherlands		85	0.6	100	36	35	0.3	140	29
Norway		50	1	100	50	30	1	150	
Spain	pH <7	50	1	100	50	30	1	150	
	pH >7	300	3	150	210	112	1	450	
Sweden		40	0.4	30	40	30	0.3	75	
Switzerland		50	0.8	75	50	50	0.8	200	
UK	pH 5-5.5	300	3	400	80	50	1	200	50
	pH 5.5-6	300	3	400	100	60	1	200	50
	pH 6-7	300	3	400	135	750	1	200	50
	pH >7	300	3	400	200	110	1	300	50
Ontario, Canada	pH \leq 6	60	1.6	120	100	32	0.5	220	14

1. The USA has not defined maximum permissible heavy metal concentrations in soil

2. EC Council Directive (1986)

3. Draft EC Council Directive (2000)

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 7.6
**Maximum Annual and Life-Time Cumulative Heavy Metal Loadings (kg/ha) to Soil for
Selected Countries and Ontario Where Biosolids are Used in Agriculture**
(after Matthews and Lindner 1996 and MOEE & OMAFRA 1996)

Country		Pb	Cd	Cr	Cu	Ni	Hg	Zn	As	F
Austria (stelemark)	Annual ⁽³⁾	1.25	0.025	1.25	1.25	0.25	0.025	5	0.05	
EU (present) ⁽¹⁾	Annual ⁽⁴⁾	15	0.15		12	3	0.1	30		
EU (proposed) ⁽²⁾	Present (Annual) ⁽⁴⁾	2.25	0.03		3	0.9	0.03	7.5		
	Medium term (Annual)	1.5	0.015		2.4	0.6	0.015	6		
	Long term (Annual)	0.60	0.006		1.8	0.03	0.006	4.5		
France	Annual	2.4	0.06	3	3	0.6	0.03	9		
Germany	Annual	1.5	0.016	1.5	1.3	0.3	0.013	2.5		
	Life-time Cumulative	90	1.6	210	150	32	0.8	330	14	
UK	Annual	15	0.15	15	7.5	3	0.1	15	0.7	20
USA	Annual ⁽⁵⁾	15	1.9		75	21	0.85	140	2	
	Life-time Cumulative ⁽⁶⁾	300	39		1500	420	17	2800	41	
Ontario, Canada	Annual ⁽⁷⁾	1.8	0.05	4.7	2.7	0.7	0.02	6.7	0.28	

1. EC Council Directive (1986)
2. Draft EC Council Directive (2000)
3. Loadings are for arable land; pasture to receive half-loads
4. Loadings are mean values over a ten-year period
5. Loading limits apply only to biosolids that are sold or given away in a bag or other container
6. Loading limits apply to biosolids with metals exceeding the Exceptional Quality concentration limits (Table 7.4)
7. Values are one-fifth of the five-year loading

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

7.5 Heavy Metals That Are Not Regulated in Ontario

The US EPA is currently assessing whether limits are needed for 14 metals/inorganics (Be, B, F, Al, Ti, V, Mn, Ag, Sn, Sb, Ba, Tl, asbestos and CN) under its Round II Risk assessment. In general, these constituents in sewage biosolids have received little research attention and information about them is sparse. Harrison, et. al. (1997) provided a comprehensive review and summary of them in terms of known impacts to humans, wildlife and domestic animals, phytotoxicity, and migration in soils.

Some of the key summary information is reproduced in Table 7.7. Based on limited information, it was concluded that "It is not likely, however, that any of the additional trace elements and compounds pose significant risk when compared to those extensively studied because of their low concentration in biosolids compared to other metals or to the earth surface and because of the characteristics of biosolids that reduce the availability of many of these elements and compounds" (Harrison et. al. 1997).

7.5.1 Concentrations in Biosolids and Soils

There has not been a concerted effort to obtain data for unregulated heavy metals in biosolids, however, limited Canadian information (Table 7.8) was collected during the survey conducted by Webber and Nichols (1995) and Webber and Bedford (1996) and considerable information (Table 7.9) was collected during a 1982/83 survey of UK biosolids (Smith 1996).

Canadian and UK data for some unregulated heavy metals in soil are presented in Table 7.9. Where comparison is possible, these soil values frequently are similar to biosolids median values.

The concentration ranges for unregulated heavy metals were generally wider in UK than in Canadian biosolids but, with some exceptions (notably Ti), median values show reasonable agreement. Increasing sewer use by-law controls in the ten-year interval between the UK and Canadian surveys probably has contributed to the narrower ranges of Canadian values.

**7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS
IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd**

TABLE 7.7
Summary of Known Impacts of Some Trace Elements and Compounds in Biosolids
(after Harrison et. al. 1997)

Trace Element or Compound	Known to Cause Impacts to Humans	Known to Cause Impacts to Wildlife and Domestic Animals	Known to Cause Phototoxicity	Known to Migrate in Some Soils
Aluminum (Al)	Possible	Not examined	Yes	Yes
Antimony (Sb)	Yes, but not severe	Not examined	Not examined	No
Asbestos	Yes	Possible	No	No
Barium (Ba)	Yes	Yes	Not examined	Not examined
Beryllium (Be)	Yes	Yes	Yes, if in a soluble form	Not examined
Boron (B)	Not examined	Yes, in excess	Yes	Yes
Cyanide (CN)	Yes, in excess	Yes, in excess	Yes	Yes
Fluoride (F)	Yes, in excess	Yes, in excess	No	Yes
Manganese (Mn)	Yes	Not examined	Yes	No
Silver (Ag)	Yes, but not severe	Not examined	Not examined	Not examined
Thallium (Tl)	Yes	Not examined	Not examined	Not examined
Tin (Sn)	Not examined	Not examined	Not examined	Not examined
Titanium (Ti)	Not examined	Not examined	Not examined	No
Vanadium (V)	Not examined	Yes, in invertebrates	Not examined	Not examined

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 7.8
Concentrations (mg/kg/ dry/wt.) of Unregulated Heavy Metals in Canadian Sewage Biosolids
 (after Webber and Nichols 1995; Webber and Bedford 1996)

Treatment Plant Location	Sb	Ba	Bi	B	Ag	Sr	Tl	Sn	Ti	V	Zr
Parkville, BC ⁽¹⁾	38	347	14	38	16	96	25	14	168	11	8
Burnaby, BC ⁽¹⁾	39	354	11	54	67	106	11	28	214	10	30
Edmonton, AB ⁽¹⁾	72	578	Nd	49	41	281	21	7	138	19	7
Calgary, AB ⁽¹⁾	86	663	8	41	69	219	7	62	107	12	7
Lethbridge, AB ⁽¹⁾	117	477	Nd	107	35	189	6	9	148	19	3
Regina, SK ⁽¹⁾	71	413	12	70	58	169	68	36	197	54	38
Winnipeg, MB ⁽¹⁾	140	403	12	91	28	121	31	26	186	23	4
Windsor, ON ⁽¹⁾	67	314	9	210	58	266	Nd	27	140	9	34
Guelph, ON ⁽¹⁾	62	556	9	58	38	584	23	39	165	8	19
Halton, ON ⁽¹⁾	64	563	14	97	5	452	86	26	118	10	8
Ottawa, ON ⁽¹⁾	80	742	14	134	8	504	131	46	242	16	7
Toronto – Main TP, ON ⁽²⁾	63	677	7	140	81	221	31	88	145	23	15
Toronto – Humber TP, ON ⁽²⁾	53	688	5	65	61	159	Nd	100	119	14	20
Toronto – Highland Creek, ON ⁽²⁾	45	300	5	145	44	126	3	394	61	21	7
Fredericton, NB ⁽²⁾	24	568	7	20	45	209	Nd	44	85	8	16
Moncton, NB ⁽²⁾	37	379	2	29	41	91	Nd	30	244	27	12

Sb = Antimony; Ba = Barium; Bi = Bismuth; B = Boron; Ag = Silver; Sr = Strontium; Tl = Thallium; Sn = Tin; Ti = Titanium; V = Vanadium; Zr = Zirconium
 Beryllium was not detected in any of the biosolids at 1 mg/kg dry wt.

(1) Values are means for two samples taken two months apart

(2) Values are means of four analyses; two replicates each for samples taken two months apart

Nd: Not detected

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 7.9

Concentrations (mg/kg dry/wt.) of Unregulated Heavy Metals in Sewage Biosolids, Soils and Earth's Crust
(after Webber and Nichols 1995; Webber and Bedford 1996)

Metal	Canada			USA			United Kingdom			
	Biosolids	Soil	Earth's Crust	Biosolids	Soil	Earth's Crust	Biosolids	Soil	Earth's Crust	
	Range	Median	Ontario Background	Typical			Range	Median	Range	Common Value
Aluminum (Al)						88000				
Barium (Ba)	300-688	517	190	9-1004		500	23-3104	363		
Cyanide (CN)			0.12 (free)	~800						
Fluorine (F)				~100		600-900	60-40000	250 ⁽²⁾	30-300	150
Beryllium (Be)			1.2	<1.5	0.37	5	1-30		0.1-40	3
Boron (B)	20-134	68		16-680		10	15-1000	30 ⁽²⁾	2-100	10
Titanium (Ti)	61-244	147				6300	355-1677	1795		
Vanadium (V)	8-54	15	91	20-400		100	7-660	26		100
Manganese (Mn)				60-3900	57-1460	850	55-13902	318		
Iron (Fe)							2480-106812	12479		
Cobalt (Co)			19				<2-617	8	1-40	10
Gallium (Ga)							<2-15	3		
Germanium (Ge)							<2-9	<2		
Bromine (Br)							4-1049	29		
Rubidium (Rb)							<2-232	16		

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 7.9 – cont'd
Concentrations (mg/kg dry/wt.) of Unregulated Heavy Metals in Sewage Biosolids, Soils and Earth's Crust

Metal	Canada			USA			United Kingdom			
	Biosolids		Soil ⁽¹⁾	Biosolids	Soil	Earth's Crust	Biosolids		Soil	
	Range	Median	Ontario Background	Typical			Range	Median	Range	Common Value
Strontium (Sr)	91-584	199					45-1335	174		
Yttrium (Y)							<2-34	7		
Zirconium (Zr)	3-38	10					14-2500	70		
Niobium (Nb)							<2-41	5		
Silver (Ag)	5-81	43	0.35	<930		100	<2-1252	5	1-3	1
Tin (Sn)	7-394	33		40-700		2	19-683	101	2-200	10
Antimony (Sb)	24-117	64	1	44 & 308		0.2	<2-572	7	2-10	
Tellurium (Te)							<2	<2		
Tungsten (W)							<2-1418	4		1
Thallium (Tl)	Nd-131	16	2.5			7000	<2-5	<2		0.1
Bismuth (Bi)	Nd-14	9					<2-10	8		
Uranium (U)							<2-2	2		

Biosolids data obtained from Webber and Nichols (1995). Webber and Bedford (1996), Smith (1996) and Harrison et. al (1997) .
 Soil and earth crust data obtained from MOEE (1997, Smith (1996) and Harrison et al. (1997)).
 Nd – Not detected

(1) The Ontario Background soil concentrations are taken from Table F of the Ontario Guideline for Use at Contaminated Sites (MOEE 1997) and represent the 98 percentile values of the Ontario-wide sampling program at rural and urban parks unaffected by local point sources of pollution.

(2) Common Value

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

7.5.2 Potential Implications

Harrison et al. (1997) presented information for the unregulated metals and compounds included in Table 7.7. and summaries of that information are provided below.

Aluminum (Al)

Soils minerals, particularly aluminosilicate clays, contain large amounts of Al (~10%) which is released and reprecipitated as amorphous hydroxide during weathering. Soluble Al is phytotoxic but the concentration in soil solution is highly dependent upon pH and is virtually zero above pH 5. Well managed agricultural soils are limed to maintain pH>5 and in Ontario biosolids may not be applied to soil with pH<6. Thus, Al in biosolids is considered a non-issue relative to agricultural use of biosolids.

Antimony (Sb)

Concentrations in biosolids exceed those in soils (Table 7.9).

Asbestos

Asbestos is a common name for fibrous silicate minerals that are resistant to acid and fire. It is a carcinogen that can cause lung cancer in humans, primarily through inhalation. Dermal contact is not an exposure route of concern because it is not likely to be absorbed through the skin. Asbestos measurements were not identified for Ontario or other Canadian biosolids and are not recommended by the consultants preparing this report. They consider it highly unlikely that asbestos fibres embedded in biosolids residues would become airborne and represent a significant carcinogenic hazard to humans or animals.

Barium (Ba)

Ba concentrations in Canadian biosolids (Table 7.8) fall in about the middle of the 9-1004 mg/kg typical concentration range reported by Harrison et al. (1997) and approximate the 500 mg/kg concentration in the earth's crust. Since soil is derived from the earth's crust, land application of biosolids is unlikely to significantly increase Ba concentrations in soil.

Beryllium (Be)

UK data (Table 7.8) indicate very low Be concentrations in biosolids which are unlikely to significantly increase concentrations in soil.

Boron (B)

The Ontario guidelines (MOEE & OMAFRA 1996) define a 1 kg/ha/year limit for B application to land growing intolerant crops and the hot water soluble B concentration in soil for these crops should be ≤ 1 mg/kg dry wt. The maximum application (8 dry tonnes/ha/5 yr.) of sewage biosolids containing the Canadian maximum B concentration (134 mg/kg dry wt. - Table 7.9) would add 1.1 kg/ha of B to soil during the year of application. This loading approximates the annual limit for intolerant crops and, depending on the soil concentration prior to land application, might cause phytotoxicity.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Cyanide (CN)

Cyanide salts are used in extraction of gold and silver from ores, in electroplating and for cleaning certain metals. No total CN data were identified for Canadian biosolids but mean concentrations in USA biosolids are about 800 mg/kg dry wt. The free form [HCN (aq)+CN⁻], is considered to be extremely toxic to humans and animals, and is unlikely to occur in biosolids because they are not acid. No free cyanide was detected in biosolids collected from 37 wastewater treatment plants in Ontario (MOE 1988). Iron cyanide complexes [Fe (CN)₆³⁻ + Fe(CN)₆⁴⁻], is the most likely forms in Ontario biosolids because of iron chloride addition for phosphorus removal, and are considered to be relatively harmless. Moreover, cyanides don't accumulate in soil because they undergo rapid microbial degradation. Thus, it is unlikely that CN in land applied biosolids represents a significant risk to human or animal health because of low concentrations in the biosolids and low persistence in soils.

