# Mercury Contamination in Aquatic Ecosystems: an Analysis of the Critical Areas

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#### Introduction

**WIRONMENTAL** pollution worldwide is an undesirable by-product of the increased demand for natural resources in modern civilization. However, since the advent of human societies, there have always been foci of environmental contamination, though nothing on the scale we see today. Practically all of the world's environments suffer from some degree of contamination. Even such remote areas as the Antarctic present concentrations of contaminants like mercury and lead, distributed on a global scale in the atmosphere, in concentrations above those expected for the region. And so the main question now asked by environmental agencies is not whether an environment is contaminated, but how severe the contamination is.

In general the pollutants that cause the most damage to the ecosystems can be sorted into two main groups. The first comprises those substances present in the effluents produced by large urban centers, especially those associated with the improper dumping of solid waste (garbage) and the inadequate or non-existent treatment of sanitary wastewater. Aquatic environments, such as rivers, estuaries and coastal regions, are those most severely affected by the contaminants generated by these sources. This group includes organic matter, which results in heightened biochemical demand on oxygen in aquatic ecosystems, and nutrient overload - particularly of nitrogen and phosphorous -, which causes the proliferation of algae and plankton in natural waters. The result of this type of environmental pollution is ecosystemic eutrophication, characterized by accelerated growth in simple photosynthetic organisms, such as algae and bacteria, and the elimination of species more sensitive to the changed environmental conditions, further exacerbating the increased biochemical demand for oxygen. The continuation of these processes of pollution causes environments to become anoxic or suboxic, in other words, with insufficient oxygen levels to support the full development of aquatic life. Poor and populous nations are the main agents of this type of pollution, whose cause is directly linked to low living conditions

among the population, with insufficient basic sanitation and sewage treatment. Classic examples of this situation are the receptive water bodies adjacent to large metropolitan areas, such as Guanabra Bay in Rio de Janeiro and the Tietê River, which runs through the city of São Paulo.

The second group, composed by pollutants from industry and mining, include toxic substances such as metals, greenhouse gases and organic pollutants, especially those generated by the burning of oil. Unlike the first group of contaminants, whose effects are generally local or regional at worst, these can affect the environment on a global scale. For example, the emission of greenhouse gases (especially carbon dioxide and methane) and metals (such as mercury and lead) into the atmosphere originates in part from energy generation and from the burning of fossil fuels. Though the effects of these contaminants are much less visible, their impact is far harder to remedy. Little is known about how natural ecosystems respond to chronic exposure to these contaminants, many of which, especially metals, are non-degradable and therefore accumulate in nature, where they continue to affect ecosystemic functioning over the course of decades or even centuries. For example, former mines like the left-over quarries from the Old Western gold rush in the USA, though in disuse since the century before last, still affect the local biota today (Mastrine et al., 1999).

In the closing decades of the last century, recognition of the danger posed by these pollutants led the majority of nations around the world to adopt rigid environmental legislation and replace highly pollutant technological processes. However, though there has been a significant reduction in emissions from point sources, such as industry, and diffuse sources, such as the improper discharge of waste from these activities into the environment, the increased demand for fossil fuels and the growth of large urban centers has given rise to a relative increase in the contribution of these so-called diffuse (or non-point) sources, whose emissions are unpredictable and produce effects that vary depending on the local characteristics. Furthermore, prevailing environmental legislation is still insufficient to deal adequately with diffuse source emissions. As a result, vast areas of the planet continue to receive chronically heavy loads of pollutants and the effects these will have on the natural ecosystems are still not wholly known.

