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## Mercury in tropical and subtropical coastal environments

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### Abstract

Anthropogenic activities influence the biogeochemical cycles of mercury, both qualitatively and quantitatively, on a global scale from sources to sinks. Anthropogenic processes that alter the temporal and spatial patterns of sources and cycling processes are changing the impacts of mercury contamination on aquatic biota and humans. Human exposure to mercury is dominated by the consumption of fish and products from aquaculture operations. The risk to society and to ecosystems from mercury contamination is growing, and it is important to monitor these expanding risks. However, the extent and manner to which anthropogenic activities will alter mercury sources and biogeochemical cycling in tropical and sub-tropical coastal environments is poorly understood. Factors as (1) lack of reliable local/regional data; (2) rapidly changing environmental conditions; (3) governmental priorities and; (4) technical actions from supra-national institutions, are some of the obstacles to overcome in mercury cycling research and policy formulation. In the tropics and sub-tropics, research on mercury in the environment is moving from an exploratory “inventory” phase towards more process-oriented studies. Addressing biodiversity conservation and human health issues related to mercury contamination of river

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## Keywords

Mercury land-based sources; Atmospheric transport; Tropical estuaries; Bacterial Methylation; Trophic Transfer; Human health

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## 1. Introduction

### 1.1 The atmospheric and riverine sources of mercury to tropical coastal environments

The oceans play a central role in the biogeochemical cycles of essential and nonessential elements through a series of abiotic processes (chemical speciation, inorganic scavenging, sedimentation) and biological processes (bio-alteration, bio-accumulation, and bio-magnification). In the last few centuries (and especially since World War II), anthropogenic perturbations to the global mercury cycle have in many cases overwhelmed the natural processes of mercury cycling (Mason et al., 1994; Mason et al., 2012). For a long time human influence on mercury cycling was detectable only at the local scale, but it is now clear that these influences are global. From land-based sources, natural and anthropogenic mercury releases reach coastal areas and from there gain access to the ocean and its trophic webs. Although changes in Hg concentrations in seawater through direct measurements are not yet fully documented in the tropics, inventories of local sources (e.g. Marins et al., 2004), the increase of mercury levels in the biota (Evers et al., 2008a), temporal trends in geochemical records (Xu et al., 2011) and mercury cycling models, all point towards the existence of a global problem (Doney, 2010), of which tropical coasts are only part.

The tropical and subtropical belt includes countries of every socio-economic level, including very poor countries to some of the ten largest global economies (Fig. 1). It is associated with some of the largest coastal population densities (e.g. India, Bangladesh, China, South-East Asia, Caribbean Islands and Nigeria), and some of the World’s megacities are on the margins of estuaries in tropical regions (e.g. Hong Kong, Rio de Janeiro, Bombay). Global mercury emission patterns (Pacyna et al., 2010) place most of the mercury atmospheric sources in densely populated coastal areas. However, nations in the tropical region are mainly emerging and developing economies, struggling with issues of population welfare, environmental protection and economic growth. In these cases, health and environment policies vary, and (mercury) pollution-oriented policies are not always in place. Therefore, coastal populations and environments are at risk in terms of direct and indirect exposure to mercury (e.g. thorough atmospheric transport-deposition, river discharge and seafood contamination).

Despite efforts to improve the situation, the linkages between river basin and coastal environmental management plans are poorly developed in countries of the tropical and subtropical belts (Barletta et al., 2010). The consequences of poorly integrated basin and estuarine management regarding water quality, and mercury pollution in particular, are transferred to (and amplified) in coastal waters (Costa et al., 2009; Evers et al., 2008a).

Each country has mercury emission characteristics related to their own socioeconomic status, but some common features exist. The three major sources of mercury are related to energy generation (industrial and domestic) through coal burning, the sum of small- and large-scale gold mining, production of non-ferrous metals and cement production. Deforestation may be another source of mercury to aquatic and coastal environments (Almeida et al., 2005). In some countries, such as China, mercury mining can also be a contributor to coastal contamination (Li et al., 2009)

Mercury and gold mining remain as major sources of environmental mercury contamination in tropical regions, putting nearly 9 million people at risk at approximately 280 of the 2100 identified contaminated sites all around the world ([www.unep.org](http://www.unep.org)). Chlor-alkali plants, while historically responsible for significant local and regional mercury contamination, are no longer a major mercury source for the environment since mercury electrodes are being replaced by the membrane-based production process and, where it has not yet been done, effluent treatment is usually in place.

The accumulation of mercury from non-point sources in remote ecosystems, where aquatic resources are extensively used by local populations, is a serious conservation and human health concern in coastal areas (Canuel et al., 2009), as for example in relatively isolated Pacific Islands (Denton et al., 2009; Chouvelon et al., 2009).

Tropical coasts harbour a variety of biomes, ecosystems, and habitats that are home to sand beaches, wetlands and flooded mangrove forests surrounding estuaries which support a rich biodiversity. Coastal reefs and seagrass meadows complete the ecoclines, leading to platform environments. Some of these continuums might disappear, or decline in areal extent. Habitat loss and changes in biological community structure may alter mercury cycling in these regions before we have a chance to fully understand the important mercury cycling processes. The consequences for the remaining elements of the coastal mercury cycle are unpredictable. Simplification of coastal tropical food webs, and consequent loss of biodiversity, may result in new patterns for mercury cycling and contamination in tropical coastal regions. Therefore, we are likely to face new and challenging human health and conservation issues in the near future.

In the absence of significant point sources for mercury contamination, aquatic biota in tropical regions are exposed to mercury contamination derived ultimately from highly seasonal atmospheric deposition (Costa et al., 2009; Barletta et al., 2012). The tropical climate is characterized not only by high air (>18°C) and water (>25°C) temperatures, but also by intense, well-defined rainy seasons (type “A” climates, from the Köppen-Geiger classification). Triggered by enhanced nutrient supply to estuaries and coastal waters, eutrophication and concomitant biological dilution tends to reduce mercury bioavailability to consumers, namely vertebrates who prey on estuarine and coastal organisms. On the other hand, mercury bioavailability can increase during the dry season, such that animals towards the top of the food web tend to show higher mercury concentrations (Costa et al., 2009; Barletta et al., 2012).

Mason et al. (1994; 2012) have shown that, in the last five centuries, anthropogenic impacts on mercury biogeochemical cycles have significantly changed both the nature of the processes and the fluxes of mercury on a global scale. Those impacts on tropical coastal ecosystems and habitats are no exception to that pattern. This paper discusses the possible nature and patterns of such changes in the tropics, focusing on coastal food webs from their base (bacteria and plankton), through filter feeders and other consumers, to apex predators (large fish, reptiles, birds and mammals) and finally to humans.

## 2. Mercury atmospheric sources to tropical and sub-tropical coasts

In tropical coastal regions, rivers and direct discharges from industrial facilities and sewage can be the dominant sources of mercury to seawater, biota and sediments. Although this is quite often the situation, direct atmospheric deposition can also be very important. Atmospheric sources of mercury might be of a more global nature (coal burning, volcanoes etc.), but others can be quite unexpectedly local, as for instance medical incinerators (Denton et al., 2011) and mishandling of e-wastes (Robinson, 2009). The mercury burden delivered to coastal environments by atmospheric transport and deposition interacts with local ecological and social patterns (Jardine and Bunn, 2010). So, it is necessary to take this atmospheric contribution into consideration, especially when inventories suggest higher levels of mercury in coastal environmental compartments than the most obvious, waterborne mercury sources, can possibly explain.