Fluoride (F⁻)

Although F is added to public water supplies at 1 mg/L to reduce tooth decay, no F data were identified for Canadian biosolids. UK data (Table 7.9) indicate that biosolids concentrations may greatly exceed soil concentrations. Excessive F concentrations in plant tissues can cause toxicity in grazing animals. Documentation of F concentrations in Ontario biosolids is warranted.

Manganese (Mn)

Mn concentrations in USA biosolids (60-3900 mg/kg dry wt) were similar to those for western Washington soils (57-1460 mg/kg). Since biosolids are applied to Ontario soils with pH > 6 it is very unlikely that they would be associated with Mn toxicity to plants that occurs in acid soils.

Silver (Ag)

Photographic processing works contribute Ag to biosolids, however, it is a valuable metal and concentrations in Ontario biosolids were uniformly low (Table 7.8). Ag does not cause serious toxic manifestations in humans or other animals.

Thallium (Tl)

Tl concentrations in Canadian biosolids were uniformly low (Table 7.8) and are unlikely to present a significant hazard due to land application. However, Tl is exceptionally toxic to humans and animals and further documentation of concentrations in biosolids is warranted.

Tin (Sn)

Sn is used as a coating on other metals to prevent corrosion and in alloys. Sn compounds are used as mordants in dyeing and as reducing agents, catalysts and fungicides. With one exception, Canadian biosolids exhibited uniformly low Sn concentrations (Table 7.8) but concentrations in excess of 500 mg/kg dry wt. can occur (Table 7.9). Research is lacking on the fate and effects of Sn in land applied biosolids.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Titanium (Ti)

Ti is a strong, low density, corrosion resistant metal used on its own and in alloys. The dioxide is used as a gemstone and paint pigment. Concentrations in Canadian biosolids were uniformly low but high concentrations can occur (Table 7.9). Findings from one research study suggest that Ti associates strongly with both organic and inorganic materials and is likely to be bound by biosolids and soil constituents.

Vanadium (V)

Vanadium concentrations in all Canadian and most UK biosolids were less than typical soil concentrations (Table 7.9).

Other Metals (Table 7.9)

Iron (Fe), like Al, is a major constituent of soil minerals and is released and re-precipitated as amorphous hydroxide during weathering. Fe is very slightly soluble within the pH range of agricultural soils and even though high concentrations may occur in biosolids (Table 7.9), frequently due to iron chloride addition for phosphorus removal or biosolids conditioning, it is considered a non-issue in relation to agricultural use of biosolids.

Several other heavy metals included in Table 7.9 (e.g., Br, Sr, Zr) can occur at elevated concentrations in biosolids but no information was identified concerning their fate and effects following land application.

7.5.3 Loadings to Ontario Land

The Ontario Guideline for Use at Contaminated Sites (MOEE, 1997) provides soil concentration limits for an extensive list of metals, inorganics and organics for property owners to restore contaminated sites to allow for agricultural, parkland, residential, commercial and industrial uses. Two sets of soil concentration limits are defined for agricultural use. The first set known as the "Background approach" limits involves use of soil quality criteria to define ambient or naturally occurring uncontaminated soil conditions. These limits were developed by determining the 98 percentile concentrations from an Ontario-wide sampling program at rural and urban parks that are unaffected by local point sources of pollution. The second set known as "Effects Based approach" limits involves use of soil and groundwater quality criteria developed to provide protection against the potential for adverse effects to human health, ecological health and the natural environment. Due to complexity in estimating the movement of metals through soil, a potential impact of metals on ground water quality was not considered for the "Effects Based Approach" limits for metals. The "Effects Based Approach" and "Background Approach" limits for selected unregulated heavy metals are presented in Rows 1 and 2, respectively, of Table 7.10.

The maximum number of biosolids applications that can be allowed to a given field before the agricultural land use criteria recommended for contaminated site clean up is reached were calculated and presented in Rows 10 and 11 of Table 7.10.

* Canadian Biosolids Survey, Webber & Nicols, 1995, Webber & Bedford, 1996

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

“Effects Based Approach” limits – Row 11, which assumes biosolids contain the highest metals concentrations reported by Canadian Biosolids Survey*, indicates that none of the unregulated metals are of concern in relation to the “Effects Based Approach” limits. Under this scenario, beryllium (Be) is the most restrictive, and it limits the maximum number of biosolids applications to a given field to 6. However, molybdenum (a regulated metal) may be more restrictive, limiting the number of biosolids applications to field to 5 (MOEE and OMAFRA 1996).

“Background Approach” limits – Row 11, which assumes biosolids contain the highest metals concentrations reported by the Canadian Biosolids Survey*, indicates that several unregulated metals are of concern in relation to the “Background Approach” limits. Based on the small amount of information available, silver (Ag) is the most restrictive and it limits the maximum number of biosolids application to any field to 1. Similarly, antimony (Sb) and thallium (Tl) limit the maximum number of biosolids applications to any field to 2 and 4, respectively (Table 7.10).

It should be emphasized that the estimates presented in Table 7.10 are based on very limited biosolids data and using the highest concentration reported by the Canadian Biosolids Survey*. Further, monitoring of biosolids for these and other unregulated metals should be conducted to obtain more extensive and reliable information.

* Canadian Biosolids Survey, Webber & Nicols, 1995; Webber & Bedford, 1996

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 7.10
Calculated Maximum Permissible and Probable Total Loadings of Selected Unregulated Heavy Metals in Land Applied Ontario Sewage Biosolids*

	Antimony (Sb)	Barium (Ba)	Beryllium (Be)	Silver (Ag)	Thallium (Tl)	Vanadium (V)
Row 1: Effects Based Criteria for Agricultural Land Use (mg/kg dry wt.) ⁽¹⁾	13	750 (1000)	1.2	20 (25)	4.1	200 (250)
Row 2: Soil Background Criteria for Agricultural Land Use (mg/kg dry wt.)	1.0	190	1.2	0.35	2.5	91
Row 3: Mean Concentrations in Rural Parkland Soils (mg/kg dry w) ^(1,2)	0.17	75	0.45	0.12	0.5	40
Row 4: Difference "Row 1 – Row 3" (mg/kg dry wt.) ⁽¹⁾	12.8	675 (925)	0.75	19.88 (24.88)	3.6	160 (210)
Row 5: Difference "Row 2 – Row 3" (mg/kg dry wt.)	0.8	115	0.75	0.23	2.0	51
Row 6: Maximum Permissible Loading Not to Exceed Row 1 (kg/ha) ^(1,3)	25.6	1350 (1850)	1.5	39.76 (49.76)	7.2	320 (420)
Row 7: Maximum Permissible Loading Not to Exceed Row 2 (kg/ha) ⁽⁴⁾	1.6	230	1.5	0.46	4.0	80
Row 8: Canadian Biosolid Median Concentrations (mg/kg dry wt.) – Table 7.8	64	417		43	16	15
Row 9: Canadian Biosolids Maximum Concentrations (mg/kg dry wt.) – Table 7.8	117	688	<1.5, 30 ⁽⁶⁾	81	131	54
Row 10: Maximum Number of Applications, of Biosolids with Median Metal Concentrations, Not to Exceed Row 6 and Row 7 Loadings, respectively ⁽⁶⁾	50, 3	404, 69		116, 1	56, 31	2667, 667

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 7.10 – cont'd
Calculated Maximum Permissible and probable Total Loadings of Selected Unregulated Heavy Metals in Land Applied Ontario Sewage Biosolids*)

	Antimony (Sb)	Barium (Ba)	Beryllium (Be)	Silver (Ag)	Thallium (Tl)	Vanadium (V)
Row 11: Maximum Number of Applications, of Biosolids with Maximum Metal Concentrations, Not to Exceed Row 6 and Row 7 Loadings Respectively⁽⁵⁾	27, 2	245, 42	6, 6	62, <1	7, 4	741, 185

*) Table provided by the Client

(1) Values in brackets are for medium and fine textured soils (MOEE 1997)

(2) Values are from MOEE (1993) - "Ontario Typical Range of Chemical Parameters in Soil, Vegetation, Moss Bags and Snow

(3) Values are 2 times those in Row 4 and assume 2,000,000 kg of soil dry wt in the 0-15 cm zone of biosolids incorporation

(4) Values are 2 times those in Row 5 and assume 2,000,000 kg of soil dry wt in the 0-15 cm zone of biosolids incorporation

(5) Based on maximum permissible biosolids application rate of 8 dry tonnes/ha/5 years. The number of years (not shown) to reach maximum permissible loadings would be – 5 x the maximum number of applications

(6) Canadian data not available - values are Table 7.8 data for US and UK biosolids, respectively

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

7.6 Fate

Many recent publications concerning the fate of heavy metals in land applied sewage biosolids were identified during the literature search and reference to them is included in the database. Much of the information was similar to that reported by Harrison et al. (1997) in their comprehensive review. There were numerous reports of increased total and extractable metal concentrations in soils resulting from land application of sewage biosolids. Similarly, there were numerous reports of increased metal concentrations in plants grown on sewage biosolids treated soils and of differential metal uptake related to plant species and variety.

Plant uptake studies generally show that metal bioavailability decreases in order of the following fractions: water-soluble > exchangeable > specifically adsorbed > organically bound, carbonate associated, Fe-Mn oxide > acid replaceable, residual inorganic.

Canet et al. (1997) fractionated the heavy metals in biosolids-amended soils into water-soluble plus exchangeable, organically bound, carbonate associated, and residual fractions. Most of the heavy metals were present in the carbonate and residual fractions, although substantial amounts of water-soluble plus exchangeable cadmium, and organically bound copper and nickel were found.

Similarly, Sloan et al. (1997) fractionated the heavy metals in biosolids-amended soils into exchangeable (Exch), specifically adsorbed (SA), Fe-Mn oxide and acid replaceable Ox/AR), residual organic (R-Org) and residual inorganic (R-In). In both control and biosolids amended soils 60-75% of the cadmium was found in the more easily extracted Exch and SA forms. Biosolids applications also increased the percentages of Ni and Zn in the Exch and SA fractions. Biosolids applications had little effect on the Exch and SA fractions of chromium and lead. Greater than 75% of chromium, copper, nickel, lead and zinc were found in the relatively stable Ox/AR, R-Org and R-In fractions of control and biosolids-amended soils. Cadmium, chromium and copper concentrations in lettuce grown on the soils were highly correlated with ($P < 0.001$) to metal concentrations in one or more of the Exch, SA, and Ox/AR soil chemical fractions.

The above fractionation studies indicate considerable differences in bioavailability among heavy metals added to soil in sewage biosolids. For example, a considerable proportion of the Cd added to soil remains either water-soluble or exchangeable (high bioavailability); a proportion of Ni and Zn remain exchangeable and specifically adsorbed (moderate bioavailability), a proportion of Ni and Cu remains organically bound (moderate bioavailability), but by far the largest proportions of all metals except Cd occur in the residual fractions (low bioavailability). Leaching potential for the metals is highly correlated with bioavailability but the consultant is unaware of any reports of significant metal loss from soil through leaching or run-off of soluble metal. Erosion losses invariably are associated with substantial loss of soil and biosolids.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Chromium VI is a strong oxidizing agent that is highly toxic in aquatic systems and there is concern in some quarters that it may present a toxicity problem in biosolids treated soils. Thus, the Draft EC Council Directive (2000) requires that soils be subjected to the Bartlett and James (1979) "chromium oxidation quick test". Biosolids application will be prohibited on soils in which >1:1 mol of Cr VI/g of soil (equivalent to 52 mg of Cr VI/kg of soil) is produced.

The Ontario Guideline for Use at Contaminated Sites (MOEE 1997) indicates that Cr VI in soil for agricultural use must be reduced to 10 and 8 mg/kg dry wt. for fine and medium textured soils respectively (Effects Based Approach) or 2.5 mg/kg (Background Approach). These limits are 12 to 48 times less than the maximum permissible total Cr guideline (120 mg/kg) for soils receiving biosolids (MOEE and OMAFRA 1996).

The Council for Agricultural Science and technology in the USA reported that very little soluble chromium is found in soils. Hexavalent chromium may be present in soil for a short time but is reduced to trivalent chromium and then changed to low solubility compounds. They also found that hexavalent chromium does not occur in many biosolids since it is reduced to the trivalent (less toxic) state during biosolids digestion. No Cr VI data is available for Ontario biosolids. Any trivalent chromium in soils is transformed to less soluble complexes that are difficult to leach. The Council concluded that Chromium is not expected to be a limiting factor in determining the quantity of sludge that may be applied to soil producing agronomic crops because:

- Plants can tolerate relatively high levels of chromium applied in sludge.
- Plants do not accumulate chromium even when it is present in the soil at high levels.
- There is evidence that chromium is required by humans and animals and that diets are deficient in chromium in certain areas.

In 1989, a U.S. Department of Agriculture Technical committee concluded there is no scientific basis to limit application of sewage sludge-Cr based on phytotoxicity. Similarly, there is no known basis to limit sludge-Cr to protect the food chain, livestock or children from Cr-toxicity due to sludge ingestion, or to protect soil biota or wildlife. The EPA accepted this conclusion and indicated that, considering both plant toxicity and groundwater contamination, there was no risk basis for establishing Cr limits for sewage biosolids. Their analysis was extensive considering a number of different sludge use and disposal scenarios, and many potential exposure pathways and is the most comprehensive Cr study to date. Based on the above, it appears that Cr is not a significant health or environmental concern in biosolids treated soils.

