CANADIAN ENVIRONMENTAL QUALITY GUIDELINES FOR MERCURY

C. GAUDET[1], S. LINGARD[2], P. CURETON[1], K. KEENLEYSIDE[1], S. SMITH[1] AND G. RAJU[3]

[1]*Evaluation and Interpretation Branch, Environment Canada, Ottawa, Ontario, K1A 0H3.* [2]*Lingard Consulting, Ottawa, Ontario, K1K 2K6.* [3] *Raju Environmental Services, Gloucester, Ontario, K1C 4Z6.*

Abstract: CCME Canadian Environmental Quality Guidelines for mercury have been recommended or are under development for soil, water and sediments. These guidelines provide nationally consistent benchmarks for environmental quality across Canada and are intended as decision support tools in protecting and sustaining aquatic and terrestrial ecosystems in Canada and the beneficial uses they support. A Canadian water quality guideline for protection of aquatic life was recommended in 1987 as 0.1 $\mu g{\cdot}L^{-1}$. Currently, mercury guidelines for soils and sediments are under development. Preliminary calculations indicate that interim marine and freshwater sediment guidelines for the protection of aquatic life will both be 0.14 $mg{\cdot}kg^{-1}$; and that soil quality guidelines will be 2.0 $mg{\cdot}kg^{-1}$ (agricultural and residential land uses), and 30.0 $mg{\cdot}kg^{-1}$ (commercial and industrial land uses). Final recommended values are subject to change pending final approval by the Canadian Council of Ministers of the Environment.

1. Introduction

Goals for environmental quality have evolved from simple human use protection goals such as "drinkable, swimmable" water, to protection of the "ecosystem", including the human and non-human components. In response to an increasing public concern that chemical substances entering the environment were a major factor placing ecosystems at risk, the Canadian Council of Minister's of the Environment (CCME), undertook to develop nationally consistent benchmarks for environmental quality in Canada that would be protective of the long-term sustainable use of Canadian ecosystems. An initial emphasis on water quality lead to the publication of the Canadian Water Quality Guidelines in 1987 (CCREM 1987). National water quality guidelines were developed to protect and sustain not only the important human uses such as drinking water, but also freshwater life, livestock water and irrigation water. More recently, the guidelines have been expanded to encompass other important ecosystem components, mainly marine water quality, marine and freshwater sediment quality, tissue residue guidelines for protection of wildlife consumers, and soil quality guidelines.

The use and interpretation of the terms *criteria, guidelines, objectives* and *standards* vary among different agencies and countries. Environment Canada has generally adopted the term guideline as being a numerical limit or narrative statement recommended to support and maintain designated uses of the environment. This term is used interchangeably with the term criteria in describing the soil quality guidelines.

Canadian Environmental Quality Guidelines (EQGs) are based on a published national protocol which ensures consistent decision-making and quality of scientific data

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in development of national guidelines. Though protocols vary dependent on the specific exposure pathways and receptors of concern for a particular land or water use, the basic philosophy underlying guideline development is the same, i.e. to ensure that levels of contaminants in the environment pose no risk to the potential or existing range of biota, functions and interactions integral to sustaining the integrity of the ecosystem which supports a specified land or water use. To achieve this broad-based protection of a complex system, guidelines are based on conservative assumptions, such as protection of sensitive species and life stages.

Collectively, Canadian EQGs provide an important framework for protecting aquatic and terrestrial ecosystems across Canada and sustaining the many beneficial uses they support. EQGs have broad application in environmental evaluation and management. For example, they can serve as the basis for the development of objectives for the assessment and remediation of contaminated sites, as screening tools for assessing environmental quality and interpreting the significance of contaminant levels in environmental media, as goals for national and regional toxics management or rehabilitation programs, and as environmental benchmarks for international negotiations on emission reductions and trade agreements. These guidelines also play an important role in the Canadian Environmental Protection Act (CEPA) which was proclaimed in 1988 and is the basis of the federal government environmental protection legislation. Under CEPA Part 1, the Minister was given the authority to formulate environmental quality guidelines and objectives.

This paper outlines the current derivation procedures for Canadian Water Quality Guidelines, Soil Quality Criteria, and Sediment Quality Guidelines, and presents the proposed guidelines for mercury. To provide the context for development of mercury guidelines, this paper begins with a brief overview of the major sources, fate, and behaviour of mercury in the Canadian environment.

