Canadian Water Quality Guidelines for the Protection of Aquatic Life

MERCURY
Inorganic mercury and methylmercury

Mercury (CAS 7439-97-6, atomic mass. 200.6) is one of the most toxic metals found in the environment. It belongs to group IIb of the periodic table of elements and is called quicksilver because it is a silver-white liquid at room temperature. Mercury exists in three valence states (0, I and II). Elemental mercury (Hg^0) is chemically different from the other two members of group IIb metals, cadmium and zinc. Elemental Hg has a very low melting point (-39°C) compared with cadmium (321°C) and zinc (420°C). Hg^0 is quite volatile (vapour pressure 0.16 Pa at 20 °C) and is not readily soluble in water, though natural waters tend to be supersaturated with Hg^0 compared to air, resulting in its volatisation (Morel et al. 1998). Its high surface tension and uniform volume of expansion make mercury ideal for use in thermometers, barometers and other measuring devices (Smith and Rowan-West 1996).

In water, the mercurous state [Hg(I)] of mercury exists as a doubly-charged binuclear ion (dimer), Hg_2^{2+}. Hg(I) combines most commonly with inorganic molecules (Weber 1993). The chemical compounds of the mercuric ion [Hg(II)] are highly stable and much more numerous than those of Hg(I) (OECD 1994). The mercuric cation, Hg^{2+}, forms a relatively weak bond with chloride compared to bonds formed with other inorganic anions, although mercuric chloride ($HgCl_2$) may dominate when chloride salts are abundant (Alberta Environmental Protection 1992). $HgCl_2$ is more likely to be associated with sediments, with log K_{sed} values of 3.4-4.1 reported (Hurley et al. 1994). Hydroxyl anion (log K_1=10.6) has a higher affinity for Hg in the absence of organic complexing agents and will dominate in most freshwaters unless the pH is low, or chloride is high. Under reducing conditions, Hg^{2+} preferentially forms stable, largely covalent bonds with sulphide (including thiols) and selenides whenever these ligands are present (Jackson 1998).

Mercuric forms of Hg can be transformed through abiotic and biotic processes to form alkylmercury compounds such as monomethylmercury [CH_3Hg^+], dimethylmercury [$(CH_3)_2Hg$], and aryl compounds [e.g., phenyl-mercury] (Alberta Environmental Protection 1992). Monomethylmercury is commonly referred to as methylmercury and abbreviated as MeHg. It is very toxic and accumulates readily in aquatic biota (Beckvar et al. 1996; World Health Organization 1989). The dimethylmercuric form is volatile. Total mercury refers to the total concentration of all mercury species (i.e., both inorganic and organic forms).

Mercury occurs naturally, but significant amounts enter ecosystems through anthropogenic emissions, re-emissions and discharges. Natural sources of mercury include geological mercury deposits, rock weathering, forest fires and other wood burning, faults/volcanoes (land-based and oceanic), hotsprings, and a portion of the volatilisation from the oceans. The primary anthropogenic sources of Hg in Canada include: metal smelting; coal-burning power plants; municipal waste incineration; sewage and hospital waste incineration; coal and other fossil fuel combustion; cement manufacturing; and, mercury waste in landfills or storage (Pilgrim and Ecological Monitoring and Assessment Network 1998).

Mercury is used in dental amalgams, exterior paints, thermometers, barometers, and electrical products such as dry-cell batteries, fluorescent lights, switches, and other control equipment. It is used also in the electrolytic preparation of chlorine and caustic soda (chlor-alkali industry) and is an important chemical utilised globally by the gold mining industry to separate gold from other minerals into a gold-mercury amalgam. Mercury was used formerly as a seed and turf fungicide (Alberta Environmental Protection 1992).

Typically, MeHg represents less than 10% of the total Hg in surface waters, but can exceed 30% in perturbed systems such as newly formed reservoirs. In natural surface waters (freshwater and marine), concentrations of

Table 1. Water quality guidelines for mercury for the protection of aquatic life (Environment Canada 2003)*.