Mercury has been identified as a priority toxic substance in the Canadian environment and national environmental quality guidelines are under development for water, sediment and soil (Gaudet et al. 1995). Mercury concentrations in sewage biosolids may exceed 5 mg/kg (SYLVIS 1995, Webber and Bedford 1995, Webber and Nichols 1996) and are much larger than background soil concentrations of 0.02 to 0.15 mg/kg (Gaudet et al 1995). It has been demonstrated that sewage biosolids increase methyl mercury emissions from soil (Carpi et al. 1997) and this has contributed to concern about detrimental effects on crop and environmental quality. However, Granato et al. (1995) conducted studies at a dedicated sludge utilization site that had received up to 1317 tonnes/ha of sewage biosolids with a mean mercury concentration

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

of 3.3 mg/kg and found no increase in mercury concentrations of corn and wheat tissues. They concluded that uptake response slopes used in the US EPA's risk assessment models, to define a 17 kg ha⁻¹ cumulative mercury loading to soil (US EPA 1993) over-predicted mercury uptake by crops and were unnecessarily conservative.

7.6.1 Long Term Metal Availability

For several years now, there has been controversy concerning the ultimate fate of heavy metals in land applied sewage biosolids. It is recognized that the organic matter in biosolids plays a large role binding heavy metals and reducing their bioavailability. However, organic matter degrades continually in soil and there is speculation that the bioavailability of sewage biosolids heavy metals in soil will suddenly increase at some time in the future when the biosolids organic matter has disappeared.

A second school of thought on this subject is that metal adsorption capacity (e.g., iron and aluminum oxides, calcium carbonate, humic and fulvic acids) added with sewage biosolids will persist as long as the metals of concern persist in the soil and that the metals will remain in chemical forms not readily available for plant uptake. Therefore, the metal concentrations of plant tissue will reach a "plateau" as sewage biosolids mass loading increases and remain at this plateau after termination of sewage biosolids application.

Considerable effort has been directed toward determining whether the increased metal availability over time and/or plateau effects occur. Several sites in various countries have histories of very large sewage biosolids heavy metal loadings. Studies at these sites have not supported the increased metal availability over time hypothesis but it has been argued that these results cannot be trusted because records frequently are lacking or are inadequate.

Recently, a number of researchers in the USA have conducted carefully controlled studies to address the long term metal availability. Chang et al (1997) analyzed ten years of data from a field experiment in which Swiss chard was grown on soil with maximum biosolids and cadmium loadings of 2880 tonnes/ha and 176 kg/ha, respectively. Findings indicated that conditions necessary for both the plateau and the increased metal availability over time might be found but that neither was observed. The same experiment was used to study the effect of organic matter mineralization on cadmium availability in sewage biosolids treated soil (Hyun et al 1998). The organic carbon content of treated soil decreased approximately 40% during ten-years after biosolids application was terminated but there was no indication that cadmium solubility or bioavailability increased as organic carbon declined.

Logan et. al. (1997) conducted a six-year field study of cadmium, copper, nickel, lead and zinc in corn (*Zea mays* L.) and lettuce (*Lactuca sativa* L.) following one-time applications of sewage biosolids ranging from 0 to 300 tonnes/ha. Results indicated that cadmium, copper and zinc in corn exhibited a plateau type response whereas concentrations of these metals in lettuce increased linearly with biosolids application.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Brown et. al. (1998) conducted a field study to assess the bioavailability of cadmium in long term biosolids and cadmium salt amended soil. Two of the biosolids had cadmium concentrations of 13.4 and 210 mg/kg. A cadmium salt treatment with cadmium was added to soil at a rate equivalent to that added by the high cadmium biosolids applied at 100 tonnes/ha. Lettuce cadmium was compared between treatments and in relation to the soil cadmium/soil organic carbon ratio. With only 16% of added biosolids organic carbon remaining, the cadmium concentration in lettuce was not different from that of the initial

cropping. Moreover, significantly less cadmium was taken up by lettuce grown on biosolids amended soil than by lettuce grown on soil amended with equivalent rates of cadmium salt. The cadmium concentration in lettuce grown on the low cadmium biosolids treatment was not different from that grown on the control treatment. These results indicate that the potential hazards associated with food chain transfer of biosolids-applied cadmium are substantially lower than equivalent salt treatments and that the hazards do not increase over time.

The authors suggest that:

- Increases in cadmium adsorption observed in biosolids amended soil appear to be related to the inorganic complexing ability added to the soil with biosolids
- The inorganic adsorption capacity added to soil with biosolids prevents increased cadmium availability suggested by the increased metal availability with time hypothesis.

Thus, Brown et. al. concluded that evidence was not obtained for increased sewage biosolids heavy metal bioavailability due to organic matter degradation but that a "plateau effect" (plant metal concentration does not increase beyond some level) may be observed depending upon the crop being grown.

7.6.2 Heavy Metal Transport

Harrison et al. (1997) reviewed the effect of biosolids application on the movement of metals in soil and concluded that trace metals remain primarily in the zone of incorporation even during long term biosolids application at rates approaching 500 tonnes/ha.

Several recent publications report similar findings. For example, Sloan et al. (1998) reported virtually complete recovery of cadmium, copper, nickel, lead and zinc in the surface 30 cm of soil, sixteen years after biosolids application. Recovery of only fifty-nine percent of the chromium, was attributed to incomplete extraction from the soil. Berti and Jacobs (1998) observed no vertical movement of cadmium, chromium, copper, nickel, lead and zinc below the 15 to 30 cm sampling depth in biosolids treated soil but observed lateral movement which was attributed to soil movement during tillage operations.

7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Although the above information is representative of a very large preponderance of the literature, occasional citations such as McBride et al. (1997) claim the potential for significant movement of biosolids metals out of the zone of incorporation by physico-chemical or biological processes which could contribute to groundwater contamination.

However, biosolids and heavy metal loadings to Ontario soils are miniscule compared with the experimental conditions discussed above suggesting that agricultural use of biosolids in Ontario poses no significant risk for heavy metal contamination of groundwater.

7.7 Findings and Conclusions from the Literature Review

1. Large amounts of research have focused on a few heavy metals considered to be the most hazardous and guidelines/regulations for land application of sewage biosolids have been developed to limit loadings of these constituents to agricultural land. Sewage biosolids application rates are generally agronomically based so as not to exceed crop nutrient requirements.
2. There is much less Canadian than US and international research on the effects of heavy metals in land applied sewage biosolids. However, Canadian and in particular, Ontario recommended practices are among the most conservative in the world. Considering the absence of detrimental effects in studies with high metal concentrations and application rates, it appears that recommended land application practices in Ontario present no significant risk to humans and the environment.
3. The regulated metals can be considered Group I contaminants for which current Ontario guidelines are adequate to protect the well being of soils, crops, animals, humans and ground and surface water qualities.
4. The following unregulated metals and compounds in biosolids were considered: aluminum, antimony, asbestos, barium, beryllium, boron, cyanide, fluoride, manganese, silver, thallium and tin. Based on very limited information, it was concluded that loadings of unregulated metal in land applied sewage biosolids are unlikely to exceed "Effects Based approach" limits developed for contaminated site cleanup of soil for agricultural use, however, they may exceed the "Background approach" limits. Thus, the unregulated heavy metals are Group II contaminants requiring further research.

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7. LITERATURE REVIEW OF THE FATE AND EFFECTS OF HEAVY METALS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

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8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND

8.1 Introduction

Wastewater contains feces and urine from both humans and animals. Human pathogens enter municipal wastewater from patients in hospitals, or from any sick person or individual carrying the organisms. Animal pathogens enter municipal wastewater from meatpacking and processing facilities, street run-off and from rats and/or other vectors in contact with sewage or sewers. During the course of typical wastewater treatment, the pathogens are reduced in number but those remaining concentrate in the untreated sludge (Smith and Farrell 1996).

8.2 Fate and Effects

The four major types of pathogenic organisms in municipal wastewater and biosolids are: bacteria, viruses, protozoa and helminth worms and those of primary concern are presented in Table 8.1, accompanied by their disease/symptoms. It is evident from information on typical pathogen concentrations in sewage biosolids (Table 8.2) and infective doses (Table 8.3) that observing recommended land application practices is mandatory to minimize a human health risk. Moreover, the information in Table 8.4 indicates significant persistence of pathogens on both soil and plants. Some helminth worms may be extremely persistent because a protective shell (cyst) forms around the ova when they are stressed.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 8.1
Principal Pathogens of Concern in Municipal Wastewater and Biosolids
(after Smith and Farrell 1996)

Pathogen	Disease/Symptoms
Bacteria	
<i>Salmonella</i> spp.	Salmonellosis (food poisoning), typhoid
<i>Shigella</i> spp.	Bacillary dysentery
<i>Yersinia</i> spp.	Acute gastroenteritis (diarrhea, abdominal pain)
<i>Vibrio cholera</i>	Cholera
<i>Campylobacter jejuni</i>	Gastroenteritis
<i>Escherichia coli</i> (pathogenic strains)	Gastroenteritis
Viruses	
Poliovirus	Poliomyelitis
Coxsackie virus	Meningitis, pneumonia, hepatitis, fever, etc.
Echovirus	Meningitis, paralysis, encephalitis, fever, etc.
Hepatitis A virus	Infectious hepatitis
Rotavirus	Acute gastroenteritis with severe diarrhea
Norwalk Agents	Epidemic gastroenteritis with severe diarrhea
Reovirus	Respiratory infections, gastroenteritis
Protozoa	
<i>Cryptosporidium</i>	Gastroenteritis
<i>Entamoeba histolytica</i>	Acute enteritis
<i>Giardia lamblia</i>	Giardiasis (diarrhea and abdominal cramps)
<i>Balantidium coli</i>	Diarrhea and dysentery
<i>Toxoplasma gondii</i>	Toxoplasmosis
Helminth Worms	
<i>Ascaris lumbricoides</i> (roundworm)	Digestive disturbances, abdominal pain
<i>Ascaris suum</i> (roundworm)	Can have symptoms: coughing, chest pain
<i>Trichuris trichiura</i> (whipworm)	Abdominal pain, diarrhea, anemia, weight loss
<i>Toxocara canis</i> (cat roundworm)	Fever, abdominal discomfort, and muscle aches
<i>Taenia saginata</i> (beef tapeworm)	Nervousness, insomnia, anorexia
<i>Taenia solium</i> (pork tapeworm)	Nervousness, insomnia, anorexia
<i>Necator americanus</i>	Hookworm disease
<i>Hymenolepis nana</i> (dwarf tapeworm)	Taeniasis

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 8.2
Typical Pathogen Levels in Unstabilized/Digested Sewage Biosolids
 (after Smith and Farrell 1996)

Pathogen	No./100 mL
Virus	2,500 - 70,000
Fecal Coliform Bacteria	1,000,000,000
<i>Salmonella</i>	8,000
<i>Ascaris lumbricoides</i>	200 - 1,000

TABLE 8.3
Reported Infective Dose Data
 (after Smith and Farrell 1996)

Pathogen	Infective Dose	Range
Bacteria		
<i>Clostridium perfringens</i>	10 ⁶	10 ⁶ - 10 ¹⁰
<i>Escherichia coli</i>	10 ⁴	10 ⁴ - 10 ¹⁰
<i>Salmonella</i> (various spp.)	10 ²	10 ² - 10 ¹⁰
<i>Shigella dysenteriae</i>	10 - 10 ²	10 - 10 ⁹
<i>Shigella flexneri</i>	10 ²	10 ² - 10 ⁹
<i>Streptococcus faecalis</i>	10 ⁹	10 ⁹ - 10 ¹⁰
<i>Vibrio cholera</i>	10 ³	10 ³ - 10 ¹¹
Viruses		
Echovirus 12	HID50 919 PFU; HID1 17 PFU estimated	17 - 919 PFU
Poliovirus	1 TCID50; <1 PFU	4 x 10 ⁷ TCID50 for infants; 0.2 to 5.5 x 10 ⁶ PFU for infants
Rotavirus	HID50 10 ffu; HID25 1ffu estimated	0.9 to 9 x 10 ⁴ ffu
Protozoa		
<i>Entamoeba coli</i>	1 - 10 cysts	1 - 10 cysts
<i>Cryptosporidium</i>	10 cysts	10 - 100 cysts
<i>Giardia lamblia</i>	1 cyst estimated	NR
Helminth Worms		
	1 egg	NR

Where: HID = human infective dose; TCID50 = tissue culture infectious dose for 50% response; PFU = plaque forming units; ffu = focus forming units; NR = not reported

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 8.4
Survival Times of Pathogens on Soil and Plants
 (after Smith and Farrell 1996)

Pathogen	Soil		Plants	
	Absolute Maximum	Common Maximum	Absolute Maximum	Common Maximum
Bacteria	1 year	2 months	6 months	1 months
Viruses	6 months	3 months	2 months	1 months
Protozoa	10 days	2 days	5 days	2 days
Helminth Worms	7 years	2 years	5 months	1 months

Information concerning the environmental impacts of pathogens in land applied sewage biosolids has been comprehensively and frequently reviewed (Smith and Farrell 1996, Gaus et al. 1997, Smith 1996, Straub et al. 1993, Kowal 1985). Moreover, Smith (1966) cites sixteen additional review articles on this subject published between 1978 and 1990. These articles indicate that environmental impacts depend upon a large number of factors including, biosolids treatment and storage, temperature, moisture and light conditions during and following land application, elapsed time between land application and biosolids incorporation into soil, the degree of incorporation into soil, soil properties such as temperature, permeability, pH, and clay, organic matter and water contents, the nature of crops grown on treated land, the degree of human and animal access to treated land, etc. Despite this complexity, the review articles consistently conclude that properly managed land application of stabilized biosolids presents minimal risk to human and animal health and the environment.