Metals are contaminants present in effluents from a range of anthropic activities. Most metals, however, occur in compounds that are practically insoluble in water, which results in low concentrations being introduced to the environment even through point-source effluent emissions. Some metals are capable of occurring in stable chemical solutions, which can result in accumulation in the water column, leading to relatively high or even toxic concentrations. Among such metals are arsenic (As) and mercury (Hg), the latter being a ubiquitous component of industrial and agricultural effluents and an element naturally present in relatively high levels in a number of different regions. Of all contaminant metals, mercury is by far the most toxic, as well as the only metal capable of biomagnification in almost all food chains, that is, of its concentrations rising in accordance with the trophic levels of the species (Unep, 2002). This results in very high environmental exposure to mercury through the food chain for consumers at high trophic levels, including man. Mercury is the only metal proven to have caused human death s through environmental contamination, especially the ingestion of contaminated aquatic organisms (Barkay et al., 2003; Dorea et al., 2003). As its volatile form is chemically stable in the atmosphere, mercury gas (Hg<sup>0</sup>) can be disseminated on a global scale, affecting remote natural areas far away from the source of contamination.

Water pollution with mercury is particularly associated with the possibility of environmental methylation by the inorganic divalent form of the metal, Hg<sup>2+</sup> through bacteria and complexation with dissolved organic compounds that enable it to remain in the water column in comparatively high concentrations and seep back into the biota. As methylmercury is liposoluble, it is therefore easily absorbed by biological membranes in general and by the digestive tracts of practically all food chains. These processes abet the permanence and diffusion of mercury in the aquatic environment, as well as its dissemination to other ecosystems far removed from the contamination source. The organification of mercury therefore accelerates its bioaccumulation in the food chain and maximizes its threat to natural ecosystems and to human health (Porcella, 1994).

Some environments and environmental conditions help maximize the mechanisms of methylation and organic complexation of mercury, resulting in differentiated contamination processes. The present study will discuss the mercury dynamic in two systems where, given the dominant biogeochemical characteristics, the contamination of waters and aquatic organisms by this metal is more accentuated, implying risk even to the human populations that use their natural resources. These systems are mainland aquatic environments, including the Amazonian environments affected by gold panning and estuarial areas naturally rich in higher concentrations of dissolved organic matter.

#### Amazonian aquatic environments

In the closing decades of the last century, the socioeconomic difficulties experienced by developing countries and an increase in international gold prices led to a veritable gold rush in almost all nations in the Amazonian Basin. Amalgamation using metallic mercury was a process widely used in the enhancement and extraction of gold. As a result, large quantities of mercury were washed into the mainstems and atmosphere of the Amazonian ecosystem (Lacerda & Salomons, 1998; Malm, 1998).

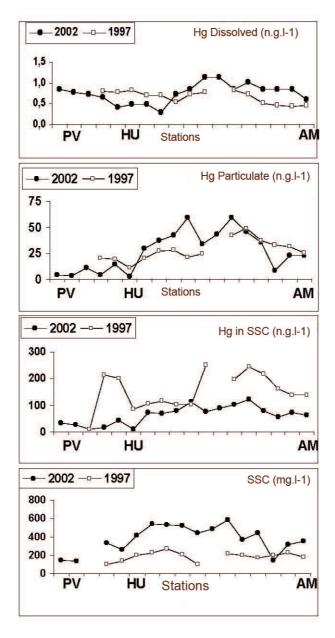
The high concentrations of mercury found in the Amazonian ecosystem were, as a general rule, attributed to gold mining (Pfeiffer & Lacerda, 1988; Malm et al., 1990; Nriagu, 1992), the presence of soils with naturally high mercury content (Roulet & Lucotte, 1995; Lechler et al., 2000), and the atmospheric transport and deposition of mercury of anthropic origin (Hacon et al., 1995, 1997; Lacerda, 1995).

Amazonian deforestation followed by the soil conversion for different uses, which began in the early 1970s and has been aggravated since the beginning of the 21<sup>st</sup> Century, is also singled out as a cause of mercury remobilization, whether of natural or anthropic origin. The erosion and lixiviation of mercury present in the soil and its re-emission into the atmosphere keep concentrations in the Amazonian ecosystem high, even as gold panning disappears (Almeida et al., 2005; Cordeiro et al., 2002; Lacerda, 1995). Another factor that favors the mobilization of mercury and possibly the production of methylmercury in the Amazon is the formation of large lakes for the generation of hydroelectricity, which largely result from the flooding of vast forested areas (Malm et al., 2004; Kehring et al., 1998; Palermo et al., 2004a,b).

