Methylmercury Contamination: Impacts on Aquatic Systems and Terrestrial Species, and Insights for Abatement

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Methylmercury (MeHg) is one of the most widespread waterborne contaminants, and through biomagnification processes has commonly been found in prey fish at concentrations toxic to piscivorous birds and mammals. In USEPA's National Fish Tissue Survey, the Lowest Adverse Effect Concentration of 0.1 g/g wet weight was exceeded in 28% of total samples and 86% of predatory fish samples (USEPA 2002). Evidence suggests that more than a quarter of all mature fish contain methylmercury concentrations above this level. This review focuses on the leading anthropogenic sources of mercury to aquatic systems, through atmospheric deposition and the environmental dynamics of the mercury methylation process in aquatic sediments. The results of extensive mercury monitoring studies are discussed, as well as the reproductive and behavioral impacts on birds and mammals feeding on fish exhibiting realistic contaminant concentrations. Recommendations are provided for continued research and the abatement of methylmercury concentrations in fish through forest management practices.

Keywords: *mercury deposition, methylmercury, sulfate reducing bacteria, aquatic sediments*

Introduction

Methylmercury (MeHg) is one of the most widespread waterborne contaminants (USGS 2001a; UNEP 2002; USDHHS and USEPA 2004). Unhealthful levels of MeHg in fish have led to the issuance of fish consumption advisories by at least 46 states (USEPA 2004). The generation, bioaccumulation, and biomagnification of MeHg within aquatic systems has been studied for decades, following the identification of severe neurological and teratogenic impacts to humans associated with the consumption of contaminated fish in Minimata Bay, Japan in the 1950s (Eisler 1987; Ninomiya et al. 1995). MeHg has been linked to potential reproductive and immune system effects in humans and wildlife (Wiener et al. 1996; USEPA 1997d; Round et al. 1998). To protect human health, the U.S. Environmental Protection Agency (USEPA) has set a generalized, default fish tissue mercury residue criterion for freshwater and estuarine fish at 0.0175 mg per kg of fish per day (USEPA 2001). With regard to management of Forest Service lands, MeHg is recognized to be a significant risk to the viability of natural systems associated with aquatic resources (Hammerschmidt et al. 1999, 2002; Gnamus et al. 2000). Of the over 40.4 million ha (100 million acres) of freshwater wetlands within the conterminous United States, over 20.2 million ha (50 million acres) were determined to be forested wetlands, as well as 10.0 million ha (25 million acres) of emergent wetlands and 7.3 million ha (18 million acres) of shrub wetlands (Dahl 2000). While National Forest lands represent only eight percent of the contiguous United States, they contribute 14 percent of the runoff (USDA FS 2000). National Forests and Grasslands contain over 240,000 km (150,000 miles) of streams and 1 million ha (2.5 million acres) of lakes (USDA FS 2004). Anglers spent nearly 50 million days fishing on National Forests in 1996, and generated US $2.9 billion (NFF 2004). Clearly, the Forest Service has a large stake in the study, prevention, and possible abatement of mercury contamination within the nation's waters.

This review focuses on the primary anthropogenic source of mercury ultimately affecting aquatic systems - coal combustion. Much smaller contributions from wildland fire, associated with Forest Service land management activities, will share that focus. Also discussed are: subsequent mercury deposition to aquatic systems, dynamics of conversion to MeHg within fresh water aquatic sediments, impacts on aquatic and terrestrial species, proposed controls and regulatory initiatives, and suggested mitigation measures.

Sources

The bulk of MeHg within natural systems originates from methylation of atmospherically deposited mercury

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species by sulfur reducing bacteria within aquatic sediments (USEPA 1997d). Long thought to originate from mercury-laden industrial point source effluent, mercury contributions from geological processes, or both, MeHg contamination has more recently been portentously linked to long-term, long-range transport of emissions from fossil fuel combustion sources throughout the industrialized world (USEPA 1997b, 1997d; UNEP 2002; Bullock 2004). Despite the uncertainties involved in determining emission inventories, it is widely accepted that anthropogenic emissions have increased relative to natural sources since the beginning of the industrial period (Fitzgerald et al. 1998). This has resulted in atmospheric deposition of elemental and oxidized mercury species, even in remote areas (Rasmussen 1994; Sorensen et al. 1994; Morrison and Watras 1996; Eisler 1998; Fitzgerald et al. 1998; USGS 2000; St Louis et al. 2001; Blett 2002; MPCA 2004). Concern exists that most National Forest lands are characterized by relatively remote freshwater systems, many showing signs of MeHg contamination (USDA FS 2000).