The literature study conducted by Washington University for the Northwest Biosolids Management Association (Gaus et al. 1997) concluded that, in general, the movement and survival of pathogens in soil depends on the characteristics of the soil and type of pathogen, but not on pathogen concentration. Important soil characteristics include moisture content, texture (clay, silt, sand), organic matter content, pH, permeability and temperature. In most cases, pathogens are retained in the upper 5 to 15 cm of soil. Higher clay content in a soil leads to increased viral retention, but the presence of humic acids (due to decomposition of organic matter), frequent drying and wetting cycles and the percolation of de-ionized water tend to decrease viral retention in the soil. Parasites are generally strained out at the soil surface because they are larger and heavier than bacteria and viruses. Very few bacteria have been detected in groundwater from biosolids-amended sites. The study also concluded that even though surface water runoff has been found to contain some indicator bacteria, bacterial contamination of surface water seems unlikely as the survival time of enteric bacteria and viruses in soil is relatively short.

Kowal (1985) concluded that the levels of bacteria such as fecal coliforms, fecal streptococci and salmonella were significantly lower at liquid sewage biosolids application sites than at wastewater spray applications sites. Spray application of biosolids did not represent a health threat for individuals more than 100 m downwind of the application site. Kowal also indicated that the survival time of organisms in aerosols depends on the application method, nozzle type and spray velocity, and the relative humidity and wind speed during application. Organisms transported in aerosols are much more susceptible to inactivation than in soil due to solar radiation, desiccation and high temperature.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Casson (1996) conducted a laboratory study to investigate the survival and transport of selected organisms (*Salmonella typhimurium*, *Shigella sonnei*, bacteriophage (MS-2) and Poliovirus type 1) in sewage biosolids amended soils exposed to a range of moisture conditions. In addition, the effect of rainfall intensity on surface transport of these organisms was examined. Casson's (1996) study concluded that:

- Two log reductions were observed after 10 to 20 days. Decay rates ranged between 0.08 log per day to 0.4 log per day.
- No relationship was established between decay rates and soil depth because of analytical variability.
- Increasing rainfall intensity caused an increase in both the runoff of total solids and concentrations of the organisms in the runoff water;
- As much as 75% of the *Salmonella typhimurium*, 30% of *Shigella sonnei* were removed at a rainfall intensity of 12.7 cm/ hr (poliovirus and bacteriophage were not studied).

8.3 Epidemiological Studies

Epidemiology is a branch of medical science that deals with the occurrence, distribution and control of disease in populations. There are two types of epidemiological study:

- Prospective studies that involve choice of the population to be studied as well as the conditions to be investigated.
- Surveillance studies that involve a general survey of a given event (i.e the occurrence of diarrhea in a given area).

A prospective epidemiological study of landspreading sewage biosolids (4 to 10 dry tonnes/ha/year) was conducted by the Ohio University and Ohio Farm Bureau Federation in Ohio during the five-year period 1978 to 1982 (US EPA, 1985). A major objective of the study was to evaluate health risks to rural residents and their livestock and to determine if the presence of biosolids on land increased the risk of disease. Forty-seven (47) farms involving 164 people in 78 families were randomly chosen to receive application of sewage biosolids and 46 farms involving 130 people in 53 were chosen as controls families (no sewage biosolids applications). The farms were spread over 3 different geographic areas and only anaerobically digested or aerobically digested sewage biosolids were applied. All 47 treated farms participated in the experiment for at least one year, 36 farms participated for 2 years and 13 farms participated for 3 years. All 46 control farms participated for at least one year, 37 farms participated for 2 years and 13 participated for 3 years.

The ratios of males to females were about 1.4:1 and 1.3:1 for the treated farms and control farms. The majority of the people in both groups were in the 50 – 59 age range, but some were in the 0 – 19 and 60 – 69 year old ranges. On average, about 9 – 11 hours per week were spent in the field and 4 – 6 hours per week with livestock. Both groups consumed about the same amount of fruit and vegetables grown on their farms. The treated group consumed a higher percent of meat grown on their farms than the control group.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Questionnaires were completed on human and livestock health prior to the study and then on a monthly basis. Blood and fecal samples were collected for microbiological testing on a quarterly basis and Tuberculin testing was conducted annually. Intensive observations were made of beef cattle herds on the treated and control farms. Human health data were evaluated by a panel of 2 infectious disease specialists, a toxicologist and an epidemiologist. Animal (horses, sheep, cattle, avian species, swine, cats and dogs) health data were evaluated by a veterinary clinician and an epidemiologist.

The epidemiological study conducted by the Ohio University and Ohio Farm Bureau Federation in Ohio (US EPA, 1985) concluded that health risks were not significant when sewage biosolids are applied at low application rates of 4 to 10 dry tonnes/ha/year and according to the agronomic management systems used in the study. The risks of respiratory illness (runny nose, sore throat, nasal congestion or hoarseness, chest congestion and cough), gastro-intestinal illness from exposure to *Salmonellae*, *Shigella* spp. or *Campylobacter* spp. or general symptoms (fever, headache, general muscular aches and pains) were not significantly different between treated and control groups. Similarly, there were no significant differences in the health of domestic animals (constipation, diarrhea, blood in feces, cough, nasal discharge and difficult breathing) on treated and control farms. Viral infections and increase in antibodies to *Salmonellae* among household members were observed but there were no significant differences between treated and control groups. The frequency of serological antibodies and the frequency of associated illnesses were similar among treated and control groups.

As part of the same study, soil and forage samples were collected on 3 farms for parasitic ova and larvae analysis both before sewage biosolids application and 7, 14 and 28 days after sewage biosolids application. Samples were obtained from sewage biosolids treated and control pasture areas. This study concluded that the risk of parasite transmission attributable to sewage biosolids application (at rates of 2 – 10 tonnes per ha dry wt.) on cattle pasture appears to be indistinguishable from farms without such applications.

Surveillance programs involving surveys of disease outbreak events are a widely used alternative to prospective epidemiological studies. They are conducted on an ongoing basis in many countries to determine factors responsible for the events. In Ontario, disease surveillance programs are conducted by the Medical Officers of Health. A long history of disease surveillance in the UK indicates that properly managed land application of stabilized biosolids presents minimal risk to human and animal health and the environment (See Section 8.4.3 below).

8.4 Regulations

8.4.1 Ontario

Based on the considerable body of evidence that properly managed land application of stabilized biosolids presents minimal risk to human and animal health and the environment, complexity and high cost of pathogen analyses, and considerable uncertainty associated with the analytical results for pathogens, the Ontario "Guidelines for the Utilization of Biosolids and Other Wastes on Agricultural Land" (MOEE & OMAFRA 1996) require that all sewage biosolids must be "stabilized" by an Ontario Ministry of Environment approved process prior to being spread on agricultural land.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Currently approved stabilization processes include:

1. Anaerobic digestion at 35°C for 15 days followed by anaerobic storage at ambient temperature for an additional 15 days
2. Aerobic digestion at ambient temperature for a total solids retention time of 45 days. Total retention time is defined as the sum of solids retention times in the secondary biological treatment system and in the aerobic digester.

Other stabilization processes are reviewed on an individual basis. Recognizing that these processes only significantly reduce the levels of pathogenic organisms in sewage biosolids, precautionary measures to protect public health, such as restricting public access to land application sites and controlling animal grazing and cropping practices for a period of time, are included in the Ontario guidelines.

The Water Environment Association of Ontario sponsored a study to determine reduction and densities of fecal coliforms and other pathogens remaining after anaerobic and aerobic digestion of sewage biosolids (WEAO 1999). The study included 6 sewage treatment plants using aerobic digestion and 8 using anaerobic digestion. Results showed that all 14 plants reduced fecal coliforms to less than the US EPA limit of 2 million organisms per gram dry solids for sewage biosolids agricultural land application (see Section 8.4.2 below).

8.4.2 United States - Environmental Protection Agency (US EPA)

The regulatory approach for pathogens chosen by US EPA requires communities intending to use sewage biosolids in agriculture to meet certain disinfection and vector reduction conditions. Two categories of disinfection requirement (Classes A and B) are defined depending upon the intended use.

The Class A requirement specifies that all pathogens in the sewage biosolids shall be reduced to "below detectable levels" and these biosolids can be used on land with high public exposure and without site management restrictions, for example, parkland and gardens, because they present no risk to public health.

The Class B requirement specifies that the level of pathogenic organisms in sewage biosolids must be significantly reduced but some pathogens remain. Consequently, Class B biosolids can only be used on lands with limited public access such as agricultural lands, and precautionary site management measures must be exercised to insure that their use involves no greater health and environmental risk than Class A biosolids. Precautionary measures related to crop harvesting, animal grazing and public access for defined periods of time are required to allow environmental factors (e.g., sunlight, desiccation) to further reduce pathogens.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Class A Biosolids

The goals of Class A processes are to achieve the following pathogenic densities¹:

- *Salmonella* <3 MPN/4g TS or fecal coliform <1000 MPN/g
- Enteric viruses <1PFU/4g TS
- Viable Helminth ova <1/4g TS

¹ Where MPN = Most Probable Number; g = gram; TS = Total solids; PFU = Plaque forming units

Class B Biosolids

The goals of Class B processes are to achieve the following pathogen densities²

- Geometric mean fecal coliform <2,000,000 MPN or CFU/g TS

² Where CFU = Colony forming units

The technical rationale for the requirements specified above is as follows:

- Fecal coliform densities of 1,000 MPN/g of sewage biosolids are sometimes used to indicate the absence of pathogens in sewage biosolids (US EPA 1996);
- When *Salmonella* are known to be abundant in unprocessed sewage biosolids, their absence in the processed material can also indicate absence of other pathogens (US EPA 1996);
- The experimental work carried out by Yanko (1988) demonstrated that when fecal coliform densities are below 1,000 MPN/g, the incidence of *Salmonella* was low and uniformly distributed between 0 and 3;
- A fecal coliform density of 1,000 MPN/g or lower indicates that regrowth of microorganisms has not occurred to a significant extent and that *Salmonella* are unlikely to be present (US EPA 1996).

The process alternatives for achieving Class A and Class B biosolids, including the options for vector attraction reduction are presented briefly in 40 CFR 503, Standards for the Use or Disposal of Sewage Sludge (US EPA 1993) and are described in detail in (US EPA 1992, revised 1999).

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

The regulation of pathogens in the Final Part 503 Rule for both Class A and Class B sewage biosolids is not based on risk assessment because methodologies were not developed sufficiently to make such calculations. The fecal coliform limit for Class B sewage biosolids was based on a survey of fecal coliform reductions and densities remaining in anaerobically and aerobically digested sewage biosolids taken from a large number of plants across the U.S. Additional considerations were that there were no reported/found cases that link epidemic outbreaks to sewage biosolids application. Recently, a framework consisting of three phases; problem formulation, analysis (which includes characterization of exposure and human health effects) and risk characterization has been developed and preliminary pathogen risk assessments have been conducted (Colford et al. 1999, US EPA 1996). However, much additional information on pathogens in biosolids and their fate and effects following land application sewage is required for the risk assessment approach to be practical.

8.4.3 United Kingdom

Much has been done to minimize the potential transmission of pathogens by reducing infectivity of sewage biosolids through effective treatment processes and then matching efficiency of pathogen removal to operational restrictions on sewage biosolids application practices and land use (Smith 1996). These requirements have been implemented in the UK through the Sludge (Use in Agriculture) Regulations (SI 1989) which are supported by a Code of Practice (i.e., recommended procedures) for sewage biosolids use (DoE 1989). Examples of effective sewage biosolids treatment processes described in the Code of practice are presented in Table 8.5. Surveillance of human and animal disease in the UK has shown these measures to be effective in preventing infection from sewage biosolids-borne pathogens following land application (Pike and Carrington 1986).

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 8.5
Relative Efficacies of Various Methods of Sewage Biosolids Treatment in Reducing
Numbers of Different Pathogens or Their Period of Survival
 (after Smith 1996)

Process	Relative Reduction		
	Good	Moderate	Poor
Raw Biosolids Storage		Viruses Bacteria	<i>Ascaris ova</i> <i>Taenia ova</i> , <i>Cryptosporidium</i> oocysts
Digestion	Viruses <i>Entamoeba ova</i> <i>Heterodera ova</i> <i>Cryptosporidium</i> oocysts	Hookworm ova Bacteria <i>Taenia ova</i>	<i>Ascaris ova</i> ³
Composting	Viruses Bacteria Fungi Helminth ova		
Lime Treatment ¹	Bacteria		<i>Ascaris ova</i>
Heat Treatment ²	Viruses Bacteria <i>Cryptosporidium</i> oocysts Helminth ova		
Irradiation	Viruses Bacteria	<i>Ascaris ova</i>	

^{1,2} Include N-Viro process

² Includes Swiss Combi process

³ Anaerobic digestion at temperatures >36°C will inactivate depending upon exposure

In the last three years, more than ever before, there has been public concern in the UK about food production methods and food safety, and land application remains vulnerable to adverse publicity because of sewage biosolids origin and composition (Davis 1999). Odour nuisance is often the trigger but public health and the environment are major concerns. Continued land application cannot be guaranteed simply by compliance with all relevant legislation and guidelines even though they are more restrictive than for any other wastes (industrial and farm) spread on the land. Instead it depends on the willingness of farmers to allow their land to be used and on public acceptance.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

Recently, positive action was taken to restore confidence in land application and gain support from farmers, their customers and the public at large. The key to this action was recognition that the problem was market and customer driven and that market demands for food safety exceeded the regulatory legislation and guideline requirements. An agreement called "The Safe Sludge Matrix" (1999) was developed between Water UK (for the water utilities) and the British Retail Consortium (BRC) representing major retailers wanting assurance that biosolids recycling to land is safe. The negotiations leading to the agreement included input from various other stakeholders such as farm organizations, food manufacturers, food processors and government agencies.

The agreement consists of a table (Table 8.6) linking crop types with clear guidance on the minimum level of treatment for sludge products applied to that crop or rotation. Main points in the matrix are:

- Rapid phasing out of untreated sludge on land with immediate effect from 31 December 1998 for grazing land and for grass/maize silage.
- From 31 December 1998, treated sludge may not be applied to the surface of grassland, and there are more stringent controls for growing vegetable crops on sewage biosolids treated land.
- There is provision for land application of advanced treated sewage biosolids which are virtually pathogen-free.