The GEOS-Chem model (Holmes et al., 2010) was used to estimate wet and dry deposition of oxidized Hg(II), particulate Hg<sub>p</sub>, and gaseous elemental Hg(0) to the tropical oceans on a seasonal and annual basis as a function of longitude. For this simulation of atmospheric mercury deposition in the tropics, we used the Streets et al. (2009) global anthropogenic inventory for 2006 partitioned into 17 regions; emissions within each region follow the GEIA 2000 distribution (Pacyna et al., 2006). In addition, we included Hg(0) emissions from artisanal gold mining (total 450 Mg a<sup>-1</sup>; Hylander and Meili, 2005). Anthropogenic emissions total 2050 Mg a<sup>-1</sup>. Biomass burning emits 300 Mg a<sup>-1</sup> following the distribution of CO from biomass burning. The model calculates soil, vegetation, snow, and ocean emissions (1200 Mg a<sup>-1</sup>, 260 Mg a<sup>-1</sup>, 210 Mg a<sup>-1</sup>, and 2000 Mg a<sup>-1</sup>, respectively) with coupled surface reservoir sub-models. Global mercury emissions are 8300 Mg a<sup>-1</sup> if we include gross ocean Hg(0) emissions or 6300 Mg a<sup>-1</sup> if we include only net ocean emission. These emission totals for anthropogenic and natural sources are within 15% of recent UNEP estimates for 2005 (Pacyna et al., 2010; Pirrone et al., 2010), but uncertainties for individual source types are estimated to be 25–50% (Pacyna et al., 2010).

On an annual basis (Fig. 2), Hg deposition is highest in the western Pacific (14 μg m<sup>-2</sup> yr<sup>-1</sup>) and lowest in the eastern Pacific (9 μg m<sup>-2</sup> yr<sup>-1</sup>). The high deposition rates in June-October in the western Pacific (Fig. 3) are due to elevated wet deposition, while wet and dry deposition are roughly equivalent the rest of the time. Dry deposition of elemental Hg generally accounts for about 10–20% of the total deposition.

Very few wet or dry mercury deposition measurements exist in the tropics (Mason et al., 1992; Lamborg et al., 1999), with none published in the scientific literature in the last

decade. Atmospheric concentration measurements in the tropics come mainly from transient ship cruises, with very few long-term monitoring sites (Sheu et al., 2010; Müller et al., 2012). Observations in this region are essential for evaluating and improving our understanding of mercury inputs to tropical terrestrial and marine ecosystems. In addition, global modelling efforts would benefit from research on the rate and mechanisms of the oxidation and reduction reactions for atmospheric mercury species, rate coefficients for gas-aerosol partitioning, mechanistic controls for natural land emissions, and field observations in remote regions, especially the southern hemisphere.

### 3. Mercury inputs to tropical coastal food webs

In many cases, mercury reaches coastal habitats through river discharge into estuaries. From there, a number of abiotic processes influence mercury cycling in tropical coastal waters (Costa and Liss, 1999; 2000; Paraquetti et al., 2007; Muresan et al., 2008a). Mercury methylation and uptake by the biota is also a concern since the average mercury levels in biota increased (Evers et al., 2008a), following expected biogeochemical trends (Mason et al., 1994; Mason et al., 2012).

Both primary producers and higher plants are seldom used in mercury research in tropical coastal environments, but occasionally appear in the literature in combination with other environmental compartments (water, sediments, primary consumers) (Stavros et al., 2008). A few studies used mercury measurements on mangrove tree leaves to assess the importance of the mangrove detritus food web on local and regional mercury cycles. Although these are frequent food items for charismatic organisms such as marine turtles (Guebert-Bartholo et al., 2011a) and the manatee (Stavros et al., 2008), studies on other multicellular tissues such as macroalgae and seagrass are rare.

Mercury (Hg) is an environmental pollutant that accumulates in aquatic ecosystems, generating health concerns for consumers of contaminated organisms (wildlife and humans). The most common organic form of Hg, methylmercury (MeHg), is of particular concern because it is a potent neurotoxin and biomagnifies in aquatic food webs. Thus, it becomes concentrated in the tissues of higher trophic level organisms (Wolfe et al., 1998). From a toxicological point of view, the key process that facilitates mercury entrance in coastal food webs is mercury methylation.

#### 3.1 Bacteria and mercury transformations in tropical coastal environments

The microbial community plays a critical role in the biogeochemical cycling of mercury, influencing its chemical speciation, reactivity, and bioavailability through mediation of processes such as methylation, demethylation, oxidation, and reduction. For some of these processes, the relevant bacterial groups, the important environmental factors, the biochemical pathways, and even the genes involved have been described in the literature (e.g. mercury demethylation and reduction). However, for other reactions (e.g. mercury methylation) our knowledge is still incomplete, despite the number of studies that have been conducted (Benoit et al., 2001; Barkay et al., 2003; Merritt and Amirbahman, 2009; Barrocas et al., 2010).

The key process that drives mercury bioaccumulation is mercury methylation. Most of the research concerning this process has been conducted in freshwater ecosystems, both in temperate and tropical areas. Huguet et al. (2010) found high Hg methylation potential in planktonic microorganisms in the anoxic water column and downstream waters of a reservoir in French Guiana, supporting the hypothesis that Hg methylation occurs under moderately anoxic conditions. Such conditions can occur in bottom waters and sediments where mixing and oxygenation are restricted, but also in microenvironments (i.e. biofilms) within an otherwise oxic water column. Coelho-Souza et al. (2006) reported the contribution of cyanobacteria (e.g. *Microcystis aeruginosa* and *Synechocystis* sp.) and bacteria associated with its mucilage to Hg methylation in an oligohaline tropical coastal lagoon in Brazil. Also, mercury methylation rates measured in periphyton associated with the roots of aquatic macrophyte in tropical freshwater systems (e.g. Amazonian lakes and rivers) were higher than rates measured elsewhere, demonstrating the relevance of these microenvironments (Mauro et al., 2002, Achá et al., 2011). When comparing Hg methylation rates in periphyton associated with aquatic macrophytes from a tropical and a temperate system, Guimarães et al. (2006) attributed the higher rates in the tropical ecosystem to higher bacterial production rates. Others studies reported higher methylation potential in tropical aquatic systems due to relatively high Hg(II) levels in the water column combined with elevated sulphate-reducing bacteria (SRB) activity, which was promoted by high temperature and organic substrate concentration and consequently low dissolved oxygen (Muresan et al., 2008b).

Despite the importance coastal zones play in the global mercury cycle, very little process-oriented Hg research has been done in coastal ecosystems, especially in the tropics (Whalin et al., 2007). New research to improve our understanding of the biogeochemical cycling of Hg in aquatic systems must take into account the prominent role played by the microbial community.

### 3.2 Estuarine, coastal and marine plankton: the base of the food web

The trophic transfer of mercury along marine food webs has been recognized as an important process, influencing bioaccumulation and the geochemical cycling of mercury (Fisher and Reinfelder, 1995). The trophic transfer factors along the food web are a useful tool to assess the biomagnification of mercury from one trophic link to another. However, bioavailability and chemical speciation (especially free ions) influences mercury toxicity and its bioaccumulation by organisms in the marine environment (Wang and Rainbow, 2005).

Primary producers can represent an important exposure route for mercury (Watras et al., 1998) to primary consumers, such as zooplankton and filter-feeding bivalves (Okay et al., 2000; Anandraj et al., 2008). Autotrophic and heterotrophic organisms in the microbial loop play key roles in the transfer of mercury into marine food webs (Fenchel, 2008), influencing the biogeochemical cycling of the aquatic ecosystem, as well as being major contributors to elemental cycling and vertical fluxes (Fisher et al., 2000). Dissolved MeHg, which is the most biologically available organic form of mercury, is bioaccumulated up to a million times in microscopic particles, including phytoplankton and bacteria, via adsorption to cell surfaces in the water column (Mason et al. 1996; Miles et al., 2001). These MeHg-enriched

particles are then consumed by zooplankton, which in turn are a primary food source for larval, juvenile, and some adult fish (Hall et al., 1997).

The microbial loop is expected to be an especially important feature in the ecology of tropical waters, due to the warm temperatures, high dissolved organic carbon (DOC) concentrations, and year-round high solar insolation. One important impact of the microbial loop on mercury cycling (Fig. 4) in the water column is the acceleration of organic matter mineralization and the regeneration of nutrients to fuel primary production (Fenchel, 2008). The accumulation of methyl mercury in prokaryotes (bacterio-plankton) will impact the transfer of mercury into food webs, due to the fact that bacterio-plankton can serve as food for zooplankton and small fish (Fenchel, 2008). The nature of metal binding in microplankton may significantly affect trophic transfer (Ng et al., 2005).