2. Mercury in the Canadian Environment

Mercury is relatively ubiquitous in the environment and is found in almost every environmental compartment including air, volcanic gases, fresh water, sea water, soils, mineral ores, lake and river sediments, and living organisms. Sources of mercury are both natural, via weathering and degassing of the earth's crust, and anthropogenic (D'Itri 1990).

In Canada, anthropogenic release of mercury to the environment has been estimated at approximately 31 tonnes annually (Jaques 1987). The major form of this release is as atmospheric emissions. Although mercury is no longer mined in Canada, the major source of mercury emissions is base metal recovery (45.2% of the total). Power generation and the combustion of coal, petroleum products, wood and natural gas contribute the second largest source of emissions (25.8% of the total).

In Canada, the general terrestrial concentrations of mercury are in the range of 0.02 to 0.15 $mg{\cdot}kg^{-1}$, with an average of 0.05 $mg{\cdot}kg^{-1}$ (Jonasson and Boyle 1972; McKeague and Kloosterman 1974; Environment Canada 1979) with elevated levels adjacent to anthropogenic point sources such as mining sites, reaching several hundred $mg{\cdot}kg^{-1}$

(Jonasson and Boyle 1972). Naturally elevated levels have also been found in British Columbia due to cinnibar deposits and in areas of Quebec and Ontario near areas of gold, copper or zinc mineralization (Environment Canada 1979). Flooding of terrestrial environments to form hydroelectric reservoirs results in the formation of methylmercury compounds in soil which become available to organisms within the soil, sediment, and water column (Louchouarn *et al.* 1993).

Sediments are often a major sink for mercury compounds in aquatic environments where it can be rendered virtually inactive (deep sediments) or converted to methylmercury, principally by sulphur-reducing bacteria (Bigham and Henry 1993; ENVIRO TIPS 1984). Mercury levels in Canadian lakes are generally below 0.3 $\mu g \cdot g^{-1}$ except in the Flin Flon area in Manitoba where Harrison and Klaverkamp (1990) reported high levels of 3.77 to 6.39 $\mu g \cdot g^{-1}$. Background sediment concentrations of mercury in Canada have been found to range from 0.01 $mg \cdot kg^{-1}$ to 1.6 $mg \cdot kg^{-1}$ with the mean concentration being 0.075 $mg \cdot kg^{-1}$ (Friske 1994). However, elevated concentrations have been noted in Ontario, Labrador, Northern Manitoba and in Yukon streams due to a variety of factors including the presence of massive sulfide and black shale deposits, glaciations resulting in transport and deposition, and faulted and techtonically-active terrains.

Several recent studies indicate that background mercury concentrations range from 1 to 20 $ng \cdot L^{-1}$ in freshwater (Kudo et al. 1982; Bloom 1989; Mierle 1990). In an acidified watershed in central Ontario, Mierle (1990) observed a positive correlation between aqueous mercury and dissolved organic carbon with total mercury concentrations below 5 $ng \cdot L^{-1}$ except during low flow periods when they exceeded 20 $ng \cdot L^{-1}$. Anthropogenic sources may elevate these levels considerably. For example, the Wabigoon River in Ontario received an estimated 10 metric tons of mercury from chlor-alkali operations with total mercury levels in water reaching up to 370 $ng \cdot L^{-1}$ reported near the outflow (Jackson et al. 1982), with levels typically in the 20-40 $ng \cdot L^{-1}$ range (Parks et al. 1989).

3. Canadian Environmental Quality Guidelines for Mercury

Mercury has been identified as a priority toxic substance in the Canadian environment, both through CEPA and by the CCME, and national environmental quality guidelines for mercury have been finalized, or are currently under development, for water, sediment and soil. Mercury has become an increasingly important issue in recent years since it is not only toxic in its inorganic form, but methylation greatly enhances it mobility and bioavailability. Since the primary site of methylation appears to be the sediments, and the sediments also act as a significant sink for this contaminant, the need for sediment quality guidelines has become apparent. In the following section, we briefly outline the derivation procedure for each of these guidelines using mercury as an example. Unless these values have been approved and published under the auspices of the CCME, they are not to be considered as final, recommended national guidelines.

