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Perspectives

Mercury cycling and human health concerns in remote ecosystems in the Americas

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Abstract

Fish constitutes a praised—and sometimes irreplaceable—component of the daily diet for numerous communities throughout the Americas. These populations can be exposed to mercury (Hg)—ubiquitous in fish flesh—to levels potentially harmful to their health. Mercury is subjected to various chemical transformations once released to the environment. From diffuse loading on land and waters to accumulation in the aquatic food web, this paper draws a general picture of the main processes influencing the Hg cycle in different ecozones. Contrarily to other pollutants, the amount of Hg found in fish tissues is not clearly related to the extent of Hg loading in a given ecosystem. In the Arctic, the sudden stripping of gaseous elemental Hg from the atmosphere—known as the Hg depletion events—and its subsequent transformation into methylmercury (MeHg) seems to be one of the driving force behind Hg accumulation in freshwater aquatic organisms and marine mammals. In boreal environments, numerous watershed-based processes influence the transport and methylation of Hg deposited from atmospheric source and selectively control the amount of MeHg transferred from source to fish. In the tropics, small-scale gold mining operations have induced the release of considerable amount of Hg to the environment. However, the extensive deforestation of the tropical forest and the subsequent erosion and lixiviation of land-deposited Hg down to tropical rivers appears to have a greater impact on the observed wide-scale fish Hg contamination.

Mercury accumulation in remote ecosystems do represent a significant and complex environmental issue, considering: (a) the high levels of Hg monitored in fish and marine mammals flesh; (b) the large-scale occurrence of such worrying Hg bioaccumulation patterns; (c) the extensive use of aquatic resources by numerous remote communities; (d) the fact that this issue shall persist at least on the mid-term, even if strong measures are taken to lessen Hg anthropogenic loadings to the global atmosphere. Although there are no simple answers to the potential threats to human health caused by Hg accumulation, several political actions to lessen human exposure to Hg are possible and are discussed here.

Keywords: Mercury, methylmercury, fish consumption, land use management, mercury depletion events

TABLE OF CONTENTS

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1. CONTEXT

Several populations living in remote areas of the Americas depend upon small-scale fishing activities for meeting their protein needs. These populations comprise for example subsistence fishers from First Nations communities of the Canadian boreal forest and the Canadian Arctic, sports fishers frequently using the fish resource of lakes and rivers not far from their homes, and riparian communities settled on the shores of large watercourses in the Amazon. For these people, fish and/or marine mammals constitute irreplaceable components of daily meals, either in response to traditional values, limited availability of other protein sources and/or economic dependency. Many of these populations enjoy simple life styles, in harmony with the relatively pristine environments in which they evolve, far from direct industrial contamination point sources. Despite this fact, several scientific evidences prove that the health of these communities might be threatened by high levels of methylmercury (MeHg) in fish and marine mammal tissues.

Fish eating communities are often not aware of the dangers related to MeHg, or even of its existence in their milieus. The sole concept of "contaminant" is frequently missing from First Nations regional languages and dialects. Moreover, the presence of mercury (Hg) in the global ecosystems is ubiquitous, following its emission to the atmosphere—mainly from coal combustion, waste incineration, ore refining or from natural sources such as volcanoes or wind erosion—its transport and diffuse deposition on lands and water plans, its transformation into MeHg occurring in wetlands and aquatic systems, and its accumulation throughout the aquatic food chain (ICMGP, 2006; Gbor *et al.*, 2007, Munthe *et al.*, 2007). In this context, it is difficult to properly inform and arm inhabitants from isolated communities about the Hg threat, especially since these people are usually not responsible for the pollution.

The environmental and health impacts in remote ecosystems of global Hg cycling constitute a concrete example of environmental injustice and have focused the international attention on the following questions:

A. Does Hg accumulation in remote North American ecosystems have an impact on human health?
B. Can the extensive gold mining activities in remote tropical regions of South America constitute a potential time bomb, threatening the health of numerous riparian populations, far away downstream from gold extraction sites?
C. How could these situations evolve and what could be done to protect the health of fish eating populations?

As a result, scientific efforts have provided a general picture of Hg pathways from the Brazilian tropics to the Canadian Arctic. This paper synthesizes the main pathways undertaken by Hg in its journey from skies to fishes through specific regional case studies and presents pieces of answers toward lessening exposure to MeHg through fish consumption endured by remote populations relying on local fish resources. These examples clearly demonstrate that, for a given ecosystem, the Hg loading is often not strictly correlated with the local/regional contamination of the biota by MeHg. In fact, the environmental characteristics and human uses of aquatic ecosystem, often dictate its overall response (or sensitivity) to various levels of Hg loadings. In turn, mercury contamination of the ecosystems, monitored by the accumulation of mercury in top predator fish species, can lead to health deteriorations. Related impacts on human health depend on the economic/social/traditional dependency of communities to the fish resource, on exposure to other persistent bio-accumulative contaminants and on metabolic ability to handle xenobiotics (defined here as the human vulnerability to Hg). These multiple parameters provide partial answers on how exposure to MeHg can be lessen in remote populations relying on local fish resources.