During the heyday of the gold rush, high concentrations of mercury were measured in different compartments of the Amazon. After 1998, gold panning went into decline and even disappeared from most of the formerly mined areas. However, high mercury concentrations continued to be measured in fish stocks and the human populations that consume them (Bastos & Lacerda, 2004; Bastos et al., 2006; Barbosa et al., 2003), which suggests extensive remobilization of mercury deposited in the region's ecosystems. Some studies have attributed mercury remobilization to changes in soil use, particularly the removal of forest cover through slash-and-burn and subsequent conversion to pasture and/or other agricultural purposes (Lacerda er al., 2004; Almeida et al., 2005). As such, expanding deforestation to make way for cattle-ranching pasture may therefore be responsible for keeping mercury concentrations at such high levels. Figure 1 compares the concentrations of suspended solids, dissolved mercury, particulate mercury and mercury present in suspended material along a 1,600km stretch of the Madeira River between Porto Velho (RO) and its meeting with the Amazon River.

The comparison of findings for 1997 and 2002 clearly shows the influence of deforestation on the hydrochemistry of the Madeira River. The concentrations of suspended solids, particularly downstream of Humaitá, which receives run-off from the main soya expansion frontier in Amazonas state, increased noticeably between these two periods, from roughly 200 mg.L<sup>-1</sup> in 1997 to nearly 600 mg.L<sup>-1</sup> in 2002 (Figure 1).

Mercury concentrations in the particulate material in suspension downstream of Humaitá in 2002 (~100 ng.g<sup>-1</sup>) showed a reduction in relation to 1997 (~500 ng.g<sup>-1</sup>), suggesting the entrance of material less rich in mercury when compared with that still under the effects of gold panning in 1997. There were also increases in both dissolved fractions and particulate fractions of mercury contents in the waters. Once again, this was most clearly verifiable downstream of Humaitá. This result strongly suggests that alterations in soil use in the region spurred the remobilization of mercury deposits in soils and this process is responsible for keeping average mercury levels high in the waters of the Madeira River, especially downstream of Humaitá.





Concentrations of dissolved mercury, particulate mercury, mercury in suspended matter and solid content in suspension along the Madeira River between Porto Velho (PV), Humaitá (HU) and the meeting with the Amazon River (AM), in 1997 and 2002.

Mercury associated with particulate material in suspension tends to sediment, while dissolved mercury is usually more bioavailable for reincorporation into the biota. The mercury methylation process, which significantly enhances its bioavailability and accumulation throughout the food chain, is favored by slightly acidic anoxic or suboxic aquatic environments with high concentrations of dissolved organic matter and intense microbiological activity. In stagnant pools, riverside and flooded lakes and manmade reservoirs these biogeochemical characteristics are more likely to occur. Amazonian reservoirs, like those in the Brazilian southeast, have been the subject of numerous studies in virtue of their biogeochemistry and hydrodynamics, which are particularly favorable to the mechanisms of mercury methylation. The purpose of these studies has been to understand the dynamics of the processes involved.

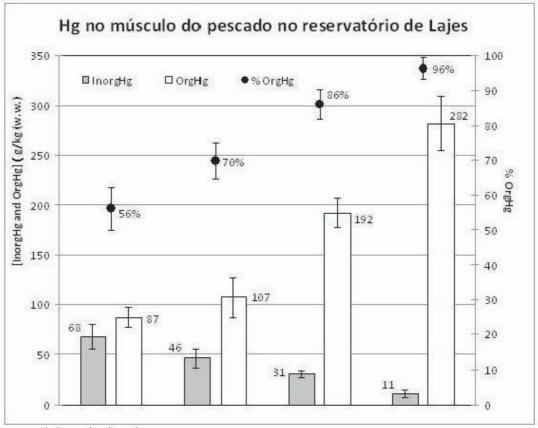
More recent reservoirs, besides offering the abovementioned conditions, all highly propitious to mercury methylation, also have an important additional and readily methylatable source of organic mercury in the remobilization of the soil and vegetation flooded to form the new artificial lakes. However, we will not be able to go into this aspect in greater detail in the present study. The high mercury mobilization in surrounding areas, associated with good conditions for the organification of mercury, could result in increased levels in reservoir fish stocks for periods lasting from five to thirty years (Palermo et al., 2002, 2004a,c; Malm et al., 2004). Studies conducted in reservoirs that are also aquatic systems, given their more stable hydrodynamics, have afforded a better understanding of mercury bioaccumulation processes in Brazilian mainland aquatic systems.