By taking account of customer requirements (i.e., market demands), the "The Safe Sludge Matrix" increased confidence in sewage biosolids recycling to land.

8.4.4 European Community

The need to treat sewage biosolids to reduce the likelihood of spreading pathogens into the environment and to build consumers' confidence in land application is well recognized (Draft EC Council Directive 2000). A structured approach, similar to that in the UK Safe Sludge Matrix, has been developed in which three types of biosolids treatment; A, B and C are defined (Table 8.7). Treatment rigour decreases and restrictions on use increase in the order A, B and C (Table 8.8). The restrictions ensure that using the treatment types B and C biosolids involves no greater public health and environmental risk than using treatment type A biosolids.

The European Community treatment types A and C biosolids approximate the US EPA (1993) Classes A and B biosolids, respectively.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 8.6
“The Safe Sludge Matrix (1999)”

Crop Type	Untreated Sludges	Treated Sludges ¹	Advanced Treated Sludges ²
Fruit	N	N	Y
Salads	N	N (30 month harvest interval applies)	Y
Vegetables	N	N (12 month harvest interval applies)	Y
Horticulture	N	N	Y
Combinable & Animal Feed Crops	Y Target end date 31/12/99	Y	Y
Grass - Grazing	N	N (Deep injected or ploughed down only)	Y
Grass - Silage	N	Y	Y
Maize - Silage	N	Y	Y

Note: Y All applications must comply with the current DoE Code of Practice and certain additional crop and harvest interval restrictions.

N Applications not allowed (except where stated conditions apply)

1. A range of different treatment processes are used to reduce the fermentability and possible health hazards associated with sewage sludge. These rely on biological chemical or heat treatment. The most common form of treatment is anaerobic digestion.

2. Advanced treatment is a term used to describe treatment processes that are capable of virtually eliminating any pathogens that may be present in the original sludge.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 8.7
Recommended European Cropping Practices According to Degree of Sewage Biosolids Treatment
(after Draft EC Council Directive 2000)

Crop Type	Treatment type A	Treatment type B	Treatment type C
Pastureland	Yes, no grazing in the six weeks following spreading	No	No
Forage crops	Yes	Yes, no harvesting in the six weeks following spreading	No
Arable land	Yes	Yes	Yes, deep injection or immediate ploughing down
Fruit and vegetable crops in contact with the ground	Yes	Yes, no harvest in the 3 months following spreading	No
Fruit and vegetable crops in contact with the ground eaten raw	Yes	No	No
Parks, green areas, city gardens, all urban areas where the general public has access	Yes, only well stabilized and odourless sludge	No	No
Silviculture areas where the general public has access	Yes	Yes, no access to the public in the 3 months following spreading	Yes, no access to the public in the 10 months following spreading
Forests and woods	No	No	No
Land reclamation	Yes	Yes, no access to the public in the 3 months following spreading	Yes, no access to the public in the 10 months following spreading

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

TABLE 8.8
Processes to Achieve European Biosolids Treatment Types A, B and C
 (after Draft EC Council Directive 2000)

<p><u>Treatment type A</u></p> <ul style="list-style-type: none"> • In-vessel composting maintaining a temperature of 60°C in all material for 1 week; • Windrow composting maintaining a temperature of 50°C for 15 days with at least 1 turning of the windrow; • Thermal drying ensuring that the temperature of the sludge particles is higher than 80oC with a reduction of water content to less than 10% and maintaining a water activity above 0.90 in the first hour of treatment; • Thermophilic aerobic stabilization at a temperature of at least 55oC for 20 hours as a batch, without admixture or withdrawal during the treatment; • Thermophilic anaerobic digestion at a temperature of at least 53oC for 20 hours as a batch, without admixture or withdrawal during the treatment; • Thermal treatment of liquid sludge for a minimum of 30 minutes at 70oC followed by mesophilic anaerobic digestion at a temperature of 35oC with a mean retention period of 12 days; • Conditioning with quicklime reaching a pH of 12 or more and maintaining a temperature of at least 55oC for 2 hours; • The process shall be initially validated through a 5 log₁₀ reduction of a test organism such as Salmonella Senftenberg W 775. • The treated sludge shall not contain Salmonella spp in 50 g (wet weight) and have less than 5x10³ CFU/g of <i>Escherichia Coli</i>.
<p><u>Treatment type B</u></p> <ul style="list-style-type: none"> • Thermophilic aerobic stabilization at a temperature of at least 55oC with a mean retention period of 20 days; • Thermophilic anaerobic digestion at a temperature of at least 53oC with a mean retention period of 20 days; • Conditioning with slaked lime ensuring a homogenous mixture of lime and sludge. The mixture shall reach a pH of 12.5 directly after liming and keep a pH of at least 12 for 24 hours.
<p><u>Treatment type C</u></p> <ul style="list-style-type: none"> • Mesophilic aerobic digestion at a temperature of 35oC with a mean retention period of 12 days for primary digestion followed by a secondary stage with a minimum retention period of 14 days; • Extended aeration at ambient temperature as a batch, without admixture or withdrawal during the treatment period¹; • Storage in liquid form at ambient temperature as a batch, without admixture or withdrawal during the storage period².

¹ The minimum time length of the treatment shall be laid down by the competent authority taking into consideration the prevailing climatic conditions in the area where the treatment plant is located.

² The minimum time length of the treatment shall be laid down by the competent authority taking into consideration the prevailing climatic conditions in the area where the treatment plant is located.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

8.5 WEAO Biosolids Stakeholder Workshop – Concerns and Recommendations

A high level of concern was expressed to the WEAO Biosolids Stakeholder Workshop about the potential for disease transmission resulting from land application of sewage biosolids. Stakeholders participating in the Workshop represented the well-informed public and their opinions warrant serious consideration.

Pathogen concerns arise from a variety of issues including: recent *E. coli* contamination of the Walkerton ON water supply; evidence that microbial contamination of tile drainage water may occur rapidly following sewage biosolids or animal manure application; fear that surface water and private water wells may be contaminated; fear of aerial disease transmission and perceived inadequate monitoring and control of sewage biosolids application.

Public acceptance is crucial to the success of land application of sewage biosolids programs. Involving the public in exploring pathogen issues is suggested to improve understanding and acceptance of land application. It is recommended that:

- A task force involving medical experts, farmers, public representatives, and biosolids generators and regulators should be formed to explore pathogen issues and develop a protocol for establishing pathogen criteria for biosolids and land application site management criteria to insure the safety of farmers and the general public. Criteria development may require epidemiological information on the health effects of land application.

Information, not presently available, for pathogens in Ontario sewage biosolids and their fate following land application will be required to satisfy the task force objective. The following research programs are recommended to obtain this information:

- A survey should be conducted to develop a representative database of pathogen information for land applied sewage biosolids in Ontario.
- Small plot field studies should be conducted to determine pathogen persistence in soil and the incidence and extent of surface and groundwater contamination with pathogens following sewage biosolids application to Ontario agricultural land.

8.6 Conclusion from the Literature Review

Pathogens in land applied sewage biosolids are Group II contaminants requiring additional research.

8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

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8. LITERATURE REVIEW OF THE FATE AND EFFECTS OF PATHOGENS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND - cont'd

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9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND

9.1 Introduction

The range of organic compounds known to exist in sewage biosolids is quite extensive and diverse. Trace quantities of these compounds can potentially be transferred into agricultural soils by the spreading of sewage biosolids. The environmental consequences of trace organic pollutants in sewage biosolids-amended soils have been only investigated to a limited extent in part because analytical techniques which can be used to identify and quantify very low concentrations of organics have only been developed in recent years and are often labour intensive and expensive (Smith 1996).

The Literature Review will be presented in three parts. The first part (Section 9.2) reviews general properties and notes issues surrounding organic contaminants. The second part (Section 9.3) provides the US perspective and the third part (Section 9.4) notes studies in Canada and how they apply to organic contaminants.

9.2 Fate

Several authors in the US and elsewhere (Davis et al. 1984; Drescher-Kaden et al. 1992; Jacobs et al. 1987; Lester 1983; Overcash 1984; Rogers 1987; Sweetman 1991) have reviewed the occurrence, fate and potential impact of trace organic contaminants in sewage biosolids and sewage biosolids-amended agricultural soils. Table 9.1 (Smith 1996) presents a synopsis of the findings. The consensus of opinion based on the available information is that organic contaminants in sewage biosolids applied to agricultural lands are unlikely to cause significant environmental impact.

Table 9.1 (Smith 1996) also provides a summary of major transport and fate pathways of the common organic compounds found in sewage biosolids (Smith 1996). The main transport and fate pathways include adsorption, volatilization and degradation. Many of the organic compounds shown in the table can become strongly adsorbed onto soil organic matter and/or undergo degradation which can result in a reduction in the potential for plant uptake. With the exception of these five compounds: monocyclic aromatics, chlorobenzenes, short-chained halogenated aliphatics, aromatic and alkyl amines, and phenols, none of the other organic compounds found in biosolids have the potential to leach into groundwater. Chlorobenzenes, short-chained halogenated aliphatics, and aromatic and alkyl amines tend to be persistent in soils and thus may be leached into the groundwater; the other compounds are rapidly degraded and thus should not result in any significant environmental impact (Smith 1996).

The following values presented by the MOEE (1991) can generally be used to determine the environmental persistence and bioaccumulation potential of organic compounds:

Element Name	EndPoint and Units	Low Potential	Moderate Potential	High Potential
Environmental Persistence	$t_{1/2}$ (days)	<10	< 50 to 100	> 100
Bioaccumulation Potential	BCF Log K_{ow}	< 20 < 2	> 500 to 15000 >4.0 to 6.0	> 15000 > 6.0

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.1
Synopsis Of The Properties, Occurrence, Fate And Transfer Of The Principal Organic Contaminant Groups Found In Sewage Sludge And Sludge Amended Soils
 (after Smith 1996)

Compound Group	Physico-chemical properties	Concentration in sludge	Degradation	Leaching potential	Plant Uptake	Transfer to animals
Polynuclear Aromatic Hydrocarbons (PAHs)	Water soluble/volatile to lipophilic	1-10 mg/kg	Weeks – 10 yrs Strongly absorbed by organic matter in soil	None	Very poor Foliar absorption	Possibly but rapidly metabolised Not accumulated
Phthalate acid esters	Lipophilic, hydrophobic and non-volatile	1-100 mg/kg	Rapid with half life of <50d	None	Root retention Not Translocated	Very limited
Linear alkylbenzene sulphonates (LAS)	Lipophilic	50-15000 mg/kg	Very rapid	None	None	None
Alkylphenols	Lipophilic	100-3000 mg/kg	Rapid < 10 d	None	Minimal Root retention	Minimal
Polychlorinated biphenyls (PCBs)	Complex, >200 congeners, low water solubility, highly lipophilic and semi-volatile	1-20 mg/kg	Very persistent Half life several years Strongly absorbed by soil organic matter	None	Root retention Foliar absorption Minimal root uptake Minimal translocation	Possible into milk/tissues by soil ingestion Long half-life
Polychlorinated dibenzodioxins and furans (PCDDs/PCDFs)	Complex, 75 PCDD congeners, 135 PCDF congeners, low water solubility, highly lipophilic and semi-volatile	< few µg/kg	Very persistent Half life several years Strongly absorbed by soil organic matter	None	Root retention Foliar absorption Minimal root uptake Minimal translocation	Possible into milk/tissues by soil ingestion Long half-life
Organochlorine pesticides	Varied, lipophilic to hydrophilic, some volatile	< few mg/kg	Slow > 1 year Less by volatilization	None	Root retention Translocation not important	Via soil ingestion, persistent in tissues
Monocyclic aromatics	Water soluble and volatile	< 1 – 10 mg/kg	Rapid	Moderate	Limited due to low persistence Rapidly metabolised	Rapidly metabolised
Chlorobenzenes	Water soluble/volatile to lipophilic	< 0.1 – 50 mg/kg	Low MW types lost by volatilization Higher MW types persistent	High to low	Possibly via roots and foliage May be metabolized	Important for persistent compounds

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.1 –cont'd
Synopsis Of The Properties, Occurrence, Fate And Transfer Of The Principal Organic Contaminant Groups Found In Sewage Sludge And Sludge Amended Soils
 (after Smith 1996)

Compound Group	Physico-chemical properties	Concentration in sludge	Degradation	Leaching potential	Plant Uptake	Transfer to animals
Short-chained halogenated aliphatics	Water soluble and volatile	0 – 5 mg/kg	Low MW types lost by volatilization	Moderate	Foliar absorption Possible translocation	Low
Aromatic and Alkyl amines	Water soluble and low volatility	0 – 1 mg/kg	Slow	High	Possible	Low
Phenols	Varied, lipophilic, high water solubility and volatile	0 – 5 mg/kg	Rapid	Moderate to low	Possible via roots and foliage	None

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

Table 9.1 also shows that uptake and translocation of organic contaminants by plants occurs only to a limited extent or not at all from sewage biosolids amended soil. From the table it can be seen that uptake is minimal for the polyhalogenated compounds and for polyaromatic hydrocarbons. Kampe and Leschber (1989) showed that there was no transfer of chlorinated hydrocarbons, hexachlorobenzene, polychlorinated biphenyls or polyaromatic hydrocarbons to crops even when very large amounts of sewage biosolids were applied to soil (340 t/ha dry solids) and where resulting polychlorinated biphenyl concentrations were five to seventeen times higher than the levels which would normally be found through normal application of sewage biosolids. The authors concluded that these organic contaminants are not limiting to the use of sewage biosolids in agriculture.

Wild and Jones (1992) determined that crop uptake of polyaromatic hydrocarbons from sewage biosolids into root crops (especially carrots) did not pose a risk to the human food chain even at high concentrations. It has been determined that carrots may become slightly contaminated by polychlorinated biphenyls due to partitioning from soil to the skin of the carrot (Offenbacher 1992), but this occurs only to a limited extent and would be removed by peeling. Since most root crops other than carrots are not good polychlorinated biphenyl accumulators, O'Connor et al (1990) suggest that potential human exposure to polychlorinated biphenyls is very low under the common recommended sewage biosolids application practice for agricultural soils.