The burning of fossil fuels, primarily coal, is the best characterized and most dominant anthropogenic source of mercury emissions (USEPA 1997d; Seigneur et al. 2003), representing over 30% of mercury emissions from domestic sources and 29% of global anthropogenic emissions (Seigneur et al. 2004). Approximately 40% of the 68 metric tons (75 tons) of Hg from the coal burned domestically remains in the ash and scrubber residues, while 60% is emitted to the atmosphere (ICR 2000). The ratio of mercury deposition from international and domestic sources varies considerably by location within the United States (Bullock 2004). It is estimated that North American anthropogenic emissions account for only 1.5-3% of global emissions (Bullock 2004; Seigneur et al. 2004). However, modeling studies estimate that 95-96% of mercury deposition in the eastern United States and 88-90% in the western United States comes from anthropogenic sources (Levin et al. 2000; Bullock 2004).

Comparing estimates of long-term accumulation rates of total mercury within soil of northwestern Ontario to measured flux rates within litterfall and throughfall, St. Louis et al. (2001) found they were similar, suggesting that inputs of total mercury originated from atmospheric deposition. Hanisch (1998) indicated that 40% of mercury deposition could be attributed to anthropogenic sources. Short range modeling data reported by USEPA (2001), using the ISC-3 model, indicated that from 7-45% of primary, inorganic mercury (Hg^{II}) emissions, originating from eastern U.S. sources with relatively lower stacks, are deposited within 50 km of their source. Similar western source emissions were modeled at rates from 2-38%. Regional differences were attributed to differing frequencies of precipitation events. Round et al. (1998) reported that most mercury emissions from taller stacks, including elemental mercury (Hg^0) oxidized to Hg^{II}, are deposited within 1000 km of the source. Electric utilities are recognized as the largest single source category of mercury emissions in North America (Seigneur et al. 2004), but several other source categories are known to emit large amounts of mercury (Dvonch et al. 1999).

There are distinct regional differences across the United States with respect to mercury input values per 1012 Btu for various coal types (ICR 2000; USGS 2001b). Initially, it may be assumed that the environmental impact of mercury emissions from the burning of any given coal type may be a factor of its mercury content relative to its Btu value. In addition, the ratio of various mercury species within combustion source emissions will influence the percentage of mercury initially available for methylation within natural systems (Seigneur et al. 2004). Within 14 regions, the USGS (2001b) lists Gulf Coast lignite as exhibiting the highest mercury to Btu value (20 lb Hg per 1012 Btu [9 kg Hg per 293 million kwh]) followed in order of magnitude by coal from the northern Appalachian, southern Appalachian, western interior, Fort Union, and Pennsylvania anthracite deposits. ICR (2000) data, based strictly on reported values from the utility sector, found similar regional rankings, but significantly lower mercury to Btu ratios.

While anthropogenic emissions of oxidized and elemental mercury are widespread, they are only a fraction of total global contributions (Grumet 2000). Seigneur et al. (2003) and Lindberg and Stratton (1998) identified speciation ratios of anthropogenic emissions (the relative fractions of Hg^0, Hg^{II}, and particulate mercury or Hg^P), as critical to the environmental fate of that mercury. Anthropogenic emissions vary widely in the percentage of Hg^0, Hg^{II}, and (Hg^P) forms, with Hg0 representing the vast majority of the worldwide atmospheric mercury load (USEPA 1997c; Round et al. 1998). Hg0 remains in the atmosphere for up to a year (USEPA 1997c). Conversely, Hg^{II} species, whether emitted as primary pollutants or transformed within the atmosphere from Hg^0, are much more water soluble, more easily deposited during rain events (Seigneur et al. 2003), and are principally the mercury species which undergo methylation within natural systems (USEPA 1997a). Seigneur et al. (2003) reported that up to 50% of Hg^{II} species were depleted by rainfall, whereas less than 10% of Hg^0 species were depleted. An elevated mercury deposition pattern, associated with summertime thunderstorms, became evident at ten sites across Florida in periods ranging from 2 to 5 years (Guentzel et al. 2001). This was estimated to be >50% of the mercury deposition