Plankton is the trophic level in the water column that has the functional characteristic of being the first link in the food chain to incorporate bioavailable mercury directly from the water. It therefore constitutes an excellent compartment through which to monitor aquatic systems. The Eduardo Penna Franca Radioisotope Laboratory at the Carlos Chagas Filho Biophysics Institute of the Federal University of Rio de Janeiro has amplified its studies by analyzing the distribution of different forms of mercury (total and organic) in samples of phyto- and zooplankton and from each trophic level of the fish stock in order to investigate how mercury behaves in aquatic ecosystems.

Studies at numerous Brazilian reservoirs, both in the southeast (Santana, Vigário and Lages) and in the Amazon (Tucuruí, Samuel and Balbina), have demonstrated how mercury associated with suspended (and sedimented) particulate matter and with plankton largely occurs in its inorganic form at the start of the reservoirs (upstream), with growing concentrations of the organic form detectable closer to the dam, which would suggest an extreme intensification of processes of organification within the reservoirs themselves. The most critical conditions are those found in the deep water areas near the dam walls. However, these conditions are indeed reflected downstream of the dam too. Here we encounter a third negative consequence of dams: a trophic leap among planktivorous fish that start to feed off items of animal origin (Malm et al., 2004; Palermo et al., 2004b).

Figure 2 presents data for the Lages reservoir, one of the oldest in Brazil (approximately a hundred years), where we can observe veritable textbook cases of various interesting aspects of mercury behavior in aquatic systems. The inorganic mercury present in the lakebed sediment and suspended particulate matter is basically incorporated by the detritivorous fish, but also on a smaller

scale in planktivores. Omnivorous fish feed on various different organisms, while piscavores are more specialized to a fish-based diet. As inorganic forms of mercury are not readily absorbed by fish digestive tracts (~10%) and have a low biological half-life, they tend not to accumulate in the food chain. The methylmercury, on the other hand, is almost entirely absorbed by the digestive tract at all trophic levels and has an enormous half-life, making it ripe for biomagnification.

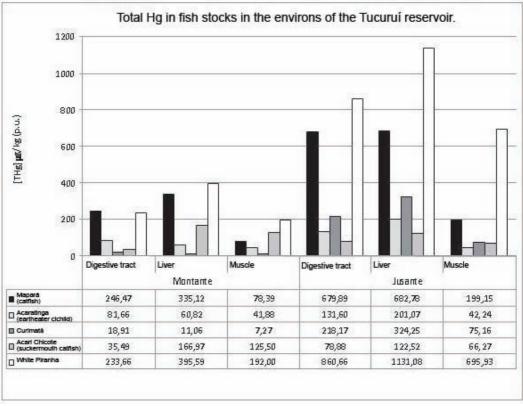


Source: Adapted from Palermo (2002).

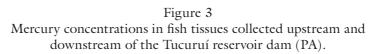
#### Figure 2

Concentration of forms of mercury in the fish food chain in the Lages reservoir.

Figure 3 shows what happens in a new reservoir (the Tucuruí in the Amazon), near the dam, where there is an accumulation of dissolved organic matter and generally anoxic conditions (i.e., perfect for methylation). However, downstream of the dam, where there is a change in feeding habits (based on the examination of stomach contents), planktivorus fish (in this case, the mapará catfish) start to ingest much more material of animal origin, and therefore present levels of contamination similar to those found in fish at the top of the downstream food chain (white piranha). As a result, the whole downstream food chain presents magnified mercury concentrations.