2. EXPOSURE TO METHYLMERCURY AND HUMAN HEALTH

The formal identification of the clinical symptoms induced by MeHg poisoning was made at the end of the 1950s, following an episode of wide scale environmental contamination that occurred in the city of Minamata, Kyūshū district, Japan. For more than 30 years, the Chisso Corporation, a chemical factory located in the middle of the city, dumped its untreated wastewaters in the Minamata Bay, including several tons of Hg, which was used as a catalyst for the production of acetaldehyde. The symptoms of MeHg intoxication include ataxia, numbness in the hands and feet, muscle weakness, narrowing of vision, loss of cognitive functions and coordination of movements (NRC 2000). Extreme exposure to MeHg in Minamata also provoked numerous cases of insanity, paralysis, coma and the death of more than 900 community members.

With the exception of major disasters or spills, Hg usually does not produce dramatic immediate health effects, but slow insidious changes in biological functions, reflecting diminishing well-being and affecting the quality of life (Clarkson, 1990; NRC, 2000; Grandjean, 2007). Evidence of physiological changes associated with exposure to low dose of MeHg were reported by extensive studies carried out in the Brazilian Amazon (Lebel *et al.*, 1996 and 1998, Mergler, 2002) and among children from the Faroe Islands (Grandjean *et al.*, 1997; Murata *et al.*, 1999; Sørensen *et al.*, 1999, Weihe *et al.*, 2003; Debes *et al.*, 2006). These early health deteriorations include partial narrowing of the visual field, loss of manual dexterity, loss of sensitivity in hands and feet, partial loss of color perception, alteration of cognitive functions and learning deficiency at childhood (NRC 2000). Early health alterations constitute not only a warning for increased risk of eventual illness, but, more important, reflect diminished well-being and functional capacities. This can have dramatic effects in situation of self-sustained economy, particularly when coupled to poverty, malnutrition, poor living conditions, inadequate health care and schooling.

Human exposure to MeHg in remote populations of the Americas derives primarily from the consumption of freshwater and marine fishes, or marine mammals, all important sources of many essential elements for these people and an integral part of their culture and economy (Wheatley and Paradis, 1996, Van Oostdam *et al.*, 1999, Muckle *et al.*, 2001; Mergler *et al.*, 2007). In all cases, the extent of exposure to Hg greatly depends on the specific fish species and marine mammal consumption patterns. The fish species, size and consumption frequency are key parameters when it comes to determining the acceptable limits for steady-state exposure (ALSSE) to MeHg (e.g. 0.1 to 0.5 ug.kg $bw^{-1}.day^{-1}$). This is an important issue in aboriginal gill-net based fisheries, as well as in recreational—or economy-related fisheries which tend to target the larger specimens of piscivorous species, those usually bearing the highest Hg levels, for regular consumption. ALSSE can be exceeded by eating only two meal per month of northern pike (typical Hg level of 0,7 to 1,0 ppm) caught in Northern Québec, or by eating one single meal of beluga muscle (1,0 to 1,5 ppm Hg)—greatly praised by the Inuit communities. Let's now look at the main pathways leading Hg, from atmospheric sources to fish and marine mammal tissues.

3. MERCURY SOURCES AND CYCLING IN NORTHERN ECOSYSTEMS

It is widely admitted that, with the exception of specific geological features or local contaminant point sources, the most important source of Hg to Northern environments is the global atmospheric pool. Even though the atmospheric Hg loading is diffuse and occurs at low rates in these regions, numerous boreal and Arctic ecosystems can be described as Hg-sensitive environments because they efficiently: (1) transform the Hg atmospheric load into bioassimilable MeHg; and (2) biomagnify MeHg along food webs, yielding contaminant levels in top fish predator species and/or marine mammals unsafe for regular human consumption. It is possible to distinguish between two major processes that drive the Hg burden from the atmosphere down to aquatic ecosystems. While airborne chemical reactions seem to dominate Hg deposition patterns in the Arctic (figure 1), land processes appear to be of greater importance in the boreal ecozone (figure 2).

3.1. ARCTIC ENVIRONMENTS: THE MERCURY DEPLETION EVENTS

The chemical speciation of Hg in the atmosphere is rather complex. One can distinguish between two main categories according to the oxidation state of the Hg molecules: Hg^0 (or Gaseous Elemental Mercury—GEM) and HgII (or Reactive Gaseous Mercury—RGM) species. Several recent observations, made in the Arctic and sub-Arctic atmosphere, point to a cyclic depletion of GEM levels that corresponds with enrichment in RGM, which then also rapidly vanishes from the atmosphere. The most plausible explanation for this phenomenon, known as Mercury Depletion Events (MDE), is as follow:

The periodic appearance of polar sunrise provokes the evaporation of bromine species from saltwater. Bromine then reacts with atmospheric ozone to form bromine oxide (BrO), which can transform GEM species into RGM, readily stripped from the atmosphere (Boudris *et al.*, 2000; Ebinghaus *et al.*, 2002, Skov *et al.*, 2004; Lindberg *et al.*, 2002):

$$Br_{(dis)} \xrightarrow{h\nu} Br_{(gas)} + O_3 \longrightarrow BrO + O_2 + GEM \longrightarrow RGM$$

This process is thought to deposit between 90 to 450 tons Hg per year on Arctic and sub-Arctic environments (Poissant *et al.*, 2008). The MDE have drawn much attention from the scientific community with over 200 papers published on the issue during the past decade (Poissant *et al.*, 2008). Since the mechanisms behind the MDE were clearly identified, other studies demonstrated that the majority of the MDE- deposited Hg through MDE is evaporated back to the atmosphere under the form of GEM through the action