in southern Florida. During a single summer rain event, a northern Wisconsin monitoring station received two thirds of its annual mercury load (L. Bruss, Section Chief, Wisconsin DNR–Bureau of Air Management, Personal communication, 2003).

2003). The propensity of atmospheric Hg^{II} to be depleted by rainfall, and the indication that vegetation may act as a sink for Hg^{II} species, suggests that elevated ecosystem exposure to Hg^{II} may be possible near a major mercury emitter (Lindberg and Stratton 1998). Table 1 lists mercury particle speciation profiles for a number of anthropogenic source categories.

While a number of studies have markedly increased our understanding of both total Hg and MeHg cycling within forested watersheds (Bishop et al. 1998; St. Louis et al. 2001; Hintelmann et al. 2002; Munthe and Hultberg 2004), few studies address the role of wildland fire in mercury cycling. One of the more spectacular examples of gaseous mercury transport within the plume of a large wildfire occurred during July 2002, when researchers using carbon monoxide (CO) as a tracer of a plume originating from a series of boreal wildfires in northern Quebec calculated a strong correlation between mercury levels and CO at Harvard Forest in western Massachusetts (Singler et al. 2003). Average flux rate for these boreal fires was determined to be 1.5 µg Hg per hectare, resulting in an annual Canadian wildfire emission rate of 3.5 metric tons Hg, equaling 30% of average Canadian anthropogenic emissions. Singler et al. estimated annual global boreal wildfire emissions to be 22.5 metric tons.

Table 1: Emission speciation profiles for various anthropogenic mercury source categories. Adapted from Round et al. 1998, with biomass fire data taken from Friedli et al. 2003.

	Speciation (%)[a] of Mercury Emissions		
Source Type	**Hg^0**	**Hg^{II}**	**Hg^P**
Electric utility fossil fuel boilers	50	30	20
Non-utility fossil fuel combustion	50	30	20
Municipal waste combustion	20	60	20
Medical waste incineration	20	60	20
Chlor-alkali factory	70	30	0
Other point sources[b]	80	10	10
Biomass fires[c]	>95	negligible	<5
Area sources[d]	100	0	0

[a] Hg^0 symbolizes elemental mercury; Hg^{II} symbolizes divalent, oxidized mercury; Hg^P symbolizes particulate mercury.

[b] Includes residential boilers, sewage sludge incinerators, wood-fired facilities, lime manufacturing, mercury compounds production, cement manufacturing, and secondary mercury production. However, a number of potentially important source categories (such as refineries) are not included because emissions estimates for these sources are currently lacking.

[c] From Friedli et al. 2003.

[d] Because most area sources do not involve combustion sources, emissions were assumed to be 100% Hg^0 for this source category. However, a small (but presently unknown) fraction of area source emissions may be in divalent or particulate forms.

Using fuels from across the United States, Friedli et al. (2003) demonstrated in the laboratory that nearly all mercury from biomass fires may be emitted as elemental mercury. Friedli et al. also sampled smoke from a small wildfire with a research aircraft, and determined that wildfires may emit a larger percentage of Hg^P than was determined from laboratory studies. This additional particulate mercury was likely released from fire-heated soils. Mercury concentrations ranged from 14 - 71 µg Hg/kg (dry mass) fuel for the laboratory burns and 112 µg Hg/kg for the wildfire. It is evident that forests act as sinks for atmospheric mercury, and wildland fire emissions contain mercury deposited on and incorporated in fuel, as well as that which may be released from fire-heated soil (Friedli et al. 2003). Forest Service researchers are currently researching mercury mobility and accumulation in fish responding to wildland fire in the Boundary Waters Canoe Area of northern Minnesota's Superior National Forest (Kolka 2003). There is also some evidence that mercury may be mobilized during soil disturbances from construction of logging roads or large fire breaks (Munthe and Hultberg 2004).