Aquatic life	Guideline value ($ng \cdot L^{-1}$)
Freshwater	
Inorganic Mercury	26
Methylmercury	4†
Marine	
Inorganic Mercury	16†
Methylmercury	NRG‡

*May not protect wildlife that consume aquatic life; see text for details.
†Interim guideline. May not protect fully high trophic level fish.
‡No recommended guideline.

total mercury range from <1 to 20 ng·L^{-1} while concentrations of MeHg are usually less than 1 ng·L^{-1}, though concentrations up to 4.1 ng·L^{-1} have been reported in the Gatineau River (Fisher et al. 1984; Kelly et al. 1997; Mierle 1990; Mierle and Ingram 1991; Schintu et al. 1989). Water draining from wetland areas tend to have higher concentrations of MeHg (mean of 0.626 ng·L^{-1}) than water from watersheds lacking wetlands (mean of 0.03 ng·L^{-1}) (Kelly et al. 1995; St. Louis et al. 1994). Mean MeHg concentrations increased from less than 0.1 to greater than 1 ng·L^{-1} following the creation of an experimental reservoir; total mercury levels remained relatively constant at 2.5-3.0 ng·L^{-1} (Kelly et al. 1997; Paterson et al. 1998). Concentrations of total mercury and methylmercury in the ranges of those found in natural surface waters have been measured in rain and snow samples as well.

The net production of methylmercury in aquatic environments is a balance between methylation and demethylation; both may occur through abiotic and biotic (microbial) processes. Several factors influence the rate of net MeHg production including: the concentration and availability of Hg^{2+}; composition of the microbial population; nutrient and mineral substrate; pH; temperature; redox potential; dissolved and particulate organic matter (DOC and POM); salinity; iron; and sulphate. Bacterial activity increases with increasing temperature and available biodegradable organic carbon. Thus, methylation rates tend to be highest in surface sediments with freshly deposited organic matter, and in warm shallow sediments where abundant bacterial activity takes place (Ramlal et al. 1986; Winfrey and Rudd 1990). Often, newly created reservoirs increase temporarily the amount of methylmercury in aquatic systems, including food chains, due to the accelerated microbial methylation of existing inorganic Hg forms caused by decomposing flooded vegetation (Abernathy and Cumbie 1977; Schetagne et al. 1999).

Methylmercury is of special concern not only because of its toxicity, but also because of its tendency to biomagnify in upper trophic levels of aquatic food webs. Mercury compounds bind strongly with sulphydryl groups in proteins. MeHg passes easily through the digestive wall and bioconcentrates in tissues, whereas inorganic Hg is more likely to be excreted (Boudou and Ribeyre 1985). Organisms at lower trophic levels usually contain the lowest proportion of total mercury as MeHg and uptake is primarily a passive process occurring by adsorption to or absorption within the cell (Beckvar et al. 1996). Aquatic plants contain a low percentage of MeHg, typically less than 50% of total Hg. Invertebrates often contain about 50% MeHg and 50% inorganic mercury (Hildebrand et al. 1980). Diet is the most important route of uptake of MeHg for organisms higher in the food chain, like piscivorous fish (e.g., walleye, lake trout), aquatic birds (loons, herons), piscivorous mammals (mink, otters), and marine mammals. These animals contain a very high proportion of total Hg as MeHg in muscle tissue (90 – 100%).

Numerous chemical and physical variables of surface waters determine the potential for mercury to bioaccumulate in fish (Environment Canada 2002). In particular, low pH (<6), low alkalinity (acid-neutralizing capacity 50 μeq·L^{-1} or less), and low calcium (<5 mg·L^{-1}) lakes are associated with elevated mercury concentrations in fish (Grieb et al. 1990; Spry and Wiener 1991).

Water Quality Guideline Derivation

The Canadian water quality guidelines (WQGs) for inorganic mercury and methylmercury for the protection of aquatic life were developed based on the CCME protocol (CCME 1991). Insufficient data exist to derive a marine water quality guideline for methylmercury.