Source: Adapted from Malm et al. (2004).



The suckermouth catfish, a detritivore that generally incorporates inorganic mercury from lakebed sediments below the dam, presents lower values as a result of the high lixiviation capacity of waters rich in dissolved organic matter discharged into the deep upstream. It has been shown that the lowest mercury values in the sediment in the whole aquatic system are to be found downstream. The two species of omnivore present higher mercury values because of proven dietary changes.

### Mercury availability in estuaries

Various Brazilian estuarial regions have registered problems with metal contamination, especially mercury. Estuaries are regions of interface between the mainland and the ocean that present unique physico-chemical characteristics that can lead to the accumulation of pollutant substances. Estuaries serve as mediums for the transport of pollutants from the mainland to the sea, acting as biogeochemical reactors that alter the bioavailability of mercury. One of the key processes is the complexation of mercury with dissolved organic matter, which increases the solubility and stability of mercury in water, favoring its incorporation



Gold panners on the Juma River, in Novo Aripuanã, Amazonas State.

into food chains and its exportation to adjacent coastal waters (Paraquetti et al., 2004).

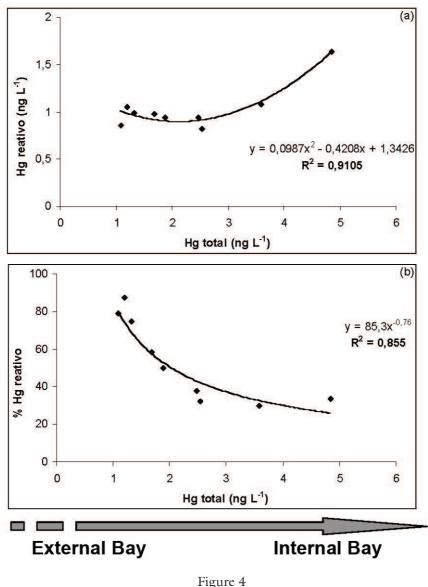
Researchers from the Millennium Estuaries Institute have been studying these processes in Sepetiba Bay, Rio de Janeiro, for many years. Sepetiba Bay receives a considerable volume of urban and industrial effluent from the environs and a significant contribution channeled from the Paraíba do Sul Riverbasin, which crosses the most industrialized region of the country. Studies of the waters re-routed from this river to Sepetiba Bay show that as much as 30% of the total mercury load that reaches the bay comes from the waters of the Paraíba do Sul (Molisani et al., 2007).

Studies on mercury transport, distribution and speciation in the waters of Sepetiba Bay show that there is a relative rise in soluble reactive species of mercury along the estuarial gradient of the bay, suggesting that the Sepetiba acts as an exporter of bioavailable forms of mercury to the nearby continental shelf. The same has been verified in other Brazilian estuaries, even those where there are no significant point sources of the element (Martins et al., 2002).

Annual mercury flow into the bay was estimated at 201 kg/year, 94.5% in the form of particulate mercury and the remaining 5.5% as fractions of dissolved mercury. In general, the total mercury content in Sepetiba Bay can be divided into two fractions: one particulate fraction, which represents 45% of total mercury content in the bay, and a dissolved fraction, which represents 55% of total mercury content. 40% of the dissolved fraction is composed of organic forms of mercury, with 13% reactive mercury (Hg<sup>2+</sup>) and 1% elemental mercury (Hg°). These results show that the particulate fraction dominates (45%?) mercury transport in the bay, while organic mercury represents the predominant form in the dissolved phase (Paraquetti et al., 2004).