Due to their short life cycles, plankton respond quickly to environmental changes in the water column and phytoplankton community structure can reflect increased eutrophication and changes in water quality in aquatic systems (Linton and Warner, 2003). In tropical Brazilian aquatic ecosystems, cyanobacteria, a significant component of the marine nitrogen cycle and an important primary producer in many areas of the ocean, are associated with MeHg production; however showing low methylation potentials (Coelho-Souza et al., 2006).

Coastal eutrophication can affect the cycling of carbon, nitrogen and phosphorus, increasing primary production and planktonic biomass, and changing the biogeochemical cycling of mercury. Thus, coastal eutrophication can affect metal uptake, toxicity and trophic transfer in aquatic food webs (Wang et al., 2001). Under bloom dilution, as the algal biomass increases, the concentration of MeHg per cell decreases, resulting in a lower dietary input to the grazer community (zooplankton) and actually reducing bioaccumulation in algal-rich eutrophic systems (Pickhardt et al., 2002; Chen and Folt, 2005).

Organisms at the base of food web have been subject of several studies of the biological effects of contamination on plankton in coastal ecosystems worldwide (Barwick and Maher, 2003; Stewart et al., 2008; Telesh, 2004; Thompson et al., 2007). The literature includes studies in tropical marine ecosystem (Knauer and Martin 1972; Hirota et al., 1979; Zindge and Desai, 1981; Hirota et al., 1983; Thongra-ar and Parkpian, 2002; Al-Majed and Preston, 2000; Al-Reasi et al., 2007; Kehrig et al., 2009a, b; 2010; 2011).

Zooplankton have the ability to accumulate both inorganic and organic forms of mercury from ingested food and/or directly from the dissolved phase (Wang and Fisher, 1998). Copepods have been shown to assimilate MeHg about twice as efficiently as inorganic mercury ( $Hg_{inorg}$ ) (Lawson and Mason, 1998). Kainz and Mazumder (2005) demonstrated that variations in zooplankton MeHg concentrations can be better predicted assuming a bacterial diet rather than a phytoplankton diet. Some variations in plankton community structure, mainly zooplankton, have been associated with variations in the concentrations of mercury in fish tissues (Chen and Folt, 2005). Thus, physical and biogeochemical characteristics of the aquatic environment that affect growth dynamics of phytoplankton and the zooplankton communities that depend on them may also affect uptake of mercury into the pelagic-based food web (Stewart et al., 2008).

In the Tropical Pacific Ocean,  $Hg_{tot}$  in major zooplankton taxonomic groups was measured in order to obtain background “control” data from unpolluted waters. The maximum value of  $Hg_{tot}$  ( $0.125 \mu g Hg.g^{-1}$  dry wt.) was found in ctenophore samples from Fiji and the minimum ( $0.019 \mu g Hg.g^{-1}$  dry wt.) in thaliaceans from Northeastern Australia. However, neither a pronounced regional difference nor a characteristic difference in connection with the food web was identified (Hirota et al., 1979). The  $Hg_{tot}$  values in the Hirota et al. (1979) study (average:  $0.058 \mu g Hg.g^{-1}$  dry wt.) were lower than those found previously in zooplankton samples ( $0.081$  to  $0.448 \mu g Hg.g^{-1}$  dry wt., average:  $0.130 \mu g Hg.g^{-1}$  dry wt.) collected between Hawaii and Monterey, California, USA (Knauer and Martin, 1972).

The concentrations of  $Hg_{tot}$  in copepods from Minamata Bay were higher (over  $0.4 \mu g Hg.g^{-1}$  dry wt.) than those in the other regions investigated in a study of three inland seas in Western Japan (Hirota et al., 1983), suggesting that Minamata Bay was still polluted by mercury. Copepods collected in Seto-Naikai had the lowest concentrations ( $0.08 \mu g Hg.g^{-1}$  dry wt.), indicating that this environment was not polluted by mercury.

Composite zooplankton samples, without any separation according to size classes or taxonomic groups, were collected from the Inner Gulf of Thailand from 1976 to 1977. These samples had low values of  $Hg_{tot}$  ( $0.00295 \pm 0.0007 \mu g.g^{-1}$  on wet wt), ranging from  $0.002$  to  $0.0045 \mu g.g^{-1}$  (Thongra-ar and Parkpian, 2002).

Concentrations of  $Hg_{tot}$  and MeHg were measured in zooplankton samples collected with a  $250 \mu m$  mesh net in the Gulf of Oman. Total Hg in copepod samples ranged from  $0.010$  to  $0.037 \mu g g^{-1}$  wet wt. The mean concentrations were  $0.020 \pm 0.008$  and  $0.022 \pm 0.008 \mu g g^{-1}$  for the Matrah and A’Seeb sites, respectively. The MeHg fraction of the  $Hg_{tot}$  ranged from 1 to 19% (Al-Reasi et al., 2007). The concentration range reported in the Al-Reasi et al. (2007) study was wider than the range determined by Al-Majed and Preston (2000) for samples from Kuwait Bay ( $0.003 - 0.010 \mu g Hg. g^{-1}$  wet wt.). MeHg found in these samples accounted for 25% of  $Hg_{tot}$ . However, these reported concentrations for  $Hg_{tot}$  are well below those reported for polluted water bodies such as Bombay harbour ( $0.103-0.139 \mu g g^{-1}$ ) (Zindge and Desai, 1981).

In Guanabara Bay, in southeastern Brazil, Kehrig et al. (2009a, b; 2010; 2011) studied mercury cycling as it is influenced by plankton along an ecological gradient from the inner estuary to coastal waters. The authors demonstrated a change in patterns of mercury cycling according to changes in the plankton community structure. MeHg and  $Hg_{tot}$  were determined in three size classes of autotrophic and heterotrophic microorganisms,  $1.2-70 \mu m$  (seston, SPM),  $70-290 \mu m$  (microplankton) and also the  $>290 \mu m$  plankton components (mesoplankton). Higher MeHg and  $Hg_{tot}$  concentrations were significantly correlated with increasing size class. The increasing ratios of MeHg to  $Hg_{tot}$  (44% for seston, 60% for microplankton and 77% for mesozooplankton) reflects a transfer from the lowest trophic level, primary producers (cyanobacteria and diatoms), protist bacteriophagous (tintinnids) to primary consumers (copepods, nauplii, cladocerans and fish larvae – mesozooplankton), suggesting that biomagnification is occurring throughout this food chain.



### 3.4 The filter feeders, another way up to the top

Mollusks, especially bivalves, are the archetypal sentinel organisms. This group feeds by filtering particles from the water column. Bivalves are often preyed upon by fishes and marine birds. So, their participation in mercury transfer along trophic webs in coastal waters includes a flux through the benthic community, in addition to the pelagic pathway. Being a large taxonomic group, their role in coastal trophic webs varies widely. In this way, they might either decrease or increase the number of trophic steps as Hg moves from bacteria/plankton to the apex of the aquatic food web (and to humans).. Their convenient individual size and ease of sampling have guaranteed bivalves a privileged position in the study of mercury bioavailability. These animals are also the main target of governmental and non-governmental monitoring programs, and are widely used in human health risk assessments, due to their frequent consumption by most socio-economic groups in coastal areas.

The North American National Oceanic and Atmospheric Administration (NOAA) Mussel Watch program has been collecting and publishing information on mercury and other priority contaminants in mussels since 1986. The program also covers tropical coasts such as the Gulf of Mexico, Hawaii and Puerto Rico. The sample collection strategy includes species that feed on different average particle sizes (e.g. Mytilidae, Oysters) which allows a broader understanding of the mercury transport pathways in coastal ecosystems. The concepts developed under the Mussel Watch Program have been expanded to other countries including France, Australia, and Japan (conf. Costa et al., 2000). Coordinated worldwide experiments remain difficult to conduct, but have been attempted in the past (e.g. Mussel Watch International – Latin America covered from Mexico to Patagonia). Although such an approach may or may not be adopted as a governmental priority in tropical countries,  $Hg_{tot}$  and MeHg concentrations in bivalves are relatively well known, and other temporal series are starting to form, as in Brazil for instance (Costa et al., 2000; Kehrig et al., 2001; 2002; 2006). The species chosen for these studies have responded to water quality and mercury levels changes in a proportional manner, as has been demonstrated for  $Hg_{tot}$  availability and accumulation during translocation experiments (Fig. 5).