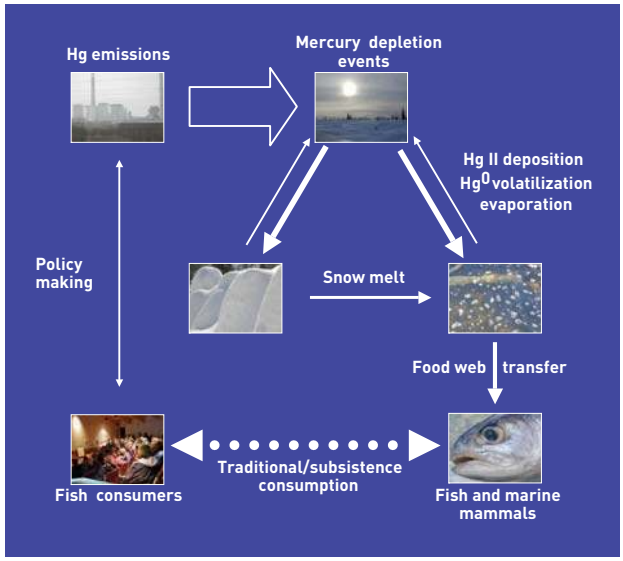

Figure 1. Mercury cycling in the Arctic

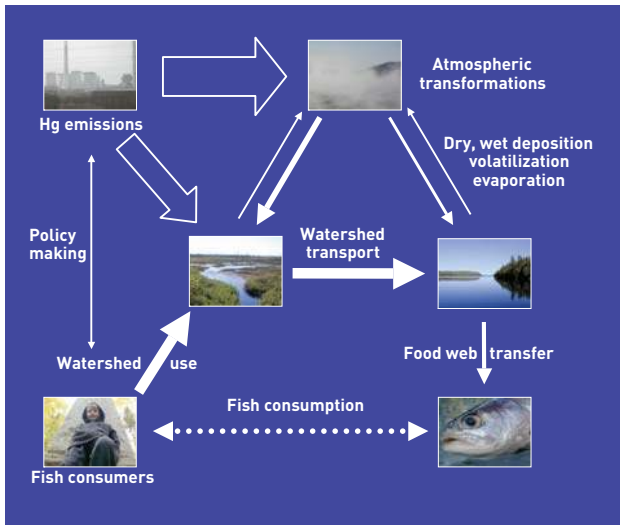

Figure 2. Mercury cycling in boreal environments

of sunlight (Ariya *et al.*, 2004; Skov *et al.*, 2004; Lindberg *et al.*, 2007; Poissant *et al.*, 2008). The remaining deposited RGM burden appears to be available for assimilation by the freshwater biota, once carried to Arctic lakes by snowmelt and runoffs (Scott, 2001; Ariya *et al.*, 2004). Mercury trapped in Arctic freshwater lakes following MDE and snowmelt, is thought to induce of Hg-related metabolic abnormalities such as lower testosterone levels and dysfunctions of reproductive capabilities observed among fathead minnows and northern pikes cohorts (Hammerschmidt *et al.*, 2002, Drevnick and Sandheinrich, 2003).

This picture somewhat differs for the Arctic saltwater environments. Outridge *et al.* (2008) reported that on a yearly basis, the total Hg flux entering the Arctic Ocean from all sources—MDE, wet Hg deposition, coastal erosion—is almost in balance with the total Hg mass removed from the ocean through evaporation, sedimentation and seawater exports, with a small yearly increase of 0,3% of the total Hg mass. This small net Hg gain by the Arctic Ocean may be enough to explain the progressive increase of the MeHg levels in the marine biota during the 20th century. However, this fragile balance will probably be modified by climate change which is likely to induce: (1) increased coastal erosion, river outflow and Hg transport; (2) changes in the ocean's biological productivity which might affect Hg bioavailability and MeHg bioaccumulation. (Outridge *et al.*, 2008; Poissant *et al.* 2008). The outstanding capacity of Hg to concentrate from one environmental compartment to the other is well known. This is particularly true in the case of complex saltwater food webs. For example, biomagnification factors as high as 380 were reported between ringed seal and its usual food source, zooplankton (Dehn *et al.*, 2005).

It is difficult to state on the origin of bulk atmospheric Hg brought to the Arctic but recent work suggests that about one third or more of the current Hg emissions to the atmosphere is from direct anthropogenic source (Lindberg *et al.*, 2007). However, according to long term monitoring programs measuring atmospheric Hg levels in various locations of the Arctic, it is still difficult to identify a clear trend pointing to a recent decrease in the levels of total atmospheric Hg, despite the numerous efforts to diminish man-made Hg emissions by the industrialized countries of the World.