The protocol does not address exposure through food or bioaccumulation to higher trophic levels. As such, aquatic life that are exposed to methylmercury primarily through food (e.g., piscivorous fish) may not be adequately protected. Moreover, **these WQGs for mercury may not prevent the accumulation of methylmercury in aquatic life; therefore, through this process the tissue residue guideline (TRG; 33 μg MeHg·kg^{-1} ww) for the protection of wildlife that consume aquatic life may be exceeded** (Environment Canada 2002). Thus, if the ultimate management objective for mercury is to protect high trophic level aquatic life and/or those wildlife that prey on aquatic life, more stringent site-specific application of these water quality guidelines may be necessary (see Additional Considerations). Use and derivation of site- and species-specific water quality objectives is provided by Environment Canada (Environment Canada 2003).

Freshwater Life

Inorganic Mercury

In fresh waters, acute (24- to 96-h LC_{50}) toxicity concentrations for inorganic Hg range from 5 to 5600 µg Hg·L^{-1} in invertebrates and from 150 to 900 µg Hg·L^{-1} in fish (Biesinger and Christensen 1972; Call et al. 1983; Rehwoldt et al. 1973; Wobeser 1975). From limited acute data, algae appear sensitive with 24-h LC_{50}s from 9 to 27 µg Hg·L^{-1} for inorganic Hg (Chen and Lin 1997).

In chronic tests (7- to 21-d), invertebrates are about as sensitive to mercury as fish. Effect concentrations (EC_{50}s) in invertebrates range from 1.28 to 12.0 µg Hg·L^{-1} for inorganic Hg (Biesinger et al. 1982; Spehar and Fiandt 1986). In fish, chronic values for inorganic Hg range from 0.26 to >64 µg Hg·L^{-1} in 5- to 60-d tests (Niimi and Kissoon 1994; Snarski and Olson 1982). Amphibians are sensitive also to inorganic Hg with 5- to 21-day LC_{50}s ranging from 1.3 to 67.2 µg Hg·L^{-1} (Birge et al. 1979). Adverse effects reported commonly among all studies include growth, impaired reproduction and development, and death.

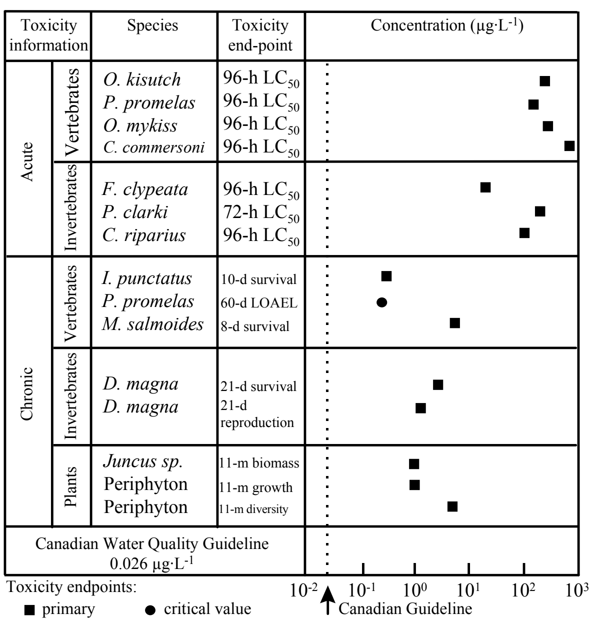

Figure 1. Select freshwater toxicity data for inorganic mercury

The guideline value recommended for inorganic Hg is based on the most sensitive LOAEL of 0.26 µg Hg·L^{-1} for juvenile fathead minnows (*Pimephales promelas)* reported by Snarski and Olson (1982). In 60-d flow-through experiments, the authors observed reduced growth (weight) of exposed offspring from exposed parents, and reproductive impairment evidenced as reduced spawning and egg production.

The LOAEL was divided by a safety factor of 10 to give a Canadian water quality guideline of 0.026 µg Hg·L^{-1} or 26 ng Hg·L^{-1}.