Many species of bivalves are being cultivated in aquaculture farms. Microbiological and sanitary aspects are the main concern with these animals and depuration in clean seawater for one to two days is a common recommendation before consumption of these organisms. However, this time does not allow for mercury, or other contaminants to be depurated. This is a growing concern, since mollusks farming is part of governmental policies all over the tropical world where they may offer animal protein when fisheries yields are no longer sufficient. The cultivation of bivalves involves less and cheaper technology than fish farming, and has faster results. Overall, mollusks aquaculture is more accessible for many coastal populations in tropical countries. Mercury levels in cultivated bivalves is usually set by local health authorities, but not necessarily are based on the WHO recommendations, and that might be in disagreement with local ecological conditions and social needs.

## 4. The consumers at risk

As the body size of aquatic organisms increases, the direct uptake of dissolved MeHg becomes less important, and trophic transfer becomes the dominant route for uptake and

accumulation of MeHg (Mason et al., 2000). In all ecosystems, including tropical and subtropical regions, the biomagnification of methylmercury in the food web of consumers strongly dictates their exposure level. In tropical vertebrates lower mercury concentrations are found in primary consumers such as the manatee (*Trichechus* spp.) (Stavros et al. 2008), low to moderate mercury concentrations in sea turtles (Day et al., 2010; Páez-Osuna et al., 2011) and many fish such as catfish species (*Cathorops spixii*; Azevedo et al., 2009; Barbosa-Cintra, 2010), and the highest mercury concentrations in upper trophic level species or apex predators.

#### 4.1 The apex predators

As long-lived apex predators, sharks are good indicators for measuring spatial and temporal trends in MeHg availability for near-shore tropical marine waters. They are not often protected by law, and capture and consumption is part of the traditional diet of coastal human populations world-wide. Sharks are well known for their ability to biomagnify MeHg and contain levels that are unsafe for frequent human consumption. Mercury concentrations in shark muscle tissue regularly exceed the United States Environmental Protection Agency (USEPA) action level of  $0.30 \mu\text{g Hg}\cdot\text{g}^{-1}$  wet wt. for many tropical and subtropical waters of the world, including Australia (Lyle, 1984), Belize (Evers et al., 2008b), Brazil (Pinho et al., 2002), and Florida (Adams et al., 1999). Individual sharks that forage on prey related to the demersal fish community tend to have higher  $\text{Hg}_{\text{tot}}$  concentration than more pelagic species (Storelli et al., 2002). In coastal waters of southern Belize, 101 sharks representing 10 species were analyzed for muscle  $\text{Hg}_{\text{tot}}$  levels (Evers et al., 2008b). The highest  $\text{Hg}_{\text{tot}}$  was recorded in bull (*Carcharhinus leucas*), blacktip (*Carcharhinus limbatus*), hammerhead (*Sphyrna* spp.), and nurse sharks (*Ginglymostoma cirratum*). Over 88% of the sharks sampled exceeded USEPA human health consumption standards. Because muscle Hg can be positively correlated with size for blacktip and nurse sharks (Evers et al., 2008b) and other shark species (Lyle, 1984; Hueter et al., 1995; Lacerda et al., 2000; Pinho et al., 2002), models for confidently predicting muscle Hg levels for species used for human consumption are promising.

Among tropical teleost fish, predatory species such as king mackerel (*Scomberomorus cavalla*), and various species of cutlassfish and grouper are apex predators that can contain elevated  $\text{Hg}_{\text{tot}}$  levels of concern to human health and sustainability of their populations. For example, the goliath grouper (*Epinephelus itajara*) is a long-lived species. In southern Belize, 40% of goliath grouper individuals exceeded the USEPA action level of  $0.30 \mu\text{g Hg}\cdot\text{g}^{-1}$  wet wt. – including all individuals over 55 cm in total length (Evers et al., 2009). While regular consumption of this and other grouper species by sensitive groups of people, such as pregnant women and young children, should be closely monitored the question of adverse impacts on this critically endangered species is also of concern, especially during complex social behaviours during spawning aggregations. The Atlantic cutlassfish (*Trichiurus lepturus*) has a pan-tropical distribution and has been widely used as a bioindicator of mercury bioaccumulation and biomagnification (Fig. 1) (Costa et al., 2009). Cutlassfish are highly valued at markets and widely consumed by communities living alongside tropical and subtropical marine ecosystems. Larger individuals may especially

pose a health threat if consumed frequently or by vulnerable human groups (Barletta et al., 2012).

Crocodylians are also at risk to elevated mercury concentrations in tropical and subtropical ecosystems, although threshold levels of concern are undefined (Rainwater et al., 2007; Vieira et al., 2011). Marine birds have also been a target for describing mercury exposure in the tropics (Kojadinovic et al., 2007; Catry et al., 2008). However, only a few species such as the white ibis (*Eudocimus albus*) and great egret (*Ardea alba*) have been used for long-term monitoring and assessment efforts for mercury in Florida (Sepúlveda et al., 1999; Frederick et al., 2004; Jayasena et al., 2011).

Many species of marine mammals inhabit tropical and subtropical coastal waters, especially porpoise and dolphins. Broad and wide-ranging prey sources likely dictate the great variability in mercury exposure documented (Seixas et al., 2007; 2008; Bisi et al., 2012; Moura et al., 2012). Although the live capture of most marine mammals is rare, studies on carcasses are becoming more frequent along tropical coasts (e.g., Amazon River mouth), helping to establish a broader ability to assess health and contamination issues for marine mammals (Moura et al. 2011).

The feeding preferences of the coastal dolphins *Pontoporia blainvillei* and *Sotalia guianensis* in south-eastern Brazil were assessed through the prey's index of relative importance (IRI),  $Hg_{tot}$  and isotopic signatures ( $\delta^{15}N$  and  $\delta^{13}C$ ) to compare their efficiency in the discrimination of prey contribution to the predators' diet. The IRI was found to be the best indicator of the dolphins' prey preferences, while  $Hg_{tot}$  and  $\delta^{15}N$  seemed to be a better indicator of trophic status when the diet was specific or made up of prey of varying sizes. The  $\delta^{13}C$  values were characteristic of a typical coastal food chain, confirming the preferred habitat area of these species (Di Benedetto et al., 2011).

#### 4.2 $\delta^{13}C$ and $\delta^{15}N$ in studies of mercury trophic transfers in tropical coastal waters

Mercury levels in fish and marine mammals depend not only on the contamination of their environment but also on several ecological factors such as diet and trophic position (Das et al., 2000). Changes in ratios of stable isotopes of carbon ( $\delta^{13}C$ ) and nitrogen ( $\delta^{15}N$ ) help elucidate trophic relationships within marine food webs and to confirm the relationships between contaminant uptake and trophic position (Cabana and Rasmussen, 1994).

Stable isotope ratios  $\delta^{15}N$  and  $\delta^{13}C$  help to identify organic matter sources in the food web (Peterson and Fry, 1987), and have been used to assess trophic level and trophic transfer of  $Hg_{tot}$  in aquatic food webs (Kidd et al., 1995; Al-Reasi et al. 2007; Ikemoto et al., 2008; Evers et al., 2009; Senn et al., 2010).

Several studies on mercury biomagnification and bioaccumulation using  $\delta^{13}C$  and  $\delta^{15}N$  in tropical marine food webs have been conducted. Al-Reasi et al., (2007) evaluated mercury biomagnification in zooplankton and fish in the Gulf of Oman. Using linear regression modelling of  $\delta^{15}N$  data to infer trophic position (slopes  $\beta_1 = 0.07$  and  $0.14$ ;  $r^2 = 0.09$  and  $0.11$ ) they reported that biomagnification of  $Hg_{tot}$  and MeHg was lower than had been observed in the Arctic ( $\beta_1 = 0.2$ ) (Atwell et al., 1998), or in temperate ( $\beta_1 = 0.29$ ) (Jarman et

al., 1996) marine and freshwater ( $\beta_1 = 0.17 - 0.48$ ) (Kidd et al., 1995) ecosystems. The lower biomagnification might be due to the fact that tropical marine ecosystems generally have high species diversity and complex food webs. However, the data sets on the relationships between trophic position determined by  $Hg_{tot}$  and MeHg, and  $\delta^{15}N$  are heterocedastic, preventing the use of some statistical analyses, even if the data are log-transformed. The safe use of  $Hg_{tot}$  and MeHg biomagnification,  $\delta^{15}N$ , biological and environmental data without mathematical manipulation requires a search for the better-fitted distributions, and only then the application of more adequate statistical analysis becomes possible (Sonesten, 2003, Barletta et al., 2012).