Environmental Dynamics

Sulfur reducing bacteria within aquatic sediments are ubiquitous, and are recognized as the primary agent of mercury methlyation within aquatic systems (Wetzel 1983; King et al. 2002; Bates et al. 2002). Benoit et al. (1999) postulate that the presence of mercury complexes such as cinnabar (HgS), the dominate neutral mercury complex in sulfidic sediment pore waters, mediates bacterial methylation of mercury by passively diffusing across bacterial membranes, and being used in sulfate reduction/ mercury methylation reactions. Other researchers point to precipitation of relatively insoluble HgS, decreasing mercury availability, and inhibiting MeHg production (Gilmour et al. 1998; King et al. 2002). Gilmour et al. also determined that those areas with sediments exhibiting high sulfur reduction rates were characterized by lower relative sulfate levels, and thus lower levels of sulfide inhibiting methylation. Wetzel (1983) reported the conditions within aquatic sediments, relative to redox potential, supporting the presence of sulfur reducing bacteria, and therefore also mercury methylation (See Figure 1). Ultimately, translocation of easily-soluble MeHg out of the sediments

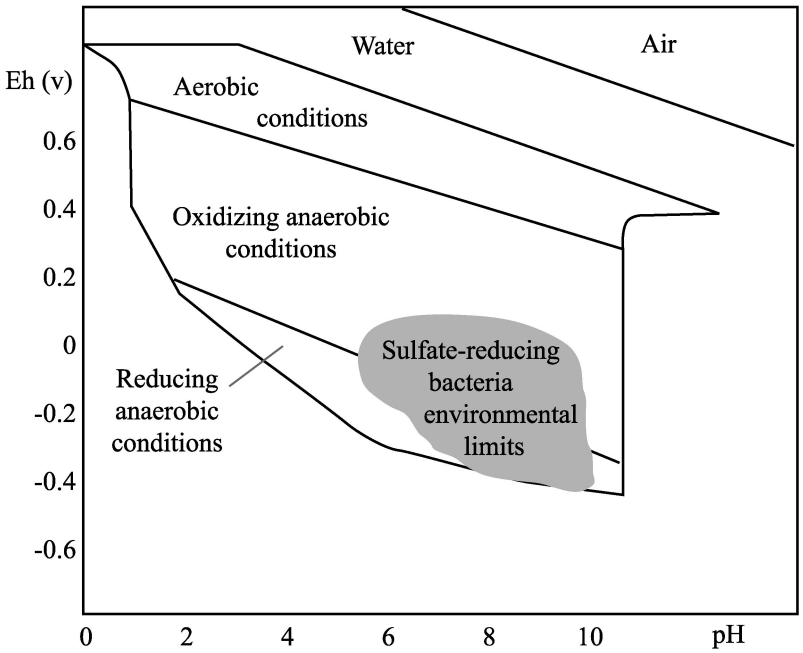


Figure 1: Environmental limits of sulfur reducing bacteria in aquatic sediments. Based on Wetzel (1983).

leads to its biomagnification within higher trophic levels (USEPA 1997d).

Due to the relative abundance of anaerobic sulfur reducing bacteria in associated sediments, wetlands and those lakes characterized by large shoal areas appear to generate methylmercury at greater rates than other freshwater systems (Sutton 1998; King et al. 2002). In addition, flooding appears to initially and significantly increase the production and export of MeHg (Kelly et al. 1997; Gerrard and St. Louis 2001) and its bioaccumulation (Paterson et al. 1998). Based on this evidence and Wetzel's (1983) indication of optimal redox conditions, the avoidance of flooding and the minimization of anthropogenic water level manipulations may offer a means to minimize MeHg production, even within existing lakes and wetlands.