3.2. BOREAL ENVIRONMENTS: ATMOSPHERIC DEPOSITION AND LAND MANAGEMENT

Several fish communities of boreal lake ecosystems also exhibit Hg levels unsafe for frequent human consumption. However, the route between Hg atmospheric deposition processes and receptor lakes somewhat differs in boreal environments, compared to Arctic freshwater lakes. Here—and except for seepage lakes—almost 90% of the total Hg load to lake waters generally transit through the watershed before reaching the lakes. Thus, the link between the extent of Hg atmospheric deposition and its bioaccumulation along aquatic food webs—or the specific ecosystem' sensitivity to Hg—rather depends on the following (see also Wiener *et al.*, 2006; Munthe *et al.*, 2007):

1. The characteristics of specific watersheds enabling an efficient transport of deposited Hg—through runoffs and erosion—from upland to methylation sites (riparian wetlands and peat lands, lake sediments).
2. The ability of specific ecosystems to efficiently transform this Hg burden into biologically available MeHg. Natural background MeHg levels do greatly vary, even within sub-regional scales. Environmental factors such as low pH, the presence of riparian wetlands, the extent of photochemical processes, high organic carbon and nutrients loadings, high biological activity, and elevated ratios of drainage basin to lake area, are favourable for MeHg production and therefore tend to yield fish with higher MeHg burdens (Wiener *et al.*, 2006; Munthe *et al.*, 2007).
3. The structure of aquatic food webs which also modulates MeHg bioaccumulation and biomagnification. Variation in invertebrate assemblages, food webs length and structure, and fish growth rates can alter the burdens of MeHg in prey and predator fish species (Parkman and Meili, 1993; Montgomery *et al.*, 2000; Gorski *et al.*, 2003; Kainz *et al.*, 2003; Simoneau *et al.*, 2005).

All these factors are largely independent of the extent of Hg atmospheric loading *per se*. Considering the above, it becomes clear that landscape management can have a significant impact on Hg transfer from catchments to lakes and/or methylation sites. Long-term experiments held in Sweden indicate that, while Hg and MeHg release from upland forest soils to surface waters is not readily affected by changes in atmospheric input (Munthe and Hultberg, 2004). However, forestry and other land-use changes causes increased erosion and releases of Hg and MeHg from terrestrial environments, also increasing Hg levels in zooplankton and fish (Porvari *et al.*, 2003; Garcia *et al.*, 2005 and 2007). Furthermore, bearing in mind that the residence time of Hg in boreal soils could range from decades to centuries, it is uncertain that a decrease in anthropogenic Hg emission would translates in a consecutive lowering of Hg levels in fish, on the short-term, despite international efforts to reduce anthropogenic Hg emissions to the atmosphere (See Box A). It is therefore plausible that many lakes located in the boreal region will remain, for decades, sensitive spots with respect to the presence of Hg.

BOX A – LINKING A DECREASE OF HG ATMOSPHERIC DEPOSITION AND HG LEVELS IN FISH: AN ONGOING DEBATE

High mercury (Hg) levels in fish living in lakes of the Northern Hemisphere have been associated with an increase of anthropogenic atmospheric Hg over the 20th century (Munthe *et al.*, 2007). During decades, governments have responded to this environmental challenge by implementing different strategies to assess Hg sources, to prevent the use of Hg in industrial

processes, in the confection of goods, and to regulate its emission to the atmosphere, especially through coal burning for the production of electricity (NAAEC-CEC, 2000; NESCAUM, 2000, 2003 and 2005; USEPA, 2005). These efforts initially paid off. The inventory of global anthropogenic Hg emissions from 1979/1980 to 1995 was marked by a sharp decrease in the 80's followed by stabilization afterward (Slemr *et al.*, 2003). The global Hg atmospheric pool responded accordingly but with a small time lag. Measurements of atmospheric Hg levels and deposition rates showed a general increase in the 80's, followed by a decrease in the early 90's; and a stabilization afterward (Slemr *et al.*, 2003; Kim *et al.*, 2005; Vanarsdale *et al.*, 2005).

The residence time of Hg in the global atmospheric pool is thought to be between several months and one year (ICMGP, 2006; Munthe *et al.*, 2007). It is thus not surprising that signs of changes to the atmospheric Hg burden are perceptible on the short term following cuts in emissions. These values are orders of magnitude shorter than the values reported for the residence time of Hg in soils which can extend from decades to centuries (UNEP, 2002).

The pathways followed by Hg to enter lakes greatly differ according to the geomorphology of the aquatic ecosystems. As seepage lakes receive most of their Hg load directly from the atmosphere, they are expected to react promptly to changes in atmospheric deposition rates. Observations to that regard were already presented by Hrabik and Watras (2003). However, direct atmospheric Hg loading to drainage lakes—e.g. lakes where most sports and commercial fisheries occur—range between 2 to 10% of the total Hg charge, the rest slowly transiting through the watersheds before reaching the lake waters (Porvari *et al.*, 2003). The forest soils covering lake watersheds constitute a tremendous pool of Hg. For instance, Grigal (2003) reported that the annual anthropogenic Hg load cumulated for 48 contiguous states of the USA (144×10^6 g) is two orders of magnitude less than the pool of Hg in the forests of those states (30 300 106 g), most of it lying in soils. Consequently, the recovery of drainage lake following Hg emission cuts on the short- to mid-term remains to be demonstrated and might have to be solely expected on the long term.

A consensus emerged among the mercury specialists attending the 8th International Conference on Mercury as a Global Pollutant—Madison (WI), August 2006—to the effect that recent efforts put toward the decrease of anthropogenic atmospheric Hg emission will eventually translate in a global decline of the Hg burden in fish. According to the Conference Declaration, the extent of this decline and the expected time frame for its occurrence will probably greatly depend on the environmental particularities of lake ecosystems (ICMGP, 2006; Munthe *et al.*, 2007). However, this general evaluation was made on past and present observation on the lake ecosystems' behaviour and atmospheric deposition regime, but it fell short of taking into account the foreseeable increases in global atmospheric Hg emission rate attributable to the strong emergence of regional economies in Asia, and to ongoing changes in environmental conditions related to Global Warming that might affect Hg methylation and bioaccumulation.