Jacobs et al. (1987) found no adverse effects of organic contaminants on the growth of crops have been observed when sewage biosolids are applied to soil at rates according to crop requirements for nutrients.

Ingestion of sewage biosolids amended soil by grazing livestock is considered to be the major route by which organic contaminants can bioaccumulate in the food chain. The lipophilic properties of the ingested organic compounds may result in significant bioconcentration in tissue fat and milk. These organic compounds are not easily eliminated from the body of the animal due to association with the fatty tissues.

Sewage biosolids has been identified as a potential dietary source of organic contaminants in grazing animals. Fries (1982) concluded that the greatest potential problems arising from long term applications of polychlorinated biphenyl contaminated sewage biosolids were through animal ingestion of contaminated soils when grazing. Alcock and Jones (1993) also suggest that significant elevation in the potential transfer of polychlorinated biphenyls to grazing livestock may result from the application of sewage biosolids to grassland.

In Germany, sewage biosolids application to permanent grassland is not permitted due to concerns over the potential for organic contaminants, in particular, polychlorinated biphenyls and dioxins, to bioconcentrate in animal tissues and milk (Sauerbeck and Leschber 1992). This is in direct contrast to the view on polychlorinated biphenyls which is based on risk assessment (US EPA 1989b). Their analysis demonstrated that there was a minimal risk to the human food chain from polychlorinated biphenyls in sewage biosolids which are applied to soils at agronomic rates. Stark and Hall (1992) also concluded that the risk of adverse effects of sewage biosolids-borne organic contaminants on animal health is low at agronomic application rates of sewage biosolids. Some exceptions may occur when sewage biosolids are highly contaminated or if animals graze on the land immediately after sewage biosolids application

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

which results in ingestion of relatively large quantities of sewage biosolids adhering to plant material (Stark and Hall 1992).

The World Health Organization (WHO) Working Group on the Risk to Health of Chemicals in Sewage Sludge Applied to Land (Dean and Suess 1985) concluded that the total human intake of identified organic pollutants from sludge application to land is minor and is unlikely to cause adverse health effects.

9.3 US Perspective

The US EPA in 1983 developed a Task Force to examine the constituents of Municipal Sewage Biosolids and determine which chemicals had the potential to pose a threat to the environment, and thus needed possible regulatory control (US EPA 1989).

This Task Force devised an environmental profile and hazard indices in order to rapidly screen the contaminants so that those most likely to pose a hazard to human health or the environment could be identified (US EPA 1985).

They developed a two tier screening system which allowed for the elimination of contaminants which did not pose a hazard for a specific pathway and for the prioritization of those contaminants that potentially may present a hazard. The first tier involved ranking the contaminants based on their hazard index value for each environmental pathway and deleting those contaminants with hazard index values less than one. This indicated that there was no potential problem under the assumed worst case scenario. In the second tier, contaminants were prioritized based on their incremental values; that is, the portion of the hazard index values that were solely attributable to their presence in sewage biosolids. The incremental values were derived by subtracting the “null” or background values from the total hazard values associated with a contaminant for the specific pathway.

The outcome of this two tiered screening approach did not provide a definitive list of contaminants that the EPA should regulate. Rather, the process identified contaminants of “potential concern” that require further analysis and evaluation. Fifty inorganic and organic contaminants were identified to undergo this process. Of the fifty contaminants, thirty two were evaluated for use in land application. Eighteen of the contaminants were organic and fourteen were inorganic.

In order to screen these compounds, models which utilize basic physical/chemical properties of organic compounds have been developed to predict the environmental fate of organics in soil, since there is a large number of these contaminants present in sewage biosolids. These models have also formed the basis of the screening methodology developed by the USEPA in their Reg 503 (US EPA 1993).

The organic contaminants evaluated in the screening process were as follows:

- Aldrin/Dieldrin
- Heptachlor
- Pentachlorophenol
- Hexachlorobenzene

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

- Hexachlorobutadiene
- Benzo(a)pyrene
- 2-Chloroaniline
- Tricresylphosphate
- Chlordane
- Toxaphene
- Lindane
- Polychlorinated Biphenyls
- Methylene Chloride
- DDT/DDE/DDD
- Trichloroethylene
- Dimethylnitrosamine
- Bis(2-ethylhexyl)phthalate
- Benzo(a)anthracene

Thirteen different hazard indices were developed to evaluate the hazard associated with each of the major environmental pathways (US EPA 1985). The pathways and effects examined included:

- Toxicity to soil biota
- Toxicity to predators of soil biota
- Phytotoxicity
- Plant uptake
- Toxicity to animals resulting from plant consumption
- Toxicity to animals resulting from sludge ingestion
- Human toxicity from plant consumption
- Human toxicity from animal ingestion
- Incidental soil ingestion by humans

The thirteen hazard indices above were calculated for two sewage biosolids contaminant concentrations and for several different cumulative application rates (i.e. 5 tonnes/ha/yr, 50 tonnes/ha/yr, and 500 tonnes/ha (for 100 yrs)) so that for a single index, a determination can be made as to the application rate at which the contaminant becomes a hazard. The lowest sewage biosolids application rate evaluated is the rate that is most applicable to the Ontario context since in Ontario, the prescribed rate is 8 tonnes/ha per 5 years. A hazard index greater than one (1) indicated that these contaminants were of concern and should be carried through to the next level of assessment (US EPA 1985).

The following provides a general description of each of the thirteen indices above:

- Index 1: Index of Soil Concentration Increment - This index indicated the degree of elevation of the contaminant in soil after the sewage biosolids has been applied. The data used in this calculation was the soil background concentration of the contaminant, the contaminant concentration in the sewage biosolids and the sewage biosolids application rate.

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

- Index 2: Index of Soil Biota Toxicity - This index compared the contaminant concentration in the sewage biosolids amended soil with concentrations that were toxic for soil organisms.
- Index 3: Index of Soil Biota Predator Toxicity - This index compared the tissue contaminant concentrations in soil organisms to contaminant concentrations known to be toxic to predators of soil biota (i.e. birds).
- Index 4: Index of Phytotoxicity - This index compared the contaminant concentration in sewage biosolids amended soils with soil concentrations that were toxic to plants.
- Index 5: Index of Plant Concentration Increment Caused by Uptake - This index calculated the incremental amount of the contaminant that was taken into the tissues of plants growing in sewage biosolids amended soils. The plants used varied according to the available data.
- Index 6: Index of Plant Concentration Increment Permitted by Phytotoxicity - This index compared the maximum plant tissue concentration associated with phytotoxicity with the background concentration in the same tissue.
- Index 7: Index of Animal Toxicity Resulting from Plant Consumption - This index calculated the potential hazard to domestic or wild animals which consume crops grown on sewage biosolids amended soils.
- Index 8: Index of Animal Toxicity Resulting from Sludge Ingestion - This index calculated the amount of contaminant in a grazing animal's diet as a result of consuming sewage biosolids amended soil or sludge adhering to forage and compared this with dietary toxic threshold concentrations for grazing animals.
- Index 9: Index of Human Toxicity/Cancer Risk Resulting from Plant Consumption - For non-carcinogens, this index compared the expected dietary intake of a contaminant from crops grown on sewage biosolids amended soils to the acceptable daily intake of that contaminant. For carcinogens, the dietary intake was compared to a daily dietary intake of the contaminant associated with an incremental cancer risk of 10^{-6} . This index assesses toddlers and adults.
- Index 10: Index of Human Toxicity/Cancer Risk Resulting from Consumption of Animal Products Derived From Animals Feeding on Plants - This index calculated the human dietary intake expected to result from contaminant intake by domestic animals given feed produced from crops grown on sewage biosolids amended soil and compares it to the acceptable daily intake of that contaminant for non-carcinogens. For carcinogens, the dietary intake was compared to a daily dietary intake of the contaminant associated with an incremental cancer risk of 10^{-6} . This index assesses toddlers and adults.

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

- Index 11: Index of Human Toxicity/Cancer Risk Resulting from Consumption of Animal Products Derived From Animals Ingesting Soil - This index calculated the human dietary intake expected to result from contaminant intake by domestic animals as a result of consuming sewage biosolids amended soil or sewage biosolids adhering to forage and compares it to the acceptable daily intake of that contaminant for non-carcinogens. For carcinogens, the dietary intake was compared to a daily dietary intake of the contaminant associated with an incremental cancer risk of 10^{-6} . This index assesses toddlers and adults.
- Index 12: Index of Human Toxicity/Cancer Risk Resulting from Soil Ingestion - This index calculated the amount of contaminant ingested by a child or adult as a result of inadvertent or intentional ingestion of sludge amended soil. The amount ingested was compared to the acceptable daily intake or a 10^{-6} risk specific intake level.
- Index 13: Index of Aggregate Human Toxicity/Cancer Risk - This index calculated the aggregate amount of contaminant in the human diet resulting from the pathways described in Indices 9-12. This value was then compared to the acceptable daily intake or a 10^{-6} risk specific intake level.

Groundwater and surface water pathways were not evaluated for land application, since it was determined by the Task Force that these pathways were not of major concern when municipal sewage biosolids was applied using good management practices. This would also be the case in Ontario, if the *Guidelines for Utilization of Biosolids and Other Wastes on Agricultural Land* (MOEE and OMAFRA 1996) were strictly followed.

Table 9.2 summarizes the Task Force's findings of the various hazard indices. It is important to note that for some of the organic contaminants hazard indices could not be calculated because of a lack of data.

TABLE 9.2
Synopsis of Contaminants Where Hazard Indices Were Greater Than One (1)
 (after USEPA 1985)

Hazard Index	Pathway	Contaminant of Concern (HI greater than 1)
Index 1	Soil Concentration Increment	Tricresyl phosphate Bis (2-ethyl hexyl) phthalate 2 chloro aniline Methylene chloride Trichloroethylene Hexachlorobutadiene
Index 2	Soil Biota Toxicity	None identified
Index 3	Soil Biota Predator Toxicity	Aldrin/Dieldrin Hexachlorobenzene
Index 4	Phytotoxicity	None identified
Index 5	Plant Concentration Increment Caused by Uptake	Polychlorinated Biphenyls
Index 6	Plant Concentration Increment Permitted by Phytotoxicity	Not evaluated
Index 7	Animal Toxicity Resulting from Plant Consumption	None identified

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.2 – cont'd
Synopsis of Contaminants Where Hazard Indices Were Greater Than One (1)
(after USEPA 1985)

Hazard Index	Pathway	Contaminant of Concern (HI greater than 1)
Index 8	Animal Toxicity Resulting from Sludge Ingestion	None identified
Index 9	Human Toxicity/Cancer Risk Resulting from Plant Consumption	Polychlorinated Biphenyls Hexachlorobenzene Chlordane Benzo(a)pyrene Aldrin/Dieldrin Toxaphene DDT/DDD/DDE Heptachlor
Index 10	Human Toxicity/Cancer Risk Resulting from Consumption of Animal Products Derived From Animals Feeding on Plants	Polychlorinated Biphenyls Toxaphene Hexachlorobenzene Aldrin/Dieldrin Chlordane DDT/DDD/DDE Heptachlor
Index 11	Human Toxicity/Cancer Risk Resulting from Consumption of Animal Products Derived From Animals Ingesting Soil	Polychlorinated Biphenyls Aldrin/Dieldrin Toxaphene Hexachlorobenzene Chlordane Heptachlor Lindane DDT/DDD/DDE Hexachlorobutadiene
Index 12	Human Toxicity/Cancer Risk Resulting from Soil Ingestion	Aldrin/Dieldrin Polychlorinated Biphenyls Benzo(a)pyrene Lindane Toxaphene Chlordane Heptachlor Hexachlorobenzene Hexachlorobutadiene
Index 13	Aggregate Human Toxicity/Cancer Risk	Polychlorinated Biphenyls Aldrin Hexachlorobenzene Toxaphene Chlordane Heptachlor DDT/DDD/DDE

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

The contaminants in Table 9.2 were carried forward by the Task Force to the second step of the evaluation; incremental ranking. As mentioned previously, the purpose of this step was to evaluate what portion of the total hazard associated with a contaminant for a particular pathway was attributable to sewage biosolids. The incremental value was determined by subtracting the null or background value for the worst case scenario from the total hazard index value for the worst case scenario. For example, for Index 3, Aldrin/Dieldrin and Hexachlorobenzene were identified as being of concern based on the pathways analysis; however, when adjusting the contaminant concentration to remove background, only Aldrin/Dieldrin are then of concern. Table 9.3 summarizes the results.