Several factors independent of mercury depostion are associated with MeHg concentrations in fish. Abiotic factors include an increase in dissolved organic matter (Sorensen et al. 1990; Babiarz et al. 2001) as associated with increased leaf fall and subsequent algal blooms (Balogh et al. 2002), fluctuation of lake levels (Wiener et al. 2002), and higher sulfate-laden inflow to shallow waters (Gilmour et al. 1992, 1998; Sutton 1998; Harmon et al. 2003; Jeremiason et al. 2003; MPCA 2004). In addition, proximity to geothermal vents, land disturbance events, and major sewage treatment plants (Sutton 1998) and some mining operations (Wiener et al. 2002) are also associated with higher MeHg levels in fish.

It is unclear how pH within the water column affects MeHg levels. Sorensen et al. (1990) determined pH was negatively correlated with fish MeHg levels, contrary the Sorensen et al. literature review and that of Sutton (1998). This discrepancy could be related to the inclusion of deeper lakes in the Sorensen et al. study. HgS nodule precipitation occurs readily under reducing conditions within the deeper areas of hypolimnetic waters, decreasing the amount of mercury available for methylation. However, if pH is high within these hypolimnetic waters, Hg can become soluble and so available for methylation (Sutton 1998).

On her comprehensive website, Sutton (1998) reported that ideal lake conditions leading to high levels of mercury methylation include: high sulfate levels, shallow well-mixed lake waters, deep lakes with a high pH, or lakes with large shallow areas. Hurley et al. (1995), in their study of 39 river sites within Wisconsin, found MeHg generation rates were positively correlated with the percentage of wetlands within a hydrologic unit, and were highest within watersheds containing greater percentages of wetland/forest sites relative to agricultural/forest sites or agricultural only sites. In their study of 80 lakes in remote northern Minnesota, Sorensen et al. (1990) also found the significant ($|r| > 0.90$), water related and positively correlated predictor variables: Al (aluminum), low sediment acid neutralizing capacity (ANC), percent of watershed in forest, lake surface area, and watershed area. High Al and low ANC have also been associated with lakes sensitive to acid deposition (Adams et al. 1991).

As part of the comprehensive METAALICUS study, Hintelmann et al. (2002) found that in a boreal forest, the initial mobility, and so the ultimate bio-availability of mercury received through wet and dry deposition, decreased markedly in a short time through methylation relative to the larger pool of stored mercury, suggesting

that there may be rapid decline in rates of MeHg bioaccumulation in fish if mercury deposition is reduced. Hintelmann et al. further suggest that this decline could take less than 10 years, although WDNR (1999) suggests 15-20 years. Potential exists for land managers and air quality regulators to influence the abatement of mercury and sulfur deposition to aquatic systems, and so abate MeHg concentrations within these systems. However, the mercury isotope marker in the Hintelmann et al. study was bound to vegetation to a much higher degree than native mercury, suggesting there may be a time delay before atmospherically derived mercury enters the mercury soil pool, and therefore before that soil pool would respond to changes in deposition.

General knowledge of the factors that contribute to MeHg bioaccumulation would not be complete without noting that abiotic and biotic MeHg degradation pathways exist within natural systems, mitigating MeHg toxicity. In addition to the abiotic processes of MeHg photodegradation in lakes (Seller et al. 1996), these include production of HgS within soil pore water (Benoit et al. 1999), and dissolution of HgS in the presence of humic and fulvic acids (Ravichandran et al. 1998). Various aerobic and anaerobic microbial populations are known to possess enzyme degradation systems that react with mercury species. These systems impart to the host some resistance to the toxic effects of MeHg, and result in the cleaving of MeHg, forming CH4 and HgII. Some bacteria possessing a more "broad spectrum" resistance have the ability to further reduce HgII, forming volatile, elemental mercury (Marvin-Dipasquale et al. 2000). However, the evident widespread bioaccumulation and biomagnification of MeHg suggests that these degradation mechanisms do not dominate within aquatic sediments.