Knowledge about the resilience of lakes and fish stocks to the stress imposed by the presence of Hg remains a key issue to be used by political stakeholders to continue to impose—and even to strengthen—costly regulations on Hg emissions to the industrial sector. However, the actual scientific evidence assessing responses to changes in atmospheric Hg loading is limited, impairing the possibilities to draw firm conclusions. This fact is partly due to a lack of long-term, consistent monitoring of environmental data. It also results from the complex biogeochemical interactions of Hg, ruled by multiple chemical, physical and biological processes that contribute, besides Hg load *per se*, to the control of MeHg levels in fish. This is why conclusions issuing from sound studies on Hg pathways from the watersheds to lakes, and its assimilation and bioaccumulation in food webs, can be of conflicting nature, while all being verifiable and accurate. Innate shifts in Hg cycling and lake metabolisms (ex.: seasonal variations in loadings, atmospheric processes, methylation efficiency, nutrients availability, etc) and/or biological processes (ex.: life-cycle changes in fish feeding habits and locations, inter-lake variations of fish growth rates, etc.) through time and space might bring misleading inferences. Moreover, many of the physical and biogeochemical processes involved in the Hg methylation and transfer from forests to lakes may be dynamically affected by changes in the climate regime (MacDonalds, 2005; Schindler and Smol, 2006)

Recent data issuing from the United States and elsewhere tend to demonstrate that local cuts in atmospheric Hg emission do have a positive short-term effect on lake ecosystems in the vicinity of point sources such as coal-fired power stations. Two recent studies by Hammerschmidt and Fitzgerald (2005 and 2006) reported strong positive relationships between rates of wet Hg deposition and Hg levels in mosquitoes and large mouth bass (*Micropterus salmoides*)—a plankton eating species often found in small lakes, pounds and swamps susceptible to react promptly to Hg emission cuts—in many American states. But this relationship seems to be weakened in New England states with little coal burning activities such as Maine and New Hampshire, right under the atmospheric Hg plume issuing from the Midwest States. Elsewhere, a study by Johansson *et al.* (2001) reported a 20% average decline in Hg levels of northern pike (*Esox lucius*) from a series of remote Swedish lakes sampled at least three years apart, from 1981 to 1995. The decline was associated with a drop of roughly 50% in Hg wet deposition during the same period. However although fish Hg concentrations dropped on average for the study lakes, increases and decreases were both observed for fish Hg levels in individual water bodies. Another study from Muir *et al.* (2005) on numerous lakes of the Canadian Arctic was unable to demonstrate any trend for Hg levels in landlocked Arctic char (*Salvelinus alpinus*) during a thirteen years span started in 1990. Finally, a survey held in 176 Minnesota lakes (MPCA 2006) reported a range of trends—increases in 44 lakes, no change in 45 and decreases in 87 lakes—for Hg in predatory fish, over a time period when deposition was known to decrease (Engstrom and Swain 1997), suggesting that multiple factors likely affect fish Hg concentrations.

A last study on the potential resilience of boreal lake ecosystems following a significant reduction of Hg atmospheric emissions deserves attention. Harris *et al.* (2007) designed a whole-ecosystem experiment to test the immediate response of fish Hg level to change in Hg deposition rates. These authors artificially increased the Hg load to a lake and its watershed by the addition of enriched stable Hg isotopes. The authors were able to correlate change in fish MeHg level to change in direct Hg deposition to the lake surface, implicitly suggesting a higher reactivity of this freshly deposited fraction. However, less than 1% of the Hg isotope added to the watershed reached the lake after a period of 3 years and the transfer rate between the lake and the watershed was still not at steady state after this period.

Considering the above, the debate on the link between atmospheric Hg deposition and fish MeHg levels still needs to be firmly concluded.

4. TROPICAL ENVIRONMENTS: GOLD MINING AND DEFORESTATION

For centuries, the capacity exhibited by Hg to form amalgamates with gold led to its use to extract the precious metal. Nowadays, most large scale commercial mining operations have adopted the cyanide amalgamation process to capture gold from raw ores. However small-scale artisanal gold mining operations (AGM) still use the secular Hg amalgamation technique to extract gold from soils and sediments. It is estimated that 13 million workers in 55 countries insure their subsistence from this activity (ILO, 1999)—about 1.6 million operating in the Brazilian Amazon alone (Malm, 1998). Several studies reported elevated Hg levels in fish caught in the Tapajós and Amazon Rivers, resulting in unsafe Hg exposure situation for many riparian population mainly constituted of subsistence fishers (see Box B) (Malm *et al.*, 1995 and 1997; Lebel *et al.*, 1996, 1997 and 1998). Regional increases of fish Hg levels were formerly thought as being directly imputable to Hg releases following AGM (Lacerda and Salomons, 1998). But an extensive three year study, held in 1994-1997 along the Tapajós River basin, sheds new light on the question (IDRC, 2003). The study examined the Hg distribution in a portion of the Tapajós River, starting nearby an extensive gold mining area, then up North to 400 km downstream. The research team initially looked for Hg gradients in the different environmental compartments of the river, but found that the amount of Hg dissolved in the water, in the suspended particular matter found in the water column, in the river sediments or even in soils from the watershed were roughly the same, either 50 km from of the AGM area or 400 km downstream (Roulet *et al.*, 1998 and 2001). A simple mass balance calculation, comparing the amount of Hg found in the riparian soils and an estimation of the amount of Hg loss through evaporation and the burning of gold-Hg amalgamate, brought the authors to suggest that at most 3% of the total Hg found in the top layers of soils was attributable to AGM; the remaining soil Hg burden probably issued from long term atmospheric deposition of Hg from volcanic origins (Roulet *et al.*, 1998b and 1999). One must know that the soils of the Amazon are ancient, perhaps more than 1 million years old, and were thus subjected to Hg accumulation for millennia.