Figure 4 presents the distribution of total mercury and its relation to the percentage of reactive mercury, that is, the fraction of total concentration present in the water immediately available for absorption into the biota, along the estuarial gradient of Sepetiba Bay. There is a direct relation between the total mercury concentration present in the water and the concentration of reactive mercury. A significant increase in total mercury concentrations occurs in the internal part of the bay, where the main rivers flow in, with measurements reaching as high as 5 ng.L<sup>-1</sup>, much higher than in the external part of the bay (~1 ng.L<sup>-1</sup>). Reactive mercury concentrations also rise in the internal part of the bay, hitting up to 1.7 ng.L<sup>-1</sup> against <1 ng.L<sup>-1</sup> in the external part (Figure 4). However, in comparison with total concentration (Figure 4b), the percentage of bioavailable (reactive) mercury is lower in the internal region (<40%) than in the external part of the bay, where it can reach 90% of total concentrations (Paraquetti et al., 2007), suggesting a significant increase in bioavailability.

Studies conducted during tidal cycles at the entrance to Sepetiba Bay suggest that, while the predominant form of mercury entering the bay is particulate, the organic mercury dissolved in the water is the main form of



Relationship between total mercury concentration and the biologically available fraction in the waters of Sepetiba Bay, Rio de Janeiro.

transport for the metal into the aquatic systems adjacent to the continental shelf or Ilha Grande Bay.

The increase in the reactivity of mercury over the course of its transport through the estuary occurs simultaneously with an increase in the concentration of organic mercury (Figure 5) and is associated with its complexation with dissolved organic matter, as demonstrated by the significant correlation between the two variables (Figure 5b) (Marins et al., 2002; Parquetti et al., 2004; Miolisani et al., 2007). Complexation with dissolved organic matter results in heightened bioavailability of mercury, even in areas without point sources of the metal, such as along tropical coastline dominated by mangroves, which are characteristically rich in dissolved organic matter (Lacerda et al., 2001). As a result, paradoxically, mercury bioavailability increases in regions farther removed from the source.

In Sepetiba Bay, relatively high levels of mercury have been recorded in fish caught in the outer rim of the bay and even in the region adjacent to Ilha Grande

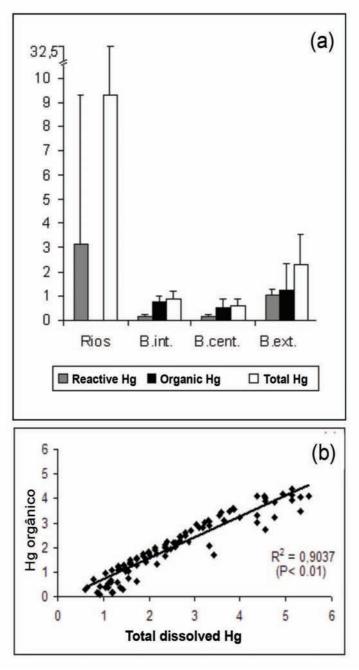


Figure 5

Distribution of mercury species in the waters of Sepetiba Bay, Rio de Janeiro, and the correlation between total dissolved and organic mercury concentrations. All concentrations are given in ng.L<sup>-1</sup>.

Bay (Marins et al., 1998), farther still from the main source of the metal through fluvial discharge into the bay. Though total mercury concentrations are higher in the inner bay areas, its bioavailability is lower.

Similar results were obtained in areas along the coast of Ceará State, characteristically void of point sources for mercury (Lacerda et al., 2007). Total mercury concentration and levels of organic mercury measured in different populations of sea-bass Cephalopholis fulva captured in coastal waters and on ocean shelves proved very different. Comparison between the two populations made it possible to investigate the effect of source proximity on mercury concentrations and speciation in liver and muscle tissue from the samples. Average total mercury concentrations in muscle (104 ng.g<sup>-1</sup> w.w.) and liver (2,865 ng.g <sup>1</sup> w.w.) and organic mercury concentrations in muscle (169 ng.g<sup>-1</sup> w.w.) and liver (1,038 ng.g<sup>-1</sup> w.w.) were much greater in the population captured off the ocean shelves. Both chemical forms of mercury were present in much higher concentrations in the liver than in the musculature. Total and organic mercury concentrations in the muscle and liver of C. fulva revealed a positive correlation with the size of the animal, which would suggest a process of bioaccumulation. However, in the coastal samples, only mercury levels in musculature revealed a correlation with animal size. Though the coastal environment is relatively rich in mercury compared with the ocean shelves, the highest concentrations were found in the oceanic C. fulva population. The significant correlation between organic mercury in muscle and animal size suggests greater mercury bioavailability in ocean waters when compared to coastal waters, which would corroborate the results obtained in Sepetiba Bay.