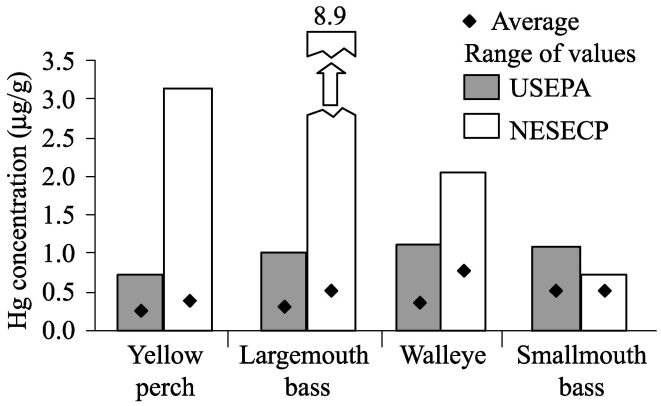
Of particular concern to the Air and Watershed Programs of the Forest Service is the correlation between the presence of sulfates and MeHg production within wetland systems. An example of note is the repeated and long-term applications of sulfur-rich agricultural fertilizers on the sugarcane fields of southern Florida, resulting in significant MeHg loadings to fish and birds downstream, within the Everglades wetland system (Bates et al. 2002). Conversely, in their study of total mercury bioaccumulation in fish, the United States Geological Survey (USGS 2001a) found a significant negative correlation of mercury bioaccumulation with sulfate in water within 20 river basins nationwide. However, nearly all collection sites were within larger streams, and it is unclear how these collection sites relate to their associated wetland systems or sulfate inputs. Preliminary results of an ongoing study within a two-hectare wetland by researchers on the Marcell Experimental Forest in northeastern Minnesota (Jeremiason et al. 2003; MPCA 2004) clearly demonstrate an increase in peat pore water MeHg concentrations following sulfate addition. While sulfate concentrations decreased following its addition, MeHg concentrations rose until pore water sulfate levels were depleted, corresponding to the findings of the USGS (2001a) that the presence of acid volatile sulfide within sediment pore water will signal the reduction of available sulfate, and thus the reduction in the production of MeHg. Most significantly for this review, further study is needed to quantify an abatement of mercury levels in fish which could stem from reductions in industrial sulfur dioxide emissions, and subsequent sulfate deposition.

Effects on Wildlife

MeHg concentrations generally increase with trophic level and with increased size and age of a given organism. Fish eventually sequestered MeHg in skeletal muscle, reducing exposure to the central nervous system (Wiener et al. 2002). Haines (1996) reported mercury concentrations in wild freshwater fish populations in 125 randomly selected lakes in Maine commonly exceeded 1 μg/g wet weight, the United States Food and Drug Administration's action level, or that level deemed unfit for human consumption (USDHHS and USEPA 2004). Half the samples from the Haines study exceeded 0.5 μg/g. USEPA's two-year, National Fish Tissue Study found concentrations exceeding 1 μg/g in only eight of 282 composite samples of predatory, freshwater game fish distributed across the United States, while the highest concentration in the 237 composite samples of bottom dwelling fish was 0.531 μg/g (USEPA 2002, 2003). However, more than 28% of the total samples exceeded 0.1 μg/g, with over 86% of predatory fish exceeding this level. Levels exceeding 1 μg/g were also common as reported in the Northeast States and Eastern Provinces' comprehensive mercury study (Tatsutani 1998), with largemouth bass (*Micropterus salmoides*) reaching 8.94 μg/g. As in all studies reviewed, predatory fish exhibited the greatest mean and maximum mercury values. Figure 2 compares data from the USEPA and Tatsutani studies.

Numerous laboratory studies have established a link between the ingestion of MeHg at sublethal levels and subtle visual, cognitive, and neurobehavioral deficits in small mammals (Wiener et al. 2002). In addition, controlled experiments with mink (*Mustela vison*) and otter (*Lutra canadensis*) have established that dietary MeHg concentrations of 1 μg/g lead to death in less than a year. In their critical review, Wiener et al. reported numerous reproductive and behavioral effects on wild avian and mammal populations associated with ingestion of realistic

Figure 2: MeHg concentrations in predatory fish: A comparison of the USEPA and NESECP studies



concentrations MeHg. For example, neurotoxic effects from sublethal exposure of eggs to MeHg can result in reduced prey capture efficiency and competitive ability of grayling (*Thymallus arcticus arcticus*) three years after exposure. MeHg content of eggs is strongly related to concentrations of the maternal fish. Tan (2003) attributes reproductive effects, based on gender, to disruption of endocrine systems by MeHg. Kamman and Burgess (2004) suggest that, in the northeastern United States, the widespread occurrence and mercury uptake characteristics of brook trout (*Salvelinus fontinalis*) and particularly yellow perch (*Perca flavescens*) may make them preferred subjects for monitoring of mercury impacts on natural systems.