However, the Hg content of the top sediment samples retrieved along this gradient suggested that the recent sediments carried 1.5–3 times as much mercury as those deposited 40 years ago (Roulet *et al.*, 2000 and 2001). This time frame correlates well with the massive colonization that the region had to face since the beginning of the 1960's following a series of initiatives from the Brazilian National Institute of Colonization and Agrarian Reform. The reform, intended to develop new agricultural regions in Brazil, had the consequence of moving tens of thousands of families from the poorer regions of northeastern Brazil to the Amazon basin. During the first 30 years of the reform, more than 350 000 families settled in these remote regions, gaining ground on the dense tropical forest through a slash and burn deforestation strategy, inducing a loss of forested soils that peaked to about 22 000 km^2 between the 1970-88 in the Amazon region (Hudson, 1998; Van de Steeg *et al.*, 2006). These facts brought the authors to conclude that the loss of Hg from soils of the Tapajós shores, following deforestation and extensive erosion, was responsible for the high levels of Hg found in local fish species, making these environment sensitive to Hg loading and local population vulnerable to Hg exposure through their dependency toward the fish resource, which is needed to complete their diet. This hypothesis of extensive soil erosion was later supported by other studies describing the enhanced transfer of terrestrial organic carbon to the river, following extensive land use on the watershed of the Tapajós River (Farella *et al.*, 2001 and 2007). Sustainable land use management strategies could again lead to an improvement of the Hg situation in this region.

BOX B – MERCURY AND GOLD MINING

The exposure patterns to Hg consecutive to small-scale gold mining (AGM) activities are multiple and somewhat differ from situations encountered in remote communities of the Northern Hemisphere. The Hg exposure experienced by traditional miners can occur either through direct handling of inorganic Hg or by breathing the toxic vapors generated during the burning of the gold-Hg amalgam. In many cases, heat separation of gold is achieved in houses, exposing worker's families to elevated levels of gaseous Hg. Mercury, under the metallic (liquid) form, is not significantly retained or transformed by the human digestive system and, when ingested under this form, it is almost completely excreted in the feces with little toxic damages to the organism (Rowland *et al.*, 1977; WHO, 2003). Similarly, skin contact with liquid mercury does not enable its entrance into the body, but rather translates in mild symptoms such as skin irritation, dermatitis or cutaneous eruptions. However vapors of elemental Hg are efficiently absorbed through the lungs and then distributed through the body. Elemental Hg can then be transformed into chemical species (Hg (II) complexes) that can accumulate in various tissues, especially the kidney and brain. Once in the body, these compounds alter the action of enzymes, and can cause serious damages to natural nervous functions if exposures are high (Jones, 1971). Vapor inhalation is generally the most important and

dangerous pathway of exposure to metallic Hg for traditional gold miners and their families. This is also true for gold dealers (and the people inhabiting in the vicinity of "gold houses"), who generally operate their business in more urban areas, purchasing amalgams from traditional miners and/or refining gold pellets still containing appreciable amounts of Hg, often in closed rooms without proper ventilation (UNIDO, 2004).

But, the inorganic Hg released by AGM can also be efficiently methylated in tropical sediments and floodplains (Roulet *et al.* 2000b; 2001b), then transferred to the aquatic food web, up to edible fish species. Although disagreement exist on the regional scope of the environmental contamination attributable to AGM, local increases in the Hg levels of fish tissues have been reported in studies dealing with this issue, also enhancing the potential exposure of communities directly involved in AGM activities, as well as those simply living in the vicinity, through their fish consumption habits (Malm *et al.*, 1995 and 1997; Van Straaten, 2000 and 2000b; Limbong *et al.*, 2003; Kambey *et al.*, 2001, IDRC, 1998 and 2003).

Mercury releases by small-scale mining occur as liquid Hg lost to the aquatic environments during the amalgamation process, and vapors entering the atmosphere. It is difficult, even locally, to evaluate the amount of Hg emitted in the environment due to AGM. Mercury losses during AGM operations largely rely on the amalgamation technique used and on the way gold is separated from the amalgam, either through the use of nitric acid, retorting or burning in open pans (UNIDO, 2004). If retorts are not used, the greatest part of the Hg introduced in the amalgamation process is released to the atmosphere. The most environmentally damaging approach to create amalgamates consist in placing Hg on sluice boxes or spreading it on the ground to put it in contact with raw grinded ore and "attract" gold, while losing a significant portion of this metallic Hg to rivers and lakes. A cleaner and more efficient approach consists of amalgamating only gravity concentrates, enabling to extract up to 90% of the initial gold content. Then excess Hg is removed through centrifuging or simply using a piece of fabric. Such amalgams have Hg contents between 20 and 40% (UNIDO, 2004).