# Conclusions

Some bodies of water with favorable conditions for the mobilization and organification of mercury deserve closer attention from researchers and environmental managers. Aquatic systems with the abovementioned characteristics – subanoxia, anoxia, low pH, high levels of dissolved organic matter - need to be preserved and protected from human meddling in their drainage lest the toxic effects of metals (especially mercury) and of other contaminants be amplified. Dissolved organic matter is capable of forming complexes with various contaminants and of remaining soluble in the water column for longer periods, thus increasing its bioavailability to the biota. Black water systems, such as the Rio Negro, already have naturally high values of mercury in their waters and consequently in the associated biota. Their drainage basins should therefore be more stringently protected.

Given the small seasonal variations in temperature, reservoirs in the Amazon sometimes spend years on end in a stratified state (with no mixing with the deep anoxic layers), and this favors the formation of critical microenvironments, some anoxic, others suboxic, all acidic and with high levels of dissolved organic matter. These microenvironments function as reactors that can maximize mercury methylation or render other toxic substances soluble. These should therefore be monitored, especially in regions downstream of dams.

The process of eutrophication caused by the discharge of sewage into watercourses is another critical point, as it can create these conditions and potentize the effects of toxic substances by making them more soluble and bioavailable, as in the case of various estuaries along the Brazilian coastline. Environmental contamination with mercury is a reality, despite the reduction in point sources over the last three decades and the implementation of more rigid environmental legislation. The large quantity of mercury emitted by human activities during the last century is still, in various situations, subject to remobilization through anthropic and natural processes and through global climate variations and changes in soil use.

Now that the load/effect ratio no longer depends on the dimension of point-source emissions but on emissions from diffuse sources and from remobilization processes working upon accumulated mercury stocks in the environment, it has become much more important to garner thorough knowledge of the biogeochemical processes that control the dynamic of mercury in the environment than about the anthropic processes that generate them in the first place. Only thus will it become possible to construct scenarios that will enable us to understand the effects of mercury contamination on the Anthropocene.

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[...] The inhabitants of these regions say that they would prefer to drink the muddy water of the Madeira than the transparent, crystalline but treacherous and disease-ridden waters of its tributaries. [...] The waters of the Madeira carry a heavy clay load. This water, when stored without disturbance, sheds its suspended particulate load, which by agency of the capillary attraction it holds over bacteria, spirits them away in precipitation, removing them from the water: it is the self-purification process of muddy waters we know of so well. The same cannot be said of the clear waters, whose bacteria remain in suspension so they can infect all those who use the source [...]

(Dr. Oswald Gonçalves Cruz in Considerações sanitarias do Rio Madeira, 1910)

#### Authors' comments

The passage quoted above, written nearly a hundred years ago, observes the capacity of waters rich in organic matter to keep disease-causing pathogenic agents in solution. What we are learning today is that the introduction of organic matter into watercourses, unpleasant in principle, may in fact mobilize toxic substances in the river or lakebed sediment and render them available anew to the biota. These alterations in aquatic environments could prove more critical than the discharge of pollutants itself. This reinforces the need to treat all effluent and exercise as little change as possible on our waters.

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*ABSTRACT* - Mercury contamination in aquatic ecosystems in Brazil is discussed by comparing the biogeochemical characteristics resulting in increasing mobilization and bioavailability to food chains and eventually increasing human exposure to mercury. We compare key Amazonian systems and some estuaries, which due to the high complexing capacity of Hg by dissolved organic matter; maintain elevated concentrations of mercury under readily bioavailable forms, able to undertake methylation and biomagnification through food chains.

Keywords: Mercury, Contamination, Amazon, Estuaries, Bioavailability.

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