The procedure and minimum toxicological data required to derive full and interim guidelines is specified in national protocols which have been developed for water, sediment, and soil (CCME 1991, 1994a,b,c). Each protocol includes criteria used to assess the acceptability of data, specified minimum data requirements for both full and interim guidelines, the derivation method and supporting rationale. Before a guideline is recommended for a substance, a complete assessment of that substance is conducted including production and uses, sources to the Canadian environment, environmental concentrations, behaviour in the environment, bioaccumulation, toxicity to both aquatic and terrestrial organisms and existing guidelines from other jurisdictions.

3.1 WATER QUALITY GUIDELINES FOR THE PROTECTION OF AQUATIC LIFE

Methods

Guidelines for the protection of aquatic life are developed both for protection of sediment benthos and biota in the overlying water column. The guidelines are defined as numerical limits designed to protect all forms of aquatic life during complete life cycles and indefinite exposure periods. The water quality guideline is derived by determining the lowest observed effect level (LOEL) for the most sensitive species and endpoint. Once the LOEL has been determined, an appropriate safety factor is applied such that the derived guideline represents a maximum concentration in water which should not be exceeded for the protection of aquatic organisms and designated uses.

Results for Mercury

Although the formal protocol for the development of Canadian water quality guidelines was established in 1991, mercury guidelines for water were adopted from other jurisdictions and published in the 1987 version. It was recommended that the total concentration of mercury in water was not to exceed 0.1 $\mu g{\cdot}L^{-1}$ based on the data of Reeder *et al.* (1979). In this study, toxic effects were examined for the most sensitive fish species identified in the literature review, the fathead minnow (*Pimephales promelas*) and the most toxic mercury species, methylmercury. It was found that levels of methylmercury in the edible parts of the fathead minnow exceeded the 0.5 $mg{\cdot}kg^{-1}$ limit for human fish consumption when exposed to 0.03 $\mu g{\cdot}L^{-1}$ methylmercury in water (Olson *et al.* 1975). Even though this concentration of methylmercury in the muscle did not cause adverse effects in the fish, the level set for human consumption would be reached before the fish were detrimentally affected. Therefore, in order to prevent this dangerous accumulation of methylmercury, levels of methylmercury in water should not exceed 0.01 $\mu g{\cdot}L^{-1}$. Assuming that methylmercury is less than 10% of the total mercury content of the water, the guideline for total mercury in water was adjusted to 0.1 $\mu g{\cdot}L^{-1}$.

Recent data has shown support for the adopted water quality guideline for mercury. Snarski and Olson (1982) have calculated the bioconcentration factor for mercury in the fathead minnow as being 4994. Since the permissible limit of mercury in fish muscle is 0.5 $\mu g{\cdot}g^{-1}$, the concentration of total mercury in water to protect fish from accumulating to this level is 0.1 $\mu g{\cdot}L^{-1}$. Chronic toxicity studies for the fathead minnow have indicated that effects occur below 0.23 $\mu g{\cdot}L^{-1}$. Therefore, the accepted WQG not only is supported by bioaccumulation data but also by chronic toxicity studies.