Many of the available estimates on environmental Hg releases attributable to AGM are based on regional Hg sales, but these numbers do not take into account either Hg recycled or bought on alternative markets. Even given the uncertainties related to these estimates, AGM represents one of the most important sources of Hg to the global environment attributable to human activities. For example, about 150 metric tons of Hg were released yearly to the environment in the Brazilian Amazon between 1979 and 1994 (Malm, 1998).

5. FRESHWATER RESERVOIRS: MERCURY HOT SPOTS THROUGHOUT THE AMERICAS

There are close to 10 000 large dams in North, Central and South America, build either to secure freshwater supply for domestic use and irrigation, or to produce electricity (WCD, 2000). These artificial lakes are attractive to populations, firstly because they often represent the only water plan available in the region, or because fish abundance and size is higher than those of surrounding lakes. Many of them are located on traditional hunting and fishing lands exploited by First Nations subsistence fisher communities.

Most studies dealing with the environmental impacts of reservoirs converge on the fact that the flooding of terrestrial ecosystems consecutive to impoundments lead to significantly increased Hg levels in fish species living in the newly created water plan (Morrison *et al.*, 1995; Rogers *et al.*, 1995; Park and Curtis, 1997; Kehrig *et al.*, 1998; Bermudez *et al.*, 1999; Lucotte *et al.*, 1999; Tuomola *et al.*, 2008). In many cases, these increases translate in fish MeHg levels unsafe for regular human consumption (Lucotte *et al.*, 1999; Nino *et al.*, 2003). The creation of reservoirs favors the recycling of the Hg burden accumulated for years in soils prior to flooding. Reservoirs also act as efficient incubators for Hg methylation (Lucotte *et al.*, 1999; Gray and Hines, 2008). Nutriments and particles leached from soils through the flooding process increase the biological productivity of these artificial aquatic ecosystems, including higher activities of bacterial consortiums involved in the transformation of Hg into bioavailable MeHg (Montgomery *et al.*, 2000). Furthermore, draw down zones which did not suffer extensive erosion, periodically flooded and dried out, typically represent environments where efficient Hg methylation can occur. Increased Hg levels in fish tissues can be detected 15 to 40 years after the initial flooding episode, depending on the type of environment impacted by flooding and on the fish species considered (Schetagne and Verdon, 1999; Nino *et al.*, 2003). Reservoirs must be regarded as sensitive environments of specific concerns for the Hg issue.

6. THE HUMAN COMPONENT: HEALTH AND PERSPECTIVES

Scientific literature does not rule out that pathogenic Hg levels in fish can also be solely the results of background (natural) Hg loading in certain lakes. Moreover, the worldwide geographic distribution of major anthropogenic Hg sources to the atmosphere has greatly evolved in the last part of the 20th century. The reported stabilization (see Box A) of the measured atmospheric Hg levels—despite efforts to decrease the emission in the industrialized countries—could be consecutive to the emergence of important new foreign sources of atmospheric Hg. In 2000, Asian countries contributed for about 54% of the global Hg anthropogenic emissions and China alone was responsible for 28% of this pool (Pacyna *et al.*, 2006). The needs of China for electricity, which is nowadays produced at over 75% through coal combustion, is expected to increase by roughly 75% by 2020, from the 2010 levels (Han *et al.*, 2000; Compton and Wu, 2005; EIA, 2006) and the overall projections for Hg atmospheric loading are not reassuring. Furthermore, the industrialization of our societies has brought other contaminants—such as PCBs and other synthetic organics—to pristine environments, which enhance the risks to health consecutive to eating fish.

In the mean time, remote communities will continue to rely on fish as a food source and they will need information to find the equilibrium between "exposure to Hg through fish consumption" vs. "land use changes in response to economic stress causing increased Hg transfer to aquatic ecosystems" vs. "health benefits related to a balanced diet, centered on fish consumption". The challenge of identifying a safe level of fish consumption is not limited to the people of remote or aboriginal origins. But it is exacerbated by the need to elaborate solutions that respect traditions and are applicable to contexts where dietary alternatives are not available, or might even create other health deteriorations: For example in the Cree communities of Northern Québec, a drastic shift from a fish-based diet to less nutritive market food—in response to information campaigns on Hg fish contamination following the creation of hydroelectric reservoirs—led to the appearance of numerous cases of diabetes and other acquired diseases (Dannenbaum *et al.*, 2008).

The establishment of so-called safe levels of exposure to Hg remains by itself a complex task. Actual guidelines are often contested and constantly reviewed in light of continuing advances on Hg toxicological effects and their threshold levels, increasing confusion among fish eating populations. Available guidelines on fish consumption do take into account specific risks encountered by more sensitive groups such as pregnant women or developing children. However, it was recently proposed that some of the proxies used to construct these recommendations are either not universal or could be misleading (Stern, 2005; Canuel *et al.*, 2006; see Box C). Once fully deciphered, these findings could modulate our perception and handling of the Hg issue.