Senn et al. (2010) combined N, C and Hg stable isotopes measurements to identify the factors influencing MeHg accumulation in fish from the northern Gulf of Mexico (USA), and concluded that trophic position measured by  $\delta^{15}N$  explained most of the variance in  $\log[MeHg]$  ( $r^2 \sim 0.8$ ). However, coastal species in the Mississippi River plume and migratory oceanic species derive their MeHg from multiple sources, and both food webs have different baseline  $\delta^{15}N$  signatures and (possibly) isotopically distinct MeHg sources.

In the Mekong Delta (South Vietnam), a significant trophic level-dependent increase was detected in concentrations of Hg and others trace metals (Ikemoto et al., 2008). Based on the trophic level assignments estimated from  $\delta^{15}N$  analysis, the authors suggest that differences in the biomagnification profiles between crustaceans and fish species might be due to differences in metal accumulation and detoxification abilities such as possession of metal-binding proteins (e.g., metallothioneins).

A tropical coastal lagoon located in Southeast Brazil that receives runoff from iron ore mining and processing activities was studied to assess the differences in trace element accumulation patterns in sediment and to investigate relationships between metal accumulation and trophic status of the organisms (Pereira et al., 2010). Trace element accumulation in biota depended not only on the trophic position of the organisms but also on the functional ecological guilds of the species involved. Gastropods (detritivores), showed higher trace element concentrations than crustaceans and fishes. Although length co-varies with the trophic position of fish species, it did not influence the concentrations of trace elements. Moreover, omnivory in fishes with fast growth may explain the general lack of correlation between  $\delta^{15}N$  and trace elements. In the adjacent marine coastal ecosystem, when species were grouped according their ecological and functional guilds. The values of  $Hg_{tot}$ ,  $\delta^{15}N$ , and trophic level increased significantly from primary producer toward carnivores.

Evers et al. (2009) studied mercury concentrations in goliath grouper *Epinephelus itajara* of Belize (Central America), and reported that the stable isotope analysis for  $\delta^{13}C$  and  $\delta^{15}N$  indicates a broad prey base and a relatively high trophic level. This study also discusses ontogenetic dietary shifts and the Hg exposure in different size classes of grouper. Through biomagnification and bioaccumulation of MeHg, older specimens are therefore at greatest risk of physiological impairment when performing complex and coordinated behaviours, such as those associated with spawning aggregations.

In marine systems MeHg uptake and biomagnification in biota depend not only on mercury input to the ecosystem but also ecological (e.g., ontogenetic phase, diet and trophic position), and environmental (e.g., rainfall, salinity, water temperature, suspended particulate matter) variables that affect MeHg sources and mobilization. If we assume that food availability in density (individuals.m<sup>-2</sup>) and biomass (g.m<sup>-2</sup>) varies through space and time (Barletta et al., 2005; 2008), thus influencing bioaccumulation and biomagnification, it is important to understand how seasonal fluctuations of the environmental variables influence food availability at different trophic levels, and consequently mercury accumulation and biomagnification fluctuations in coastal food webs.

## 5. Mercury and human health issues in tropical and sub-tropical coastal regions

On a global scale, the main pathway of Hg exposure to humans is the ingestion of contaminated seafood (2.4 µg MeHg.day<sup>-1</sup>.person<sup>-1</sup>, WHO), and this is most likely the major pathway of mercury exposure for coastal populations in the tropics and sub-tropics. Local and regional impacts may differ considerably from global averages due to variations in the speciation of mercury emissions, pollutant dispersion, and dietary habits (Fig. 1). Understanding these differences will require more detailed studies on local and regional scales.

This form of low-level exposure is probably more wide-spread than extreme cases of coastal pollution because it affects the human populations throughout their lives. Recent evidence on low-level mercury exposure also shows mercury effects on endpoints such as cardiovascular, immunologic, and birth outcomes (Karagas et al., 2012). But, with the exception of the Seychelles study, this issue has rarely been investigated by health authorities in human populations along tropical coasts. So, the delicate balance between the benefits from fish eating and the risks due to Hg exposure (Mergler et al., 2007; Oken et al., 2012) is difficult to establish.

Some of these native coastal populations have lived in relative isolation for many decades. With the expansion of urban areas, the increased value of coastal tourism and the information revolution, these groups have emerged as active stakeholders. These populations are now the target of a number of sociological studies (e.g. Diegues, 2008), but these rarely include health assessments of mercury body burdens. Therefore, their real mercury contamination is unknown, but is suspected to be significant, based on the parallels with their Amazonian counterparts. They may have any number of nutritional deficits and endemic health problems (e.g. anaemia), aggravated by extreme poverty, malnutrition, inbreeding, intestinal parasites, and other untreated illnesses, including decompression sickness (Guebert-Bartholo et al., 2011b). In these communities there has often a nearly a complete dependency on aquatic resources, including practically all animal groups occurring in tropical coastal environments. The intensification of contact with other groups can alter their dietary patterns and preferences (Sant'Anna et al., 2001), possibly due to access to other sources of animal protein that can be bought at markets (beef, pork and chicken), more easily and at better prices than fish or other seafood. These food items have become part of the culture, either by cultural colonization or because high quality fish (apex predators) are

no longer available. This dietary shift can “dilute” the mercury burden in these populations and, at the same time, divert the seafood (and its associated mercury burden) to other markets where highly-valued seafood is sold.

Many marine fisheries are near collapse due to over-fishing and environmental degradation, and aquaculture (also referred to as mariculture and/or fish-farming) has become a significant alternative for the supply of protein to human and domestic animal populations. However, as human population grows out of control, we will have to rely less on animal protein, especially seafood since most fisheries are already beyond sustainable levels. Aquaculture will never be more than a minor source of protein. The sustainability of these operations, and the safety of the food thus produced, are therefore issues of concern. For example, mariculture operations may not be aware of the mercury burden in fish food, nor the impacts of this on the Hg concentrations in the seafood that is being produced. With the exception of algae and filter feeders (e.g. *Crassostrea* spp., Mytilidae, Pecten), farmed species are generally omnivores that need some amount of animal protein in their diets, and therefore have the potential to biomagnify mercury from their feed to their final human consumers.

Mercury use in amalgamation for gold ore mining has been a major source of mercury pollution to terrestrial aquatic environments. Occupational exposure of gold miners and their families has therefore received attention from health authorities across tropical regions such as the Amazon, for example (Hacon et al., 2008 a, b). On the other hand, large-scale mining, chlor-alkali and other industrial plants, and coal burning, are the major sources of mercury to coastal and marine environments, contributing Hg contamination through river discharge and atmospheric deposition. Regulation of industrial mercury emissions is contentious because it should include the consideration of costs and benefits. The estimation of benefits is complex, partially because mercury contamination is a global phenomenon and therefore benefits need to be assessed on a global scale. However, costs are usually assigned to someone, especially local governments. Abatement of mercury pollution can be cost-effective depending on the marginal costs which, in turn depend on available control technologies and specific local conditions. For example, in terms of absolute magnitude, the social costs due to IQ loss from low-level Hg exposure are low, but given the small amount of mercury involved and the potential number of children affected, the marginal cost is high when compared to other pollutants (Spadaro and Rabl, 2008).