3.2 SEDIMENT QUALITY GUIDELINES FOR THE PROTECTION OF AQUATIC LIFE

Methods

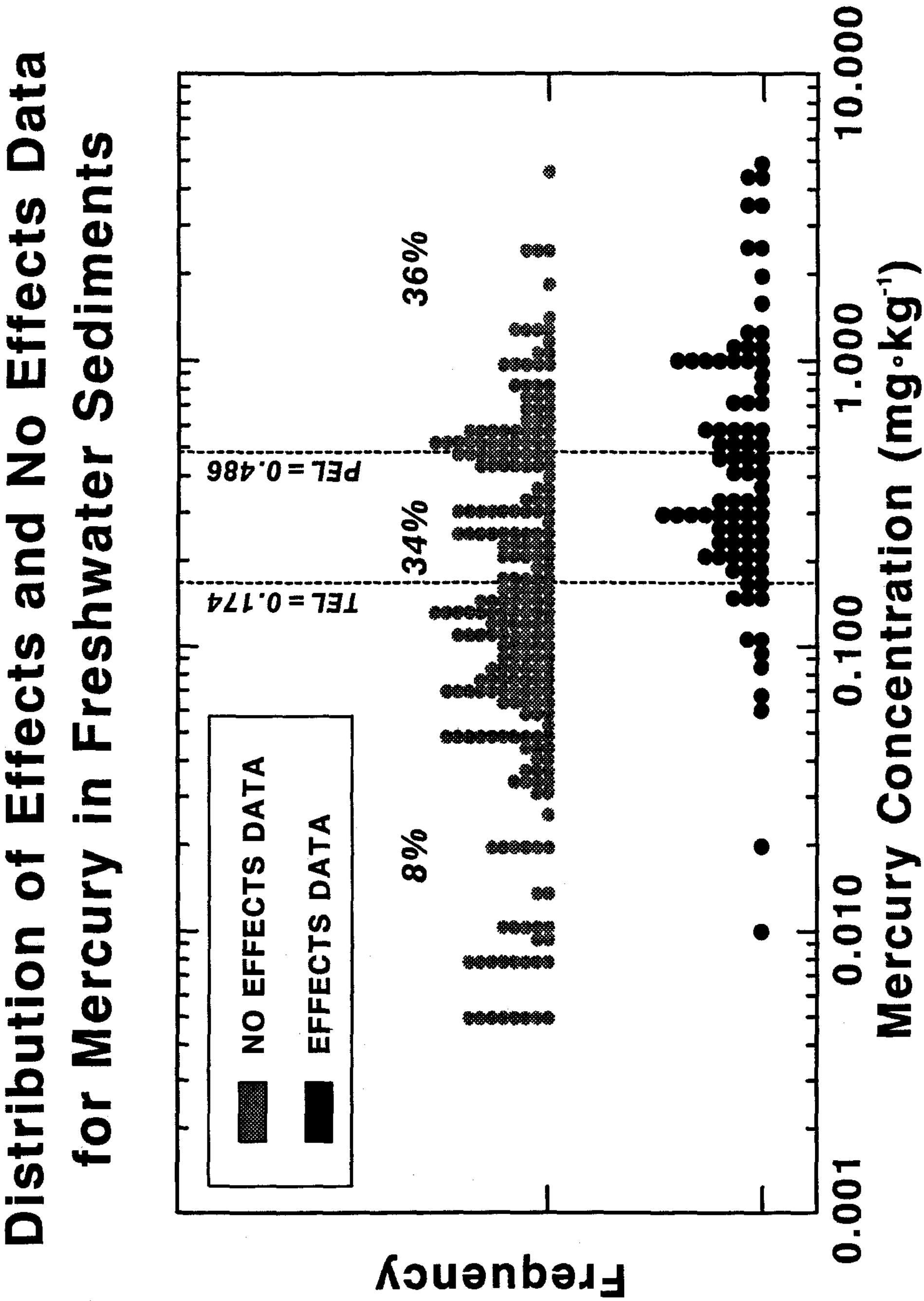
The purpose of sediment quality guidelines (SQGs) is to protect freshwater and marine (including estuarine) aquatic life associated with bed sediments. The protocol is based on two approaches: the National Status and Trends Program (NSTP) approach and the Spiked-Sediment Toxicity Test (SSTT) approach (CCME 1994b). The NSTP approach is a weight-of-evidence approach which includes data for a chemical generated from modelling (equilibrium partitioning theory), laboratory (spiked-sediment bioassay), and field studies (co-occurrence data consisting of matching sediment chemistry and biological effects data) (Long and Morgan 1990; Macdonald 1993; Long *et al.* 1994). This information is used to establish *associations* between concentrations of chemicals in sediments and adverse biological effects. All data is screened for acceptability and entered into a Biological Effects Database for Sediments (BEDS). A threshold effects level (TEL) is calculated as the geometric mean of the lower 15th percentile concentration of the effects data and the 50th percentile concentration of the no effects data set. This TEL consistently determines a range of sediment concentrations that is dominated by no effect data entries and represents the concentration below which adverse effects are not expected to occur.