Besides the development of sustainable land management strategies, other tools are proposed to remote communities to help them develop mitigation strategies and to lower their exposure level to this contaminant. For instance, it was demonstrated that proper fishing pressures imposed to boreal lakes might lead to lower Hg levels in the tissues of certain predator fish species. Lower competition for food following fish exploitation favors faster growing rates among the remaining fishes, and biodilution of their dietary Hg burden. Selective fish consumption strategies leading to a diversification of the fish size ("eat the smaller fish") and species ("don't eat the fish that eats fish") consumed can also lead to decreased Hg intake. But these elements of solutions, in order to be effective and socially acceptable, must be developed, understood, desired and implemented by (and with) the communities, while in harmony with their traditions.

BOX C – VALIDITY OF THE BIOMARKERS OF HUMAN EXPOSURE TO DIETARY MEHG

A constant and linear relationship is reported to exist between the amount of MeHg ingested from dietary source and the resulting levels of MeHg in hair and blood. Therefore hair and blood Hg levels are considered valid proxies of dietary exposure to MeHg in most epidemiological studies. These two biomarkers were also used as benchmarks for the establishment of most recommendations and guidelines on fish consumption and tolerable Hg daily intake (WHO, 1990; USEPA, 1997; NRC, 2000; FAO, 2003). However, a recent study by Canuel *et al.* (2006) reported for the first time significant intercommunity differences between the levels of MeHg dietary intake and these biomarkers of exposure, suggesting a differential in the absorption efficiency of dietary MeHg by the gastro-intestinal tract for certain communities (First Nation communities located in the province of Québec, Canada). Using detailed dietary surveys and in situ fish mercury determinations, the authors were able to precisely quantify the Hg exposure of these populations and observed a 10-fold difference between the measured hair Hg levels of the participants to the study, and the hair Hg levels calculated according to the accepted toxico-kinetic models, given their measured Hg intake. Although more research is needed to elucidate this finding, the authors proposed that genetic polymorphism affecting the GSH expression/recycling/binding to MeHg may be at the source of this observation. These results constitute a groundbreaking breakthrough in the current knowledge of the relation between Hg in fish and human exposure, and may lead to revisit the uniformed actual guidelines on safe fish consumption frequency.

Another biomarker of human exposure to Hg used to delineate guidelines on safe Hg intake was recently contested by new scientific evidence (Stern and Smith, 2003; Stern, 2005). To calculate a reference dose for exposure to Hg for pregnant women, the United States Environmental Protection Agency used a ratio between cord blood/maternal blood of 1. However, the cord blood Hg level could be as much as 70% percent higher and therefore, the actual tolerable daily Hg intake suggested by USEPA could underestimate the risks of abnormal fetus development by a factor of 2 to 3 (Stern, 2005; Trasande *et al.*, 2006).

6. CONCLUSION

Although there are no simple answers to the potential threats engendered by the presence of Hg in remote environments, it is clear that the comprehensive portrait of the subject as depicted is this paper, provides policy makers and community leaders with unique guidance on future ways to handle this important matter. Mercury accumulation in remote ecosystems do represent a significant and complex environmental issue, considering: (a) the high levels of Hg monitored in fish and marine mammals flesh; (b) the large-scale occurrence of such worrying Hg bioaccumulation patterns; (c) the extensive use of aquatic resources by numerous remote communities; (d) the fact that this issue shall persist at least on the mid-term, even if strong measures are taken to lessen Hg anthropogenic loadings to the global atmosphere. According to evidences presented here, actions should be envisioned at three levels:

1. Political interventions on: (a) the development and implementation of landscape management strategies that preserve forests and prevent extensive soils erosion

toward aquatic ecosystems; (b) the continuing implementation and strengthening of actual legislation on Hg emission control, especially at the international level, guided by the precautionary principle. Despite the facts presented before, long-term improvement of the Hg situation might be expected following significant decreases in Hg atmospheric loadings. It is also worth noting that actual control technologies limiting atmospheric Hg releases solely target RGM, and should be improved to efficiently remove all Hg species and counteract processes such as the MDE.

2. Educational local/regional interventions directed to: (a) inform communities at risk of higher Hg exposure on the potential threat to their health and on the avenues offered to them to lessen these risks, while maximizing the nutritional benefits related to fish consumption; (b) empower populations towards a sustainable handling of their own environment.

3. Continuing scientific progress on key critical gaps of our collective knowledge of the Hg cycle in the environment, metabolic pathways in the human body and fine-scale toxicological effects. Such progress can only be achieved through multidisciplinary approaches applied at the ecosystem level.

GLOSSARY

AGM: Small-scale gold mining; **ALSSE:** acceptable limits for steady-state exposure; **$Br_{(dis)}$:** Dissolved Bromine; **$Br_{(gas)}$:** Gaseous Bromine; **BrO:** Bromine oxide; **GEM (or Hg0):** Gaseous elemental mercury; **hv:** Sunlight; **Hg:** Mercury; **ICMGP:** International Conference on Mercury as a Global Pollutant; **MDE:** Mercury depletion events; **MeHg:** methylmercury; **O_3:** Ozone; **PCBs:** Polychlorinated biphenyls; **ppm:** Parts per millon; **RGM (or HgII):** Reactive gaseous mercury; **ug.kg bw^{-1}.day^{-1}:** Micrograms per kilogram of bodyweight per day

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