TABLE 9.3
Summary of Contaminants of Concern and Hazard Indices Associated with Incremental Addition to Soil
 (after USEPA 1985)

Hazard Index	Pathway	Contaminant of Concern (HI greater than 1)
Index 2	Soil Biota Toxicity	None identified
Index 3	Soil Biota Predator Toxicity	Aldrin/Dieldrin (1.5)
Index 4	Phytotoxicity	None identified
Index 7	Animal Toxicity Resulting from Plant Consumption	None identified
Index 8	Animal Toxicity Resulting from Sludge Ingestion	None identified
Index 9	Human Toxicity/Cancer Risk Resulting from Plant Consumption	Polychlorinated Biphenyls (14953) Hexachlorobenzene (4295) Chlordane (3100) Benzo(a)pyrene (2860) Aldrin/Dieldrin (1300) Toxaphene (1245) Heptachlor (15)
Index 10	Human Toxicity/Cancer Risk Resulting from Consumption of Animal Products Derived From Animals Feeding on Plants	Polychlorinated Biphenyls (64953) Toxaphene (1345) Hexachlorobenzene(1095) Aldrin/Dieldrin (100) Chlordane (180) DDT (151) Heptachlor (7)
Index 11	Human Toxicity/Cancer Risk Resulting from Consumption of Animal Products Derived From Animals Ingesting Soil	Polychlorinated Biphenyls (33947) Aldrin/Dieldrin (9090) Toxaphene (1845) Hexachlorobenzene (814) Chlordane (448) Heptachlor (396) Lindane (10) DDT (117) Hexachlorobutadiene (130)

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.3 – cont'd
Summary of Contaminants of Concern and Hazard Indices Associated with Incremental Addition to Soil
 (after US EPA 1985)

Hazard Index	Pathway	Contaminant of Concern (HI greater than 1)
Index 12	Human Toxicity/Cancer Risk Resulting from Soil Ingestion	Aldrin/Dieldrin (40) Polychlorinated Biphenyls (171) Benzo(a)pyrene (39) Toxaphene (12) Chlordane (33) Hexachlorobenzene (6.5) Hexachlorobutadiene (8.9)
Index 13	Aggregate Human Toxicity/Cancer Risk	Polychlorinated Biphenyls (109937) Aldrin/Dieldrin (11090) Hexachlorobenzene (6200) Toxaphene (4445) Chlordane (3900) Heptachlor (425) DDT (317)

Based on the results of the two tier screening process (described in Tables 9.2 and 9.3), ten of the eighteen identified organic compounds required further analysis since they could potentially pose a hazard in at least one of the 10 pathways evaluated. These contaminants are:

- Aldrin/Dieldrin
- Polychlorinated Biphenyls
- Benzo(a)pyrene
- Lindane
- Toxaphene
- Chlordane
- Heptachlor
- Hexachlorobenzene
- Hexachlorobutadiene
- DDT

These contaminants can be combined into the following groupings: pesticides such as aldrin/dieldrin, lindane, toxaphene, chlordane, heptachlor and DDT; polyaromatic hydrocarbons such as benzo(a)pyrene; polychlorinated biphenyls; and chlorobenzenes.

Polyaromatic hydrocarbons and chlorobenzenes had Hazard Index values less than one and thus do not cause adverse ecological or human health impacts (US EPA 1985).

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

In a review of 20 years of land application research with sewage biosolids, Chaney (1990) stated that except for the peel of carrots, there is little evidence for plant uptake of polychlorinated biphenyls from sewage biosolids treated soils. Transfer to the food chain is mainly through direct ingestion of sewage biosolids by cattle, this transfer is so low that sewage biosolids containing a few mg/kg polychlorinated biphenyls could be applied to agricultural soils without causing any adverse effects. Consequently the regulation of polychlorinated biphenyls in sewage biosolids was not considered necessary in the Final Part 503 due to the very low risk to the human food chain (US EPA 1993). As also noted in Section 9.2, there was a minimal risk to the human food chain from polychlorinated biphenyls in that is sewage biosolids applied to soils at agronomic rates.

In terms of the pesticides that have been highlighted for further evaluation, it has been found that the concentration loadings of pesticides in sewage biosolids applied to land are substantially lower than the pesticide concentrations applied in farming practices and as a consequence should present little quantitative risk to the environment in comparison (Jones and Wild 1991). In addition, pesticides associated with sewage biosolids tend to be less bioavailable than agrochemicals since they are strongly adsorbed to organic matter in the biosolids and soils.

9.4 Canadian Perspective

9.4.1 Organic Contaminants in Sewage Biosolids

In 1987/88, the Ministry of Environment carried out a comprehensive monitoring study to determine the concentrations of conventional parameters, metals and a wide range of trace organics in raw sewage, final effluent, raw solids and digested biosolids in 37 municipal sewage treatment plants (MOE, 1988). Trace organics monitored included volatile organics (VOCs), base-neutral and acid extractable organics (BN&As), organochlorine pesticides (OCs), polychlorinated biphenyls and dioxins and furans. Similarly, the Wastewater Technology Center conducted detailed studies in 1980-1985 and then in 1993-1994 to monitor a wide range of organic contaminants in several Canadian municipal sewage biosolids (Webber & Lesage 1988; Webber and Nichols 1995). Sewage biosolids from British Columbia, Alberta, Saskatchewan, Manitoba and Ontario were analyzed for the compounds listed in Table 9.4 in addition to polychlorinated biphenyls and dioxins and furans.

A summary of the above three data sets for organic contaminants in sewage biosolids is presented in Table 9.5. The 1980 – 1985 and 1987 data generally were single measurements whereas the 1993 – 1994 data were means of 6 measurements. The occurrences (in %), and concentration ranges and median values for the data sets can be compared directly but the geometric mean and median values cannot. It is evident from the data that the maximum concentrations of volatiles were larger in 1987 than in 1993-1994 but respective geometric mean and median values were similar. Likewise the maximum concentrations of several polyaromatic hydrocarbons, phthalate esters and other compounds in 1980-1985 and 1987 were larger than in 1993-1994 but the respective geometric mean and median values were similar. Thus, it appears that concentrations of these contaminants in a large majority of Canadian sewage biosolids remained approximately constant during the 14 year period 1980-1994.

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.4
Volatile Organic (VOC) And Base Neutral
And Acid Extractable (BN&A) Compounds
 (after Webber and Nichols (1995)).

VOC	BN&A	
1,1,1-trichloroethane	<u>Polynuclear Aromatic Hydrocarbons (PAH)</u>	<u>Nitrosamines</u>
1,1,2,2-tetrachloroethane		N-Nitrosodiphenylamine
1,1,2-trichloroethane	Naphthalene	N-Nitrosodi-n-propylamine
1,1-dichloroethane	Acenaphthylene	
1,1-dichloroethylene	Acenaphthene	<u>Phthalate Esters</u>
1,2,4-trimethylbenzene	Fluorene	Dimethylphthalate
1,2-dibromoethane	Phenanthrene	Diethylphthalate
1,2-dichlorobenzene	Anthracene	Di-n-butylphthalate
1,2-dichloroethane	Fluoranthene	Butylbenzylphthalate
1,2-dichloropropane	Pyrene	Bis-2-ethylhexylphthalate
1,2-diethylbenzene	Benzo(a)anthracene	Di-n-octylphthalate
1,3,5-trimethylbenzene	Chrysene	
1,3-dichlorobenzene	Benzo(b)fluoranthene	<u>Acid Group</u>
1,3-diethylbenzene	Benzo(k)fluoranthene	Phenol
1,4-dichlorobenzene	Benzo(a)pyrene	2-Chlorophenol
1,4,-diethylbenzene	Indeno(1,2,3-c,d)pyrene	2-Nitrophenol
2-ethyltoluene	Dibenzo(a,h)anthracene	2,4-Dimethylphenol
3-ethyltoluene	Benzo(g,h,i)perylene	2,4-Dichlorophenol
4-ethyltoluene		p-Chloro-m-cresol
Benzene	<u>Haloethers</u>	2,4,6-Trichlorophenol
Bromodichloromethane	Bis-2-chloroethylether	2,4-Dinitrophenol
Bromoform	Bis-2-chloroisopropylether	4-Nitrophenol
Chlorobenzene	Bis-2-chloroethoxymethane	4,6-Dinitro-o-cresol
Chlorodibromomethane	4-Bromophenylphenylether	Pentachlorophenol
Chloroform	4-Chlorophenylphenylether	
Cis-1,2-dichloroethylene		<u>Other Compounds</u>
Cumene	<u>Chlorinated Benzenes</u>	Nitrobenzene
Cyclohexane	1,3-Dichlorobenzene	Isophorone
Dibromomethane	1,4-Dichlorobenzene	Hexachloroethane
Dichloromethane	1,2-Dichlorobenzene	Hexachlorobutadiene
Ethylbenzene	1,2,4-Trichlorobenzene	Hexachlorocyclopentadiene
Hexane	Hexachlorobenzene	2-Chloronaphthalene
Nonane		2,6-Dinitrotoluene
O-xylene		2,4-Dinitrotoluene
Octane		1,2-Diphenylhydrazine
Propylbenzene		3,3'-Dichlorobenzidine
Styrene		
Tetrachloroethylene		
Tetrachloromethane		
Toluene		
Trans-1,2-dichloroethylene		
Trichloroethylene		
M/p-xylene		

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.5
Organic Contaminants in Canadian Sludges
(after Campbell and Webber 1994)

Contaminant	15 Canadian Biosolids 1980 – 1985			34 Ontario Stabilized Biosolids 1987			11 Canadian Biosolids 1993 – 1994	
	Occurrence	Concentration	Median	Occurrence	Concentration	Geo. Mean ^c	Range	Mean Concentration ^a
	%	mg/kg dw		%	mg/kg dw			
Volatiles								
m- & p-Xylenes				32.4	0.6 - 1417	0.8	0.4 - 3.5	1.3
Ethylbenzene				32.4	0.2 - 500	0.6	0.02 - 0.6	0.4
o-Xylene				32.4	0.3 - 37	0.5	0.02 - 1.5	0.5
Chloroform				35.3	T - 13	0.4	0.1 - 0.2	0.1
Tetrachloroethylene				5.9	2.8 - 3.4	0.3	0.03 - 0.2	0.1
Hexanol				2.9	144	12		
1,1,1-Trichloroethane				2.9	2.3	0.2	W	
Chlorodibromoethane				2.9	6.6	0.3	W	
Trichloroethylene				2.9	0.94	0.3	0.1 - 0.4	0.2
Toluene							0.2 - 42	1.3
1,2 Dichlorobenzene	27	T - 1.6	0.5				T - 0.4	0.1
1,4 Dichlorobenzene	33	T - 1.5	0.3	5.9	0.2 - 2.6	0.3	T - 0.9	0.5
Polynuclear Aromatic Hydrocarbons (PAHs)								
Acenaphthene	33	T - <3	T				0.1 - 1.4	0.6
Acenaphthylene	27	T - 5	T	2.9	6.1	1.9	W - 3.4	0.1
Anthracene	20	T - 32	1	2.9	4.0	1.3	T - 1.7	0.2
Phenanthrene	53	T - 36	2	26.5	2.9 - 28	2.2	0.9 - 11	2.4
Benzo(a)anthracene	7	<0.5		5.9	6.7 - 15	1.4	0.3 - 19 ^d	1.3 ^d
Chrysene	27	T - 23	1.5	11.8	9.7 - 14	1.5		
Benzo(b)fluoranthene	7	<0.5					0.4 - 4.9 ^e	0.8 ^e

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.5 – cont'd
Organic Contaminants in Canadian Sludges
(after Campbell and Webber 1994)

Contaminant	15 Canadian Biosolids 1980 - 1985			34 Ontario Stabilized Biosolids 1987			11 Canadian Biosolids 1993 – 1994		
	Occurrence	Concentration	Median	Occurrence	Concentration	Geo.Mean ^c	Mean Concentration ^a	Range	Median
	%	mg/kg dw		%	mg/kg dw		mg/kg dw		
Benzo(k)fluoranthene	13	<0.5-9	5	2.9	20	1.3			
Benzo(g,h,i)fluoranthene	13	T-0.3	0.2				0.1-5.2	0.2	
Benzo(a)pyrene	13	4-7.2	5.6	2.9	11	1.3	0.1-6.8	0.4	
Dibenzo(ah)anthracene	7	<13					W-3.9	T	
Fluoranthene	53	T-33	2	8.8	8.0-22	1.3	0.6-5.0	1.0	
Fluorene	60	T-3	2	8.8	2.6-4.9	1.3	0.2-2.5	0.7	
Indenol (1,2,3-cd)pyrene	7	7					T-1.8	0.2	
Naphthalene	60	T-5.8	1	14.7	2.4-26	1.5	0.5-13	1.8	
Pyrene	67	T-29	3.5	8.8	9.2-19	2.1	0.6-14	1.3	
Total PAHs			24			17		11	
Total Polychlorinated Biphenyls							W-0.3	W	
Phthalate Esters									
Butylbenzylphthalate	27	T-5.7	2.5	17.7	1.7-914	1.9	0.3-10	3.2	
Di-n-butylphthalate Diethylphthalate	80	0.2-430	10				3.6-12	7.5	
Diethylphthalate	40	T-14	2.5				T-0.6	0.2	
Dimethylphthalate	27	T-2	T				W-0.1	W	
Di-n-octylphthalate	60	T-115	7				1.2-8.1	4.1	
Bis-2ethylhexylphthalate	93	3-215	80				64-244	161	

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.5 – cont'd
Organic Contaminants in Canadian Sludges
 (after Campbell and Webber 1994)

Contaminant	15 Canadian Biosolids 1980 – 1985		34 Ontario Stabilized Biosolids 1987		11 Canadian Biosolids 1993 - 1994	
	Occurrence %	Concentration mg/kg dw	Occurrence %	Concentration mg/kg dw	Mean Concentration ^a mg/kg dw	Median
	Range ^b	Median	Range	Geo.Mean ^c	Range	Median
Other Components						
Nitrobenzene	27	T - 9	8.8	6.4 - 17	1.4	W - 5
Phenol	33	T - 68	8.8	16 - 31	2.1	0.7 - 9
Hexachlorobenzene	7	4	56	0.001 - 0.12	0.01	
Dioxins and Furans						
Octachlorodibenzodioxin			65	0.6 - 304	7.1	1.6 - 40
Heptachlorodibenzodioxin			24	1.4 - 73	5.0	0.2 - 7.1
Hexachlorodibenzodioxin			5.9	1.1 - 3.8	2.4	0.02-0.5
Pentachlorodibenzodioxin			2.9	1.3	3.2	W - 0.01
Pentachlorodibenzofuran			2.9	1.6	2.5	W - 0.05

a: Means of 6 observations, one per month during Sept. 1993 – Feb. 1994

b: Single values indicate only one measurement above minimum detection limit

c: Geometric mean- not detected was replaced with one-half minimum detection limit

d: Sum of benzo(a)anthracene and chrysene

e: Sum of benzo(b&k)fluoranthene

W: Compound not detected

T: Compound detected but not quantified

Blank: Information not available

**9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS
IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd**

Table 9.6 (Webber et. al. 1997) shows a summary of the median and highest concentrations of organic contaminants found in municipal sewage biosolids and composts. From this table, it can be seen that toluene and xylene were found in concentrations exceeding 1 mg/kg. These VOCs by nature of their physical/chemical properties tend to volatilize, degrade and disappear rapidly from soil amended with sewage biosolids (Webber et. al. 1997) and thus should not pose any significant impacts on the environment or human health. In addition, Table 9.6 also presents background concentrations of these contaminants in Ontario agricultural soils. Contaminants such as volatiles and phthalate esters have been shown to degrade in soils (O'Connor et. al. 1991) and thus are not persistent.