Methylmercury

Acute toxicity (24- to 96-h) concentrations for MeHg range from 24 to 125 µg Hg·L^{-1} in fish and from 3.5 to 6.3 µg Hg·L^{-1} in algae (no invertebrate data available) (Thomas and Montes 1978; Wobeser 1975). In chronic tests, EC_{50}s range from 0.04 to 1.14 µg Hg·L^{-1} for invertebrates and from 0.93 to 63 µg Hg·L^{-1} in fish (Biesinger et al. 1982; McKim et al. 1976; Spehar and Fiandt 1986). In side-by-side tests, MeHgCl is typically

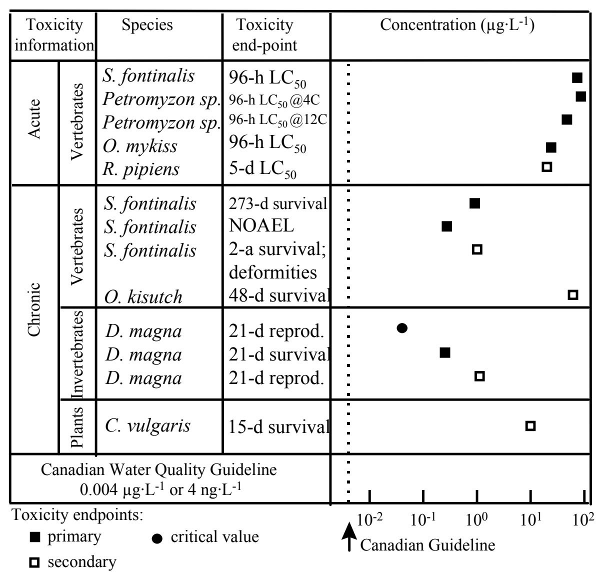

Figure 2. Select freshwater toxicity data for methylmercury.

more than ten times as toxic as $HgCl_2$ to fish, invertebrates, and aquatic plants (Biesinger et al. 1982; Niimi and Kissoon 1994; Thomas and Montes 1978).

An interim guideline was recommended for MeHg based on a combination of a high quality study that reported reproductive effects on *Daphnia magna*, and several research papers comparing the toxicity of inorganic Hg relative to MeHg. A range of measured concentrations from 0.04 to 0.26 µg Hg·L^{-1} (as MeHg) in flow-through experiments caused a significant decrease in *D. magna* production of young (Biesinger et al. 1982).

The LOAEL of 0.04 µg Hg·L^{-1} was divided by a safety factor of 10 to derive an interim Canadian water quality guideline of 0.004 µg L^{-1} or 4 ng·L^{-1}. This guideline is recommended for the protection of low trophic level freshwater life (i.e., generally trophic levels 1-2) against the adverse affects of direct exposure to methylmercury through water. This guideline may not protect high trophic level aquatic life (i.e., generally trophic levels 3 and 4) which are exposed to methylmercury primarily through food. Nor may it prevent the accumulation of methylmercury in aquatic life which could cause the tissue residue guideline (33 µg·kg^{-1} diet ww) for the protection of wildlife consumers of aquatic biota to be exceeded (Environment Canada 2002).

Marine Life

Inorganic Mercury

Data for marine waters are much more limited but trends are similar to those observed for fresh waters. A single acute (96-h) study on the effects of inorganic Hg on fish