Therefore, mercury environmental monitoring should include studies of at-risk and control groups, since there are no consistent background values against which to compare mercury levels in threatened human populations. It will be necessary to establish local/regional threshold or “background” mercury exposure levels and mercury concentrations, in order to conclude whether a population is or is not statistically different from those with similar cultural, occupational and dietary habits. Proposals for integrated projects with traditional populations of fishers in tropical coastal areas should include objectives such as: (1) the calculation of human exposure to mercury and other pollutants through seafood consumption, especially for pregnant women and children; (2) living resources and environmental services conservation to guarantee the survival of endangered species and socioeconomically important seafood stocks; (3) assessment of nutritional quality from

traditional diets in order to suggest more efficient and protective use of protein sources; (4) implementation of sustainable aquaculture projects; (5) educating local and regional “target” populations about the relative risks and benefits from seafood consumption as a complimentary option to focusing solely on reducing Hg releases to the environment (Spadaro and Rabl, 2008).

## 6. Conclusions and Recommendations

Tropical countries facing mercury pollution problems need to be involved in research and monitoring of coastal environments from sources to sinks, and must recognize the risks it poses to human health and ecosystem preservation. Because of an historical focus on mercury pollution in freshwater ecosystems, many marine habitats in developing countries within the tropics remain under-sampled with respect to mercury contamination. More process-oriented research is needed to answer questions such as: (1) How are ecosystem functions and services affected by changing mercury burdens through time and space, and how do these factors affect the various biogeochemical Hg cycling processes? (2) To what extent have we already altered the mercury cycles and increased the mercury levels in coastal ecosystems of the tropical and subtropical belts by modifying river discharge and increasing atmospheric transport and deposition?

Long term monitoring of mercury sources and cycling in these environments has been limited (Evers et al., 2008a), and should be established at ecosystem scales that also consider the impacts of human activities. Suitable locations for ecosystem-based studies have been defined through international efforts to divide the major marine habitats into “homogeneous areas” (e.g. Large Marine Ecosystems - LME, Global International Water Assessment – GIWA and the Census of Marine Biodiversity) and studies in these areas must consider the impacts from mercury pollution. New statistical approaches (e.g. long time series) are recommended for pre-existing and newly generated data, in order to further explore for information that might have a wider ecological significance. The inclusion of the most relevant environmental variables in monitoring programs (Barletta et al., 2012), distributed across broad spatial and temporal scales, and the expanded use of sentinel animal models is important (Evers et al., 2008a; Costa et al., 2009). Marine Conservation Units and fisheries grounds should be priority areas for such experiments and programs. Before sampling and monitoring programs go out in the field, the guarantee of quality data on mercury concentrations and speciation, in the myriad of environmental, biological and human matrices must be assured by all candidate laboratories that intend to participate in research and provide input to policy-makers via participation in international intercalibration exercises.

Some of the challenges to mercury biogeochemical research in tropical coastal environments are the extreme climatic and environmental conditions in the tropics (e.g. high temperature; wide fluctuations of salinity and turbidity between rainy and dry seasons) and different socioeconomic conditions, even within the same national territory. These conditions lead to high variability in many aspects of the mercury cycle. Biological turnover rates are high throughout the year. Overfishing and over-pollution are serious threats to marine biodiversity in addition to invasive species, altered water temperature, ocean acidification,

increasing frequency of hypoxia events, expansion of aquaculture operations, and maritime traffic. These are worldwide phenomena, and are threatening marine species of ecological and economic importance. Direct and indirect anthropogenic mercury inputs in the tropics need reliable identification, quantification, and monitoring. Climate change will inevitably alter rainfall regimes and river flow patterns. Deforestation and intense land use for economic development will also change the characteristics and ecological services of river basins. In combination, these will probably alter the cycling processes and bioavailability of mercury in coastal areas. Each of these areas have different sensitivities to these various threats, however, water pollution is ranked high (3<sup>rd</sup>) in most cases. Understanding the synergistic and cumulative impacts of mercury and other pollutants within the framework of environmental change due to these multiple stressors is recognized as a challenge for the future.

Climate change can also increase the erosion of soils in mangrove areas, where large amounts of organic carbon, and possibly mercury (Marchand et al., 2006), are presently stored. Connectivity between the terrestrial, coastal, and pelagic environments and their biological communities will serve to transmit the damages (Newton et al., 2012). Global climate change may have impacts on mercury cycling beyond those expressed directly at the local and regional level, and we will not be able to predict those impacts without a much better understanding of the rates and mechanisms of biogeochemical mercury cycling. For example, biotic and photochemical reduction and re-emission of mercury from the land and ocean are already significant terms in the global mercury budget. If global warming increases the reduction rates (and the subsequent re-emission rates), the mercury burden may be shifted by long-range atmospheric transport from more polluted urbanized regions (such as North America and Europe) to less-polluted areas like tropical coastal environments. Acceleration of the reduction/emission/atmospheric oxidation/deposition cycle should lower the concentrations of bioavailable Hg(II) in aquatic systems (reducing fish mercury), while higher temperatures coupled with coastal eutrophication may enhance MeHg formation by expansion of suboxic and anoxic environments (increasing fish mercury). Changes in the chemical speciation of Hg(II) and MeHg due to ocean acidification will alter their bioavailability in ways that we cannot reliably predict at present. Expansion of the tropics due to climate change (Seidel et al., 2008) will spread tropical coastal biological species, and their associated mercury contamination issues, to higher latitudes in a matter of a few centuries.

High population densities and poverty result in poor land and water resources use, including interference in trophic webs through overfishing, for example. The rapidly increasing price of land in urban centres and the economic development of some nations will set a trend towards the cremation of bodies, instead of the conventional burial. This could increase the atmospheric deposition of mercury near cremation facilities unless their emissions are tightly controlled to prevent the release of reactive oxidized gaseous mercury or aerosol mercury. Economic growth and development also involves building more houses, roads and infrastructure in general, which requires more metal and cement processing (resulting in more local and regional emissions of mercury). Landfills, a known source of mercury to coastal environments when poorly managed, will expand in numbers and in area and volume. Tourism in coastal areas is increasing. The enhancement of tourism, especially eco-



tourism, will encourage greater use, and impact upon, pristine areas, where development will inevitably occur. Such development inevitably leads to impacts on coastal environments, and steps must be taken to minimize the negative impacts of development. For example, the ecological services provided by mangrove habitat will be at least partially impaired, especially with respect to sequestration of mercury in sediments.

The increase in coastal/maritime tourism and international trade (shipping) due to the globalized economy will require more frequent and deeper port maintenance. Dredging may become a more frequent activity due to poor conservation practices in the upland river basins. Many countries in the tropics have no controls or regulations on dredging activity or dredge spoil disposal, and this may increase the risk of mercury contamination. Nations benefiting from these economic and social changes need to cooperate on a global scale to evaluate the environmental costs from altered mercury cycling and to find appropriate ways to minimize or mitigate the negative impacts. Records from museum and biological specimen collections (Hill et al., 2010) and sediment cores (Xu et al., 2011) may contribute to the assessment of global and local conditions and their quantitative and qualitative changes over time and space.

International collaboration can bring benefits that have a low-cost/high-impact effects on mercury research. *Oceans and Seas* comprise an entire section of the United Nations report “The future we want”, and the participation of marine researchers is called upon in a number of other sections regarding the issues discussed in this paper: coastal and marine pollution, oceans and human health, aquaculture, fisheries, food security, and capacity building. In paragraph 221, mercury is specifically cited: “We welcome the ongoing negotiating process on a global legally binding instrument on mercury to address the risks to human health and the environment and call for a successful outcome of the negotiations.” (UN, 2012). There is a need for research and monitoring that can answer questions such as: are we changing, for better or for worse, the mercury cycling in coastal areas? Can global actions such as the Global International Water Assessment (GIWA) and the Agreement on Agriculture (AoA) take mercury into account, enhancing the collection of existing data, the generation of new knowledge, and the transformation of this information to governments and other decision-makers?

Supra-national institutions (e.g. WHO, IAEA, UNEP, FAO) have an important role to play in the compilation of mercury emissions and for supporting basic research on Hg cycling in countries of the tropical and subtropical belts. The goals of these institutions should be focused on the transference/seeding of financial resources for research and health care, and also on the identification and organization of priority actions designed for each geographical region and for the various impacted human populations. In addition, international aid to strengthen analytical and diagnostic capabilities with respect to mercury pollution is welcome, especially when a country may not have the funding, facilities, and human expertise to evaluate its own status concerning the impacts of mercury on their people and their environment. As their missions are redefined and altered into the future, it will be important for these institutions to maintain an emphasis regarding marine mercury research, seafood advisories, environmental monitoring and source control.