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

TABLE 9.6
Estimated Permissible Sewage Biosolids Loadings to Agricultural Land In Ontario

Parameter	Observed Concentrations in Biosolids (mg/kg dry wt.) (Note c)		Concentrations in Agricultural Soils (mg/kg dry wt.) (Note a)			Loading (kg/ha) of Organics to Reach Maximum Soil Concentrations (Note b)		Maximum No. of Times Biosolids Can Be Applied Using Biosolids with Highest Organic Concentrations to not exceed (Note f)		Maximum No. of Times Biosolids Can Be Applied Using Biosolids with Median Organic Concentrations to not exceed (Note f)	
	Highest	Median	Mean	Background Based Criteria (Table F)	Effects Based Criteria (Table A)	Based on Background Criteria	Based on Effects Criteria	Background Criteria	Effects Criteria	Background Criteria	Effects Criteria
VOC											
1,2-dichlorobenzene	0.42	0.1	0.000003	0.002	0.88	0.003994	1.75994	1	524	5	2200
1,4-dichlorobenzene	0.87	0.42	0.000008	0.002	0.32	0.003984	0.639984	1	92	1	190
Benzene	0.48	0.06	0.000009	0.002	0.24	0.003982	0.099982	1	125	8	1000
Chlorobenzene	0.2	0.06	0.000005	0.002	2.4	0.00399	4.79999	2	3000	8	10000
Chloroform	0.22	0.12	0.0004	0.006	0.13	0.0112	0.2592	6	147	12	270
Ethylbenzene	0.62	0.36	0.00009	0.002	0.28	0.00382	0.99982	1	113	1	194
O/m/p-xylene	5.1	1.5	0.00025	0.002	25	0.0035	1.9995	0.1	1225	0.3	4167
Tetrachlorethylene	0.23	0.09	0.0002	0.002	0.45	0.0036	0.8996	2	489	5	1249
Toluene	42	1.1	0.0032	0.002	1	0.00336	1.99936	0.01	12	0.4	477
Trichloroethylene	0.38	0.23	0.000028	0.004	1.1	0.007944	0.779944	3	724	4	1196
BN & AS											
2-Chlorophenol	0.15	0.06	0.0014	0.1	0.1	0.172	0.172	143	143	358	358
Acenaphthene	1.6	0.53	0.003	0.05	15	0.094	29.994	7	2343	22	7074
Acenaphthylene	3.4	0.05	0.003	0.23	100	0.454	199.994	6	7353	385	499985
Anthracene	1.7	0.23	0.003	0.05	28	0.094	55.994	7	4117	51	30432
Benzo(a)anthracene & Chrysene	19	1.1	0.003	0.28	6.6	0.554	26.394	1	87	22	1499
Benzo(a)pyrene	6.8	0.33	0.00	0.1	1.2	0.194	2.394	4	44	73	907
Benzo(b&k)Flouranthene	4.9	0.76	0.003	0.05	12	0.094	23.994	2	612	15	3946

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd
TABLE 9.6 – cont'd
Estimated Permissible Sewage Biosolids Loadings to Agricultural Land In Ontario

Parameter	Observed Concentrations in Biosolids (mg/kg dry wt.) (Note c)		Concentrations in Agricultural Soils (mg/kg dry wt.) (Note a)		Loading (kg/ha) of Organics to Reach Maximum Soil Concentrations (Note b)		Maximum No. of Times Biosolids Can Be Applied Using Highest Organic Concentrations to not exceed (Note f)		Maximum No. of Times Biosolids Can Be Applied Using Biosolids with Median Organic Concentrations to not exceed (Note f)		
	Highest	Median	Mean	Background Based Criteria (Table E)	Effects Based Criteria (Table A)	Based on Background Criteria	Based on Effects Criteria	Background Criteria	Effects Criteria	Background Criteria	Effects Criteria
Benzo(g,h,i)perylene	5.2	0.24	0.005	0.2	40	0.39	69.99	9	1923	203	41661
Dibenzo(a,h)anthracene	3.9	0.03	0.005	0.15	1.2	0.29	2.39	9	77	1208	9958
Flouranthene	5	1.04	0.003	0.24	40	0.474	79.994	12	2000	57	9615
Fluorene	2.5	0.63	0.003	0.05	340	0.094	679.994	5	34000	19	134919
Indeno (1,2,3-c,d)pyrene	1.8	0.15	0.003	0.11	12	0.214	23.994	15	1666	178	19995
Naphthalene	13	2	0.003	0.05	4.6	0.094	9.194	1	88	6	575
Phenanthrene	11	2.3	0.003	0.19	40	0.374	79.994	4	909	20	4348
Phenol	9.4	2	0.005	0.1	40	0.19	79.99	3	1064	12	4999
Pvrene	14	1.2	0.003	0.19	250	0.374	2.594	3	4464	39	52083
OCs & Polychlorinated Biphenyls											
Hexachlorobenzene	0.033	0.001	(Note d)	(Note d)	0.46						
P,P-DDE (Note)	0.013	W	0.003	0.12	1.6	0.234	3.194	2250	30712		
Total Polychlorinated Biphenyls	0.28	W	0.015	0.3	0.5	0.57	0.97	254	433		
Total TEQ for (PCDD & PCDF)	0.00012	0.00002	0.0000017	0.000007	0.00001	0.0000106	0.0000166	11	17	66	104

Notes:

- a. Source: Guidelines for Use at Contaminated Sites in Ontario; 1997 and Ontario Typical Range of Chemical Parameters in soil, Vegetation, Moss Bags and Snow, 1993.
- b. Loadings calculated assuming that biosolids are applied to the surface 15 cm of soil which weighs 2×10^6 kg (2000 tonnes), maximum organics loadings and number of sewage biosolids applications permissible are conservative estimates based on no biodegradation of organics in soil.
- c. Source: Canadian Biosolids 1993-1994, data from Table 9.5.
- d. Data not available
- e. The Mean Soil and Background Based Criteria were not available for p,p-DDE and the values for DDT were used.
- f. Calculated assuming 8 tonnes per hectare of biosolids dry matter per application.
- W. No detection

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

Large concentrations of organochlorine pesticides are not expected to occur in Canadian municipal sewage biosolids because production and use of these compounds with the exception of endosulfan has been banned in Canada since the mid-1970's. Most of these contaminants did not exceed detection limits (Webber and Nichols 1995).

Large concentrations of polychlorinated biphenyls also are not expected to occur in Canadian municipal sewage biosolids since they too have been banned since the mid-1970's. From Table 9.6 it can be seen that the highest observed total polychlorinated biphenyl concentration in municipal sewage biosolids is approximately 0.28 mg/kg dry weight.

The Webber and Nichols (1995) study represents the only concerted effort to monitor dioxins and furans in a cross-section of Canadian municipal sewage biosolids and sewage biosolids composts. Mean total dioxin plus furan concentrations in Canadian sludges ranged from 0.002 to 0.036 mg/kg dry weight with a median value of 0.006 mg/kg. Mean total TEQ concentrations ranged from 0.000005 to 0.00012 mg/kg dry weight with a median value of 0.000025 mg/kg. Dioxins and furans were observed in approximately 37% of all Ontario sewage biosolids (MOE 1988). Geometric mean concentrations ranged from 0.0025 to 0.0072 mg/kg dry weight. These values are higher than those reported by Webber and Nichols (1995) but are similar or lower than concentrations found in the USA, UK, Sweden and the Netherlands (Webber and Nichols 1995). In 1996, the original study was expanded to obtain information on Eastern Canadian sludges. Four sludges were examined in Ontario (from Toronto and Halton) and two from New Brunswick (Webber and Bedford 1996). They concluded that if these sludges are applied to the land according to guidelines, no significant risk to agriculture and the environment will occur.

Table 9.6 estimates the maximum loading of organics that can be applied to agricultural land and the maximum number of times biosolids can be repeatedly applied to an agricultural field without exceeding the "Effects Based" and "Background Based" criteria for agricultural land. The estimates in Table 9.6 are conservative since it assumes that there is no biodegradation or volatilization of organics in the soil. Table 9.6 indicates that biosolids containing the median concentrations reported by Webber and Nicols (1995) can be repeatedly applied to the same field for more than 66 times (or 330 years) without exceeding the "Effects Based" criteria. Except for volatile organics, the same biosolids can be repeatedly applied to an agricultural for 6 times (30 years) without exceeding the "Background Based" criteria (restricted by naphthalene). Due to their volatile nature, volatile organics are not expected to remain in the soil very long. Their presence in biosolids is therefore, not a concern for the land application of biosolids.

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

9.4.2 Organic Contaminants in Agricultural Land

Webber and Wang (1995) conducted a study to determine the concentration of industrial organic compounds in selected Canadian agricultural soils and to assess the potential for land application of municipal sludges to cause significant polyaromatic hydrocarbons, organochlorine pesticides and polychlorinated biphenyl contamination of agricultural land. Twenty-four benchmark soils (from the Agriculture and AgriFood Canada Soil Quality Evaluation Program) and six intensively cropped southern Ontario soils were studied. All the soils exhibited low concentrations of most organic compounds as seen in Table 9.7.

TABLE 9.7
Comparison of Organic Contaminant Concentrations in Canadian Agricultural Soils and Ontario Soil Background Concentrations (mg/kg dry weight)
 (after Webber and Wang 1995)

Compound	Canadian Agricultural Soils	Sludge Amended Field	Silt loam soil and 200 tonnes/ha dw Sludge	Ontario Soil Background
PAHs	ND – 0.13	ND – 0.34	0.02 – 1.4	0.05 – 0.24
Naphthalene	ND – 1.2	0.14	0.05	0.05
Phenanthrene	ND – 0.48	0.22	0.17	0.19
Haloethers	ND	NA	NA	NA
Chlorinated benzenes	ND – 0.29	NA	NA	0.002
Phthalate esters	ND – 3.1	NA	NA	NA
Bis-2-ethylhexylphthalate	ND – 2.7			
Acid group Phenol	ND – 0.53 ND – 0.92	NA	NA	0.1 – 0.2 0.1
OC Pesticides	ND – 0.077	0.029	0.0006 – 0.004	0.05
Total DDT	ND – 72	NA	NA	0.12
Total Polychlorinated Biphenyls	ND – 0.17	0.51	0.14	0.3

Note: ND – not detected;
 NA – analyses not conducted

According to Webber and Wang (1995) the total DDT concentrations in the soil may cause some concern. They also indicated that this concentration of DDT is due to the use of agrochemicals and not the use of land application of sewage biosolids and emphasizes the persistence of this compound given that it has been banned from use since the 1970's. A laboratory study (Webber and Wang 1995) indicated that soils treated with sewage biosolids according to the recommended practice exhibited minor increases in polyaromatic hydrocarbon, organochlorine pesticides and polychlorinated biphenyl concentrations. They concluded that there is no risk to human health or the environment from these industrial organic compounds, in soils that have had sewage biosolids application according to recommended practice (see Table 9.7).

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

9.5 Regulations

The European Union is currently revising their regulation for land application of sewage biosolids (Draft EC Council Directive 2000). They propose concentration limits for organic compounds but no rationale is provided for the development of their numbers. If the proposed limits of these contaminants are compared to maximum Ontario sewage biosolids concentrations of these contaminants, then polyaromatic hydrocarbons, di (2-ethylhexyl) phthalate and nonylphenols are above these criteria. (Table 9.8) (Webber & Nichols 1995).

TABLE 9.8
Comparison of Proposed EU Limit Values and Maximum Observed Concentrations in Ontario Sewage Biosolids (mg/kg dry weight)
 (after Webber & Nichols 1995, WTI 1998)

Organic Compounds	Proposed EU Limit values	Maximum Ontario Sewage Biosolids Concentrations
Halogenated Organic Compounds	500	222 (Toluene)
Linear alkylbenzene sulphonates	2600	No Data
Bis-2-ethylhexylphthalate	100	440
Nonylphenol and Nonylphenol ethoxylates	50	700 (3 observations)
Polyaromatic Hydrocarbons	6	32 (Total PAHs)
Polychlorinated Biphenyls	0.8	0.38
Dioxins and Furans (based on TEQ)	0.0001	0.0001

9.6 Concerns Raised at the Stakeholder Workshop

During the Stakeholder Workshop particular concerns were raised re: specific groups of organic compounds namely, dioxins and furans, endocrine disruptors (mainly alkylphenol ethoxylates), and surfactants (mainly linear alkylbenzene sulphonates). Alkylphenol ethoxylates and linear alkylbenzene sulphonates were identified since in general, they represent about 94 % of the organic contaminant loading in sewage biosolids (Smith 1996). Dioxins and furans occur at very low concentrations in sewage biosolids but they are persistent and bioaccumulate and may represent a health risk to animals and humans (Wild et al. 1994).

Based on the discussion at the Stakeholder Workshop these parameters were identified for further literature research which is included in sections 10, 11 and 12 of this report.

9.7 Findings from the Literature Review

1. The WHO Working Group on the Risk to Health of Chemicals in Sewage Sludge Applied to Land has concluded that “the total human intake of identified organic pollutants from sludge application to land is minor and is unlikely to cause adverse health effects” (Dean and Suess 1985).

9. LITERATURE REVIEW OF THE FATE AND EFFECTS OF ORGANIC COMPOUNDS IN SEWAGE BIOSOLIDS APPLIED TO AGRICULTURAL LAND – cont'd

2. Based on the concerns raised at the Stakeholder Meeting, sections 10, 11 and 12 of this report provide a literature review of specific groups of organic compounds namely: dioxins and furans, endocrine disruptors (mainly alkylphenol ethoxylates), and surfactants (mainly linear alkylbenzene sulphonates).

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