While the scientific community has focused on affected salt and freshwater aquatic processes and species, wild piscivorous birds and mammals receive a greater exposure to mercury than any other receptor (USEPA 1997d). Few studies have addressed impacts on these species (Haines 1996; USEPA 1997d). Wiener et al. (2002) reported impaired reproduction in wild merlins (*Falco columbarius*), common loons (*Gavia immer*), wood storks (*Mycteria americana*), and common terns (*Sterna hirundo*) associated with ingestion of aquatic prey exhibiting elevated body burdens of MeHg. Eisler (1987) established a Lowest Observed Adverse Effect Concentration (LOAEC) of 0.1 µg/g wet weight for MeHg contaminated food fed to mallards (*Anas platyrhynchos*). Wiener et al. (2002) suggested that mercury in feathers is an indicator of mercury in other avian tissues, and Evers et al. (1998) indicated that common loons can reduce their body burden during the winter molt. Evers et al. (2003) found that common loon egg volume declined significantly as egg-Hg concentrations increased, while mercury levels in common loons were higher at eastern North American sites relative to western. In general, common loon eggs appear to be suitable indicators of MeHg availability on lakes with territorial pairs. Gerrard and St. Louis (2001) also reported adverse impacts on reproductive success of wild tree swallows (*Tachycineta bicolor*) associated with elevated aquatic MeHg levels.

Data reported for six terrestrial species rated relative rank of exposure as: Kingfisher > otter > common loon = osprey = mink ≥ bald eagle, setting reference doses for MeHg in avian and mammalian wildlife at 21 and 18 µg/ kg body weight per day, respectively (USEPA 1997d). Table 2 lists the percent of species range overlapping with regions of high mercury deposition and a generalized wildlife criterion, defined as the concentration in water that, if not exceeded, protects avian and mammalian wildlife taken from these waters. It should be noted that mercury in birds and mammals is considered to be almost exclusively in the form of MeHg.

Table 2. Impacts of methylmercury on selected bird and mammal species' ranges, with the wildlife criterion as measured in water and body tissue for each species. Data from USEPA (1997d).

Species	Percent of Range Affected	Wildlife Criterion in water (pg MeHg/L)	Kg body wt. / Kg ingested / day
Kingfisher *Ceryle alcyon*	29%	27	0.50
Mink *Mustela vison*	35%	57	0.22
Loon *Gavia immer*	40%	67	0.20
Osprey *Pandion haliaetus*	20%	67	0.20
River otter *Lutra canadensis*	38%	42	0.16
Bald eagle *Haliaeetus leucocephalus*	34%	82	0.11
Florida panther *Puma concolor coryi*	100%	NA	NA

Gnamus et al. (2000) linked mercury deposition on terrestrial plants to food intake by roe deer (*Capreolus capreolus*). A Florida panther (*Puma concolor coryi*), thought to have died of MeHg poisoning, was found to have a concentration of total mercury in its liver of 110 μg/g (Roelke 1990). Due to the numerous uncertainties involved in field experiments, more studies are needed for definitive determinations regarding effects on wild fish-eating birds and mammals.

Hopefully, the decrease in overall body burdens of mercury in Wisconsin's adult common loons from 1992-2000 (Fevold et al. 2003) represents the effectiveness of recent emission reductions associated with more stringent SO_2/acid rain regulations for coal-fired utilities.