The Spiked-Sediment Toxicity Test (SSTT) approach is a complementary procedure which will be used in the future to confirm and strengthen guidelines developed using the NSTP approach. The SSTT approach uses information on the response of test organisms to specific sediment-associated chemicals under controlled laboratory conditions (Chapman and Long 1983; Ingersoll 1991; USEPA 1992). SQGs will be developed using this approach once methodological issues have been resolved.

Results for Mercury

The distribution of BEDS data for total mercury for the freshwater environment is shown in Figure 1. The interim TEL for Mercury calculated for freshwater sediments is 0.174 $mg \cdot kg^{-1}$ dry weight. In addition to the TEL, the toxicological data can also be used to calculate a probable effects level (PEL) which represents the lower limit of the range of mercury concentrations that are almost always associated with adverse biological effects. For freshwater ecosystems, the PEL for mercury is 0.486 $mg \cdot kg^{-1}$. In marine ecosystems, the interim TEL and PEL calculated from the data are found to be 0.13 $mg \cdot kg^{-1}$ and 0.70 $mg \cdot kg^{-1}$, respectively.

The range defined by these two limits represents the range in which effects will occasionally be observed. By establishing these ranges, the likelihood of an adverse biological effect occurring at a given concentration can be estimated. This likelihood is calculated on the basis of the frequency distribution of the toxicity data by dividing the number of effect entries in a range by the total number of entries in that range, expressing this value as a percentage (Figure 1). In the case of freshwater ecosystems, only 8% of the mercury concentrations within the no effects range (0 to 0.14 $mg \cdot kg^{-1}$) are associated with adverse effects. In the possible and probable effects range for mercury, the incidence of adverse biological effects is 34% and 36% respectively. In marine ecosystems, the incidence of adverse effects is 8%, 24% and 37% in the no effects, possible effects and


Distribution of Effects and No Effects Data
for Mercury in Freshwater Sediments
Frequency
NO EFFECTS DATA
EFFECTS DATA
8%
34%
36%
TEL = 0.174
PEL = 0.486
0.001
0.010
0.100
1.000
10.000
Mercury Concentration (mg•kg-1)

probable effects range, respectively.

A full Canadian sediment quality guideline can only be recommended when the interim guideline is supported by a weight-of-evidence of the available ancillary data that links the interim sediment quality guideline with specific sediment types and/or characteristics of the sediment or overlying water column (e.g. particle size, TOC). At present, since most of the ancillary data represents means for sites, there is no correlation between the sediment concentration of mercury with any sediment or water characteristic. Therefore, only an interim sediment quality guideline can be recommended at this time.

The data presently available for the calculation of sediment quality guidelines are primarily from the United States, although Canadian data are included wherever they are available. However, a wide range of species and endpoints are employed in the tables, as well as broad ranges in sediment type, allowing the interim sediment quality guidelines to be applicable to a broad spectrum of circumstances.

3.3 SOIL QUALITY CRITERIA

Methods

Soil quality criteria (guidelines) are derived to sustain four major categories of land use in Canada - Agricultural, Residential/Parkland, Commercial and Industrial. Ecological guidelines for soil are based on protection of ecological (including domestic biota) receptors exposed either directly or indirectly to soil contaminants. Human health criteria for soil are also developed but will not be dealt with in the current paper. The ecological effects protocol (CCME 1994) accounts for exposure from direct soil contact (SQC_{SC}), contaminated soil ingestion (SQC_{SI}), and ingestion of plants grown on contaminated soil (SQC_{FI}). The ingestion procedures are generally intended to protect terrestrial wildlife and livestock from indirect exposure to bioaccumulating contaminants (log K_{ow} > 4). Within the direct contact procedure, there are three acceptable options for derivation of a criterion: 1) the Weight of Evidence approach; 2) the Lowest Observable Effect Concentration (LOEC) approach and 3) the Median Effects approach. The final determination of a guideline for the different land uses will vary as outlined in the 1994 protocol.

Depending on the available toxicity data and professional judgement, an uncertainty factor from 2 to 5 is applied to the derived value. The preliminary guideline is further "checked" to ensure that it is protective of microbial processes and groundwater. Information on natural background levels in the Canadian environment is also used to evaluate the final recommended value.