Many countries in the tropics and sub-tropics need assistance with setting up appropriate seafood consumption advisories for mercury (e.g. UNEP INC 1 to 5). Human exposure to mercury from wild and farmed fish and shellfish are likely to be quite different, and establishing appropriate consumption standards for the general population as well as at-risk populations should be part of every sustainable fisheries certification, since it is a matter of food security in many countries. Where they already exist, consumption standards for wild and farmed fish and shellfish should be evaluated and revised when necessary. The establishment of such standards is an urgent action where they do not already exist. Because of geographical variations in human cultures (dietary choices) and in climate and fauna (thus variations in mercury cycling), it is important to recognize that consumption standards have to be determined at the appropriate national, regional, and perhaps even local scales.

We will have to constantly balance the desire to preserve ecosystems and biological diversity against the needs of society (human health and food supply). Governments and NGOs must cooperate to provide the funding that is needed to address these critical scientific and sociological research issues. The eradication of Poliomyelitis through voluntary vaccination of children in most countries of the world is probably the best example where people, independent of culture, responded well to a serious health threat when correctly addressed by their governments. Improvements in information technology will continue to enhance the dissemination and availability of data (and other information) on mercury issues that will help to define the nature and scope of future ecosystem research programs as well as to translate the information to society. Scientists, governments, and non-governmental agencies need to cooperate to identify key research gaps, promote capacity building, promote opportunities for experts to meet and exchange knowledge, support low-cost, open access, publications targeted to decision makers as well as other scientists, develop new analytical and pollution-abatement technologies and environmental management techniques, and support international collaboration at all levels for all stakeholders (Costello et al., 2010).

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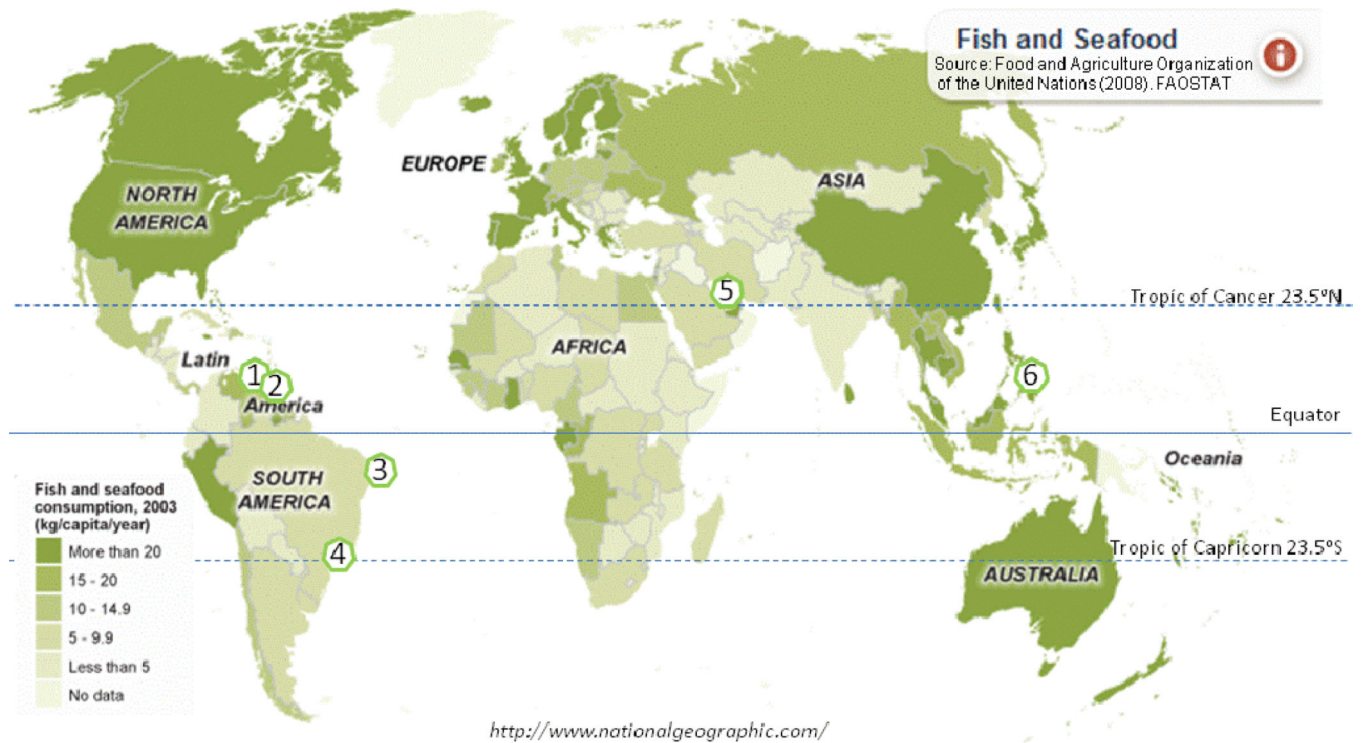
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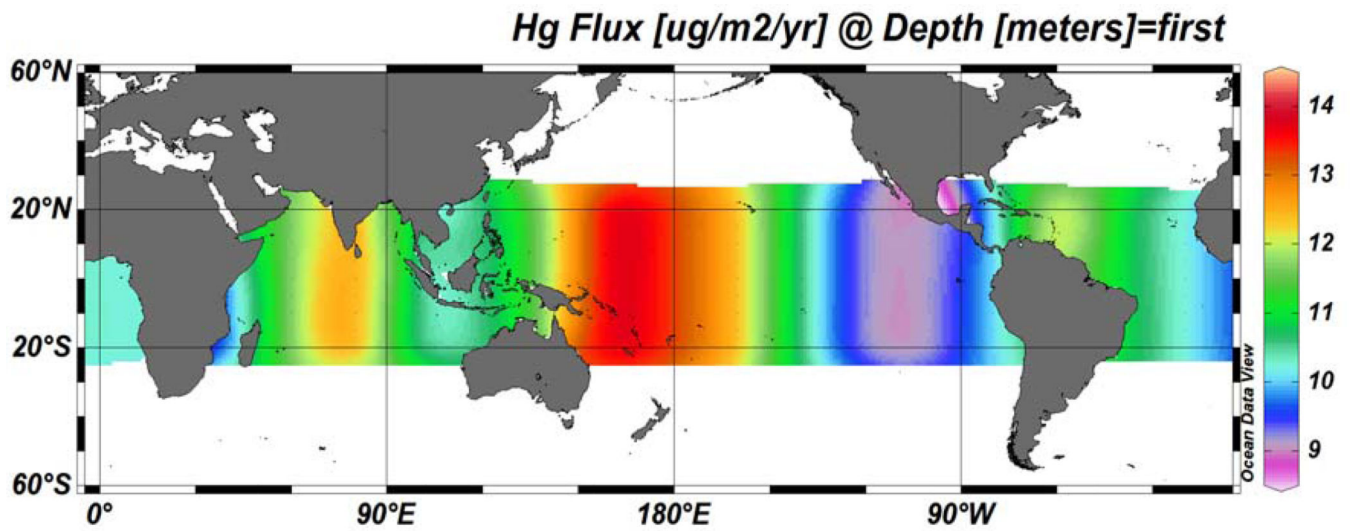
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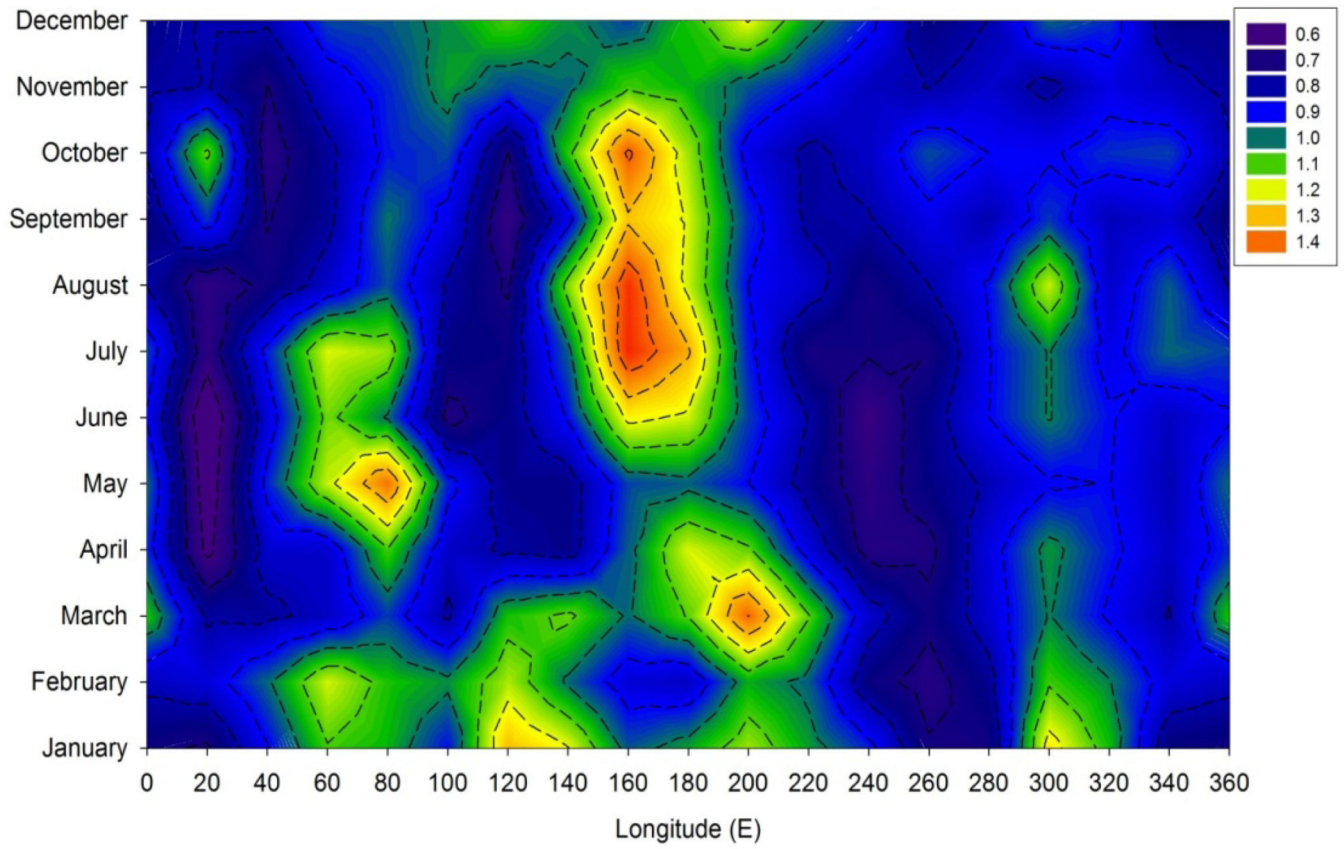
**Figure 1.**