Emission Controls

Mercury control strategies acceptable to the regulated community have been difficult to establish because of both the political issues involved and the uncertainties in the emission inventories of mercury species (Bruss 2003). However, as a result of more recent understanding of the sources and impacts of mercury deposition, USEPA, Canada, and several states have sought tighter controls on emission sources (WDNR 1999; EIA 2001). After a long and contentious review process, USEPA released its final Clean Air Mercury Rule on 15 March 2006, instituting a cap and trade program for coal and oil based power plants, over the protests of critics who sought the more stringent Maximum Achievable Control Technology standard for those sources.

As previously stated, mercury plume speciation varies greatly relative to source category (Round et al. 1998), and is an important factor in the control of mercury emissions (Laudal 2001). Coal plume mercury speciation at the inlet of particulate control devices was found to depend on the inlet temperature and the chlorine and calcium content of the coal burned (Senior 2001). For example, emissions of chlorinated HgII compounds (e.g., mercuric chloride) can be captured with particulate control devices at high efficiencies (Laudal 2001; Senior 2001), while calcium within coals can reduce the beneficial oxidizing effect of chlorine (Benson 2003). This is evident in tests performed on western coals that are low in both chlorine and calcium. However, Hg^0, representing the dominant species emitted, is not well controlled by scrubbers or other particulate control devices (UNEP 2002). Finally, Hg^p species are obviously well controlled by particulate controls.

A reading of EPRI (2004) and USEPA (1997e) suggests mercury control strategies for coal-fired sources can be divided into two general categories: (1) Fuel controls, including the burning of high ranking (high Btu) coal, burning low mercury coal, coal washing (which may reduce mercury levels by 50%), and use of alternative fuels such as natural gas; and (2) Flue gas controls, of which there are currently two leading contenders. Flue gas controls include activated carbon injection (ACI), followed by a particulate collection device, or a compact baghouse (fabric filters) added after use of an electrostatic precipitator (ESP) (EPRI 2004). A combination of these flue gas control technologies, employing ACI between the baghouse and the ESP, has been shown to be most effective under some circumstances. However, mercury removal efficiencies from the burning of lignite coal were shown to be poor (0-9%) for both ESPs and fabric filters (Sjostrom et al. 2004). As a general rule, traditional SO_2 controls also remove some Hg^{II}. The nature of mercury control strategies is too complex to be comprehensively reviewed here. For a more thorough review, inclusive of multiple source categories and technologies, see USEPA (1997e).

Conclusion and Future Research Needs

The impacts and dynamics of mercury within natural systems are still not fully understood. Recommended areas for future research that could productively contribute to this understanding include:

- Effects of soil disturbance and wildland fire on translocation of mercury relative to watershed conditions.
- Physiological and behavior effects of realistic levels of MeHg on wild populations of birds and mammals.
- Effects of sulfate deposition on in-situ mercury methylation rates under various watershed conditions.
- Relationship between acid deposition and mercury methylation rates.

For many years, comprehensive program integration has been a Forest Service goal, whether in the collection and management of data, as with recent developments within and linkages between multiple databases (e.g., the Natural Resource Information System, NRIS, and the Forest Service infrastructure inventory database, INFRA), or in the implementation of project-level management decisions. The avoidance or abatement of mercury levels in wildlife species offers an opportunity to more completely reach that goal. The following recommend actions, if implemented, could provide forest managers involved in the fields of air resources management, wildlife management, recreation, timber management, and watershed improvement, opportunities to better understand the impacts and dynamics of mercury within the natural systems they manage:

1. Monitor mercury levels in water and wildlife, as linked to implementation, effectiveness, and validation monitoring associated with various forest land management activities (e.g., management of sedimentation associated with timber harvest).

2. Rank wetlands and lakes as to their potential for elevated MeHg production rates.

3. Minimize water level manipulations of lakes and wetlands to minimize mercury methylation rates within aquatic sediments.

4. Minimize sedimentation associated with land disturbance activities.

5. Manage fish species relative to risk factors associated with proximity of mining operations, lake morphology, and landscape ecology (e.g., avoid managing for top predators in areas of high risk).

6. Comment to state and federal air regulatory officials to maximize controls on industrial mercury and sulfate emission rates near forest boundaries.

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