Results for Mercury

Sufficient data were available to develop Canadian soil quality criteria according to the CCME protocol (1994c), using the soil contact method but not for the soil ingestion or food ingestion methods (Table 1). Using the LOEC method the preliminary guideline for Agricultural and Residential/Parkland uses was calculated as 12 $mg \cdot kg^{-1}$ dry weight, based on the LOEC for lettuce (Environment Canada 1994, Table 1). Since only three (3) studies were available on only three (3) taxonomic groups, and the LOEC was from an acute toxicity study, a safety factor of 4 was applied to the LOEC resulting in a SQC_{SC} of 3

TABLE 1.

Summary of acceptable bulk soil toxicity data of mercury to terrestrial plants, invertebrates and microorganisms.

Species	Duration	Chemical Used	Test Substrate	Endpoint $mg \cdot kg^{-1}$				Reference
				NOEC	LOEC	EC_{25}	EC_{50}	
Radish (Seed emerg.)	72 h	$HgCl_2$	Artificial Soil: pH 4 to 4.2; Sand 70 to 75%;	51	103	73	103	Environment Canada 1995
Lettuce (Seed emerg.)	120 h	$HgCl_2$	Clay 16 to 22%; Silt 8 to 13%;	7	12	11	15	
Earthworm (survival)	14 d	$HgCl_2$	Moisture 80% WHC; org. matter 4.7 to 10.4%	96	194	130	181	
Microbial Processes (respiration inhibition and ATP synthesis reduction)	6 h/14 d	$HgCl_2$	Soil: pH 6.9; Organic matter 1.9 to 2.4%; Clay 7 to 25%; moisture 50 to 55% WHC	1.4 (g.m.)	14.78 (g.m.)			Zelles *et al.* 1986

g.m. - geometric mean of three studies

$mg \cdot kg^{-1}$. However, the microbial data showed that the mean No Effects Concentration for the effect of mercury on microbial processes is 1.47 $mg \cdot kg^{-1}$. The geometric mean of the two values was used to derive a final recommended guideline of 2.0 $mg \cdot kg^{-1}$ for Agricultural and Residential/Parkland uses.

For the Commercial and Industrial land uses, the geometric mean of all LOECs for ecologically relevant species and endpoints was used to recommend an initial guideline value of 62 $mg \cdot kg^{-1}$. This value was adjusted to reflect the results of the microbial check, resulting in a final recommended soil quality guideline of 30 $mg \cdot kg^{-1}$ for Commercial/Industrial land uses.

Major data gaps were identified for mercury in the terrestrial environment. Although a minimum of data exists to allow for the derivation of soil quality criteria for different land uses, there is still a paucity of data on the toxicity of mercury to soil ecosystem receptors. Thus, a great deal of research into soil mercury toxicity is required not only to improve understanding of the mechanisms, but also to validate the current soil quality criteria and improve upon it. Additional information is also needed on the background concentrations of mercury in soil. Though preliminary data indicate that mercury does not bioaccumulate in the terrestrial ecosystem, further information is required to validate this conclusion.

4. Summary and Conclusions

At present, there is sufficient data to develop water, sediment and soil quality guidelines for mercury. There is also a recognized need to develop tissue residue guidelines for aquatic ecosystems once this protocol has been finalized by the CCME. A variety of data gaps exist including insufficient Canadian data, limited knowledge on uptake of mercury species from soils and biomagnification of mercury up the food web. Further research needs to focus on these areas to expand the database used in developing the guidelines and as a means of field validating the proposed guidelines. In regards to sediment quality guidelines, relationships need to be established between concentrations of mercury in sediments, effects and sediment characteristics in order to derive full guidelines and allow for accurate predictions of toxicity at specific sites.

Environmental quality guidelines for mercury and other priority substances are important tools for environmental management, and there is an increasing emphasis in Canada on the use of environmental quality guidelines in environmental management. CCME guidelines have already been targeted as key elements of national management programs and strategies such as the Ocean Disposal Program and the National Contaminated Sites Remediation Program. These guidelines are not meant to be used independently but in conjunction with other management tools such as environmental effects monitoring, background concentrations and bioassays.

The range and scope of these national guidelines has gradually expanded, and will continue to expand to reflect our evolving understanding of the effects of contaminants in terrestrial and aquatic ecosystems and to ensure the long term sustainable use of Canada's resources.

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