More than 150 countries are in the tropical and subtropical zones, the large majority are small island states. Only a few countries have vast extensions of coastline in the tropics and sub-tropics (ex. Brazil, Australia, India, China, Mexico, USA, Chile, Peru, Angola). Seafood consumption varies widely among these nations, depending on size and capacity of fishing fleets and cultural values. Coastal pollution by mercury and other pollutants might have a role to play in this scenario at the local scale. Studies with *Trichiurus lepturus* (1) Shrestha et al., 1988 – Venezuela; (2) Mol et al., 2000 - Suriname; (3) Costa et al., 2009; Barbosa-Cintra et al., 2011 - NE Brazil; (4) Kehrig et al., 2004; Cardoso et al., 2009; Kehrig et al. 2009c; Kehrig et al., 2010; Kehrig et al., 2011; Carvalho et al., 2008; Di Benedetto et al. 2012; Muto et al., 2011; Bisi et al., 2012; Seixas et al., 2012a, b; Seixas et al., in press - SE Brazil; (5) Saei-Dehkordi et al., 2010 - Persian Gulf; (6) Prudente et al., 1997 - Philippines.

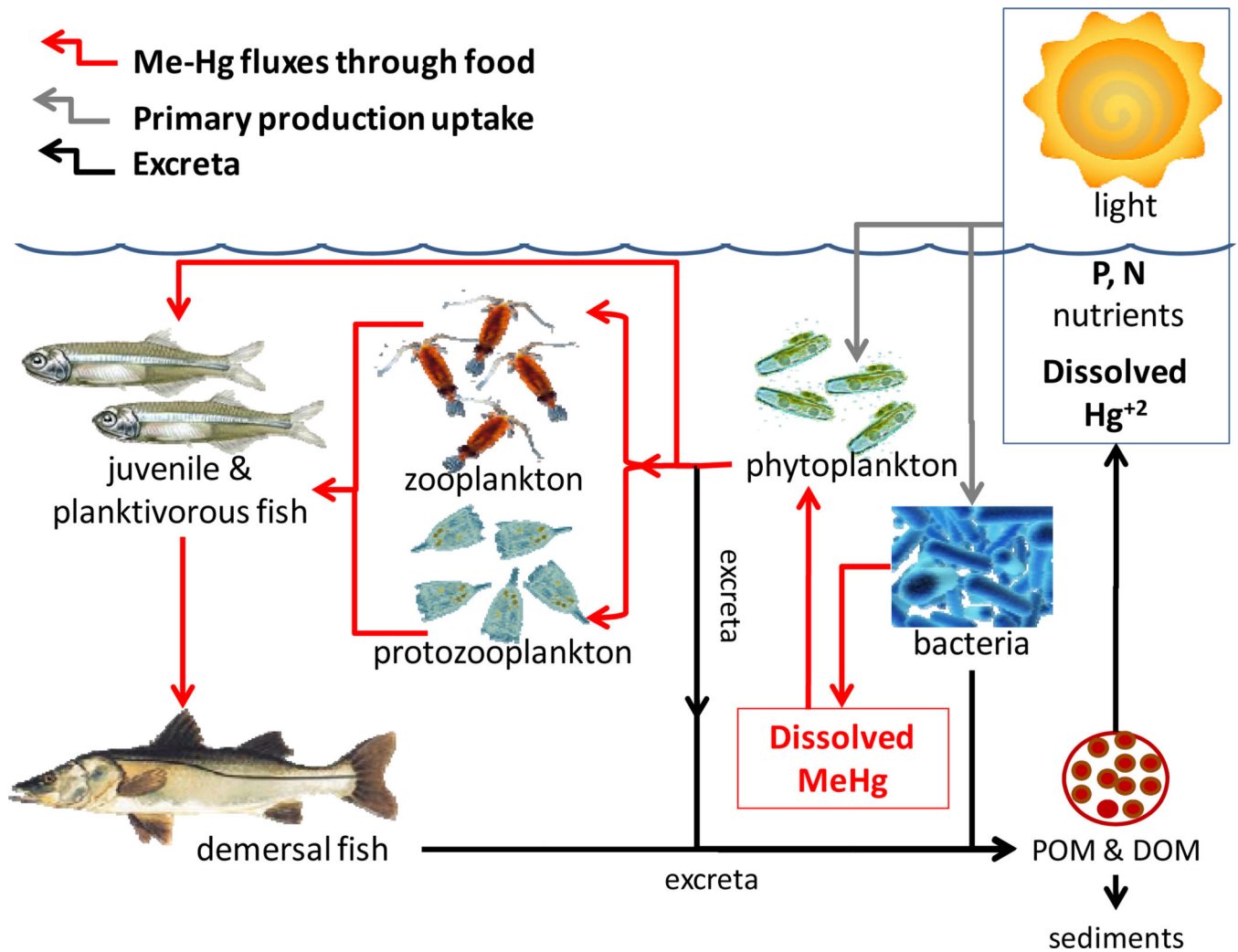


**Figure 2.**