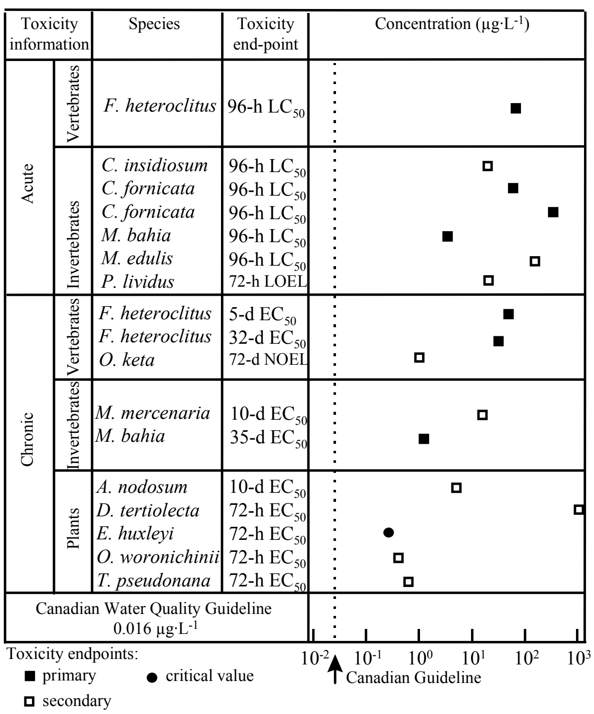

Figure 3. Select marine toxicity data for inorganic mercury.

reported a LC_{50} of 68 µg Hg·L^{-1} (Sharp and Neff 1982). For invertebrates, 24- to 96-h LC_{50} range from 3.5 to 161 µg Hg·L^{-1} (Lussier et al. 1985; Nelson et al. 1988).

Similar to freshwater studies, chronic studies on marine life report reduced growth and survival, and impaired development (e.g., increased incidence of deformities). EC_{50}s for inorganic Hg range from <5 to 55 µg Hg·L^{-1} for fish, from 1.2 to 20 µg Hg·L^{-1} for invertebrates, and from 0.16 to 1002 µg Hg·L^{-1} for plants and algae (Brown and Parsons 1978; Fisher et al. 1984; Lussier et al. 1985; Sharp and Neff 1982; Warnau et al. 1996).

The LOAEL of 0.16 µg Hg·L^{-1} was used to derived the guideline. In this static test, exposure to inorganic mercury (as $HgCl_2$) for 72-h reduced the growth of a population of the coccolithophore algae, *Emiliania huxleyi* by 50%; 0.32 µg Hg·L^{-1} halted growth completely (Fisher et al. 1984). The LOAEL was divided by a safety factor of 10 to give an interim Canadian water quality guideline of 0.016 µg·L^{-1} or 16 ng Hg·L^{-1}.

Additional Considerations

To attain the highest degree of environmental protection, all Canadian Environmental Quality Guidelines for mercury (water, sediment, tissue, and soil) should be applied concurrently.

Toxicity of mercury is negatively correlated with salinity, selenium concentration, and oxygen content, and positively correlated with temperature (reviewed by Cuvin-Aralar and Furness 1991; Heit and Fingerman 1977; MacLeod and Pessah 1973; McKenney, Jr. and Costlow, Jr. 1981; Slooff et al. 1991; Snell et al. 1991). Water hardness has a negligible effect on mercury toxicity unlike other toxic metals such as copper or aluminium, where toxicity significantly decreases with increasing hardness (Keller and Zam 1991).

The issue that faces many environmental managers is ensuring the protection of wildlife consumers of mercury-ladened fish. Calculations using reference concentrations[1] of MeHg for wildlife species and field-based bioaccumulation factors (BAFs) produced estimates of water concentrations that could protect wildlife that consume aquatic biota. These generic calculations are intended as a guide to determining site- and species-specific water quality objectives. From conservative assumptions, concentrations of MeHg below 0.007 ng Hg·L^{-1} may be

[1] A reference concentration is the concentration of MeHg in tissues of aquatic biota below which adverse effects are not expected for a given species of wildlife that consume aquatic biota (see Chapter 8). Reference concentrations for MeHg were derived for a suite of Canadian wildlife species; the lowest of which, that for Wilson's storm petrel, *Oceanites oceanicus* (33 µg/kg diet ww), was selected as the Canadian Tissue Residue Guideline (Environment Canada 2002).

required to protect all wildlife species in Canada while concentrations above 0.2 ng Hg·L^{-1} may pose a risk to wildlife species. MeHg concentrations in water between these limits may be hazardous to some wildlife depending on their feeding habits (preferred prey items, and the trophic level and BAFs of these prey items). More specific information is given in the supporting document for these guidelines (Environment Canada 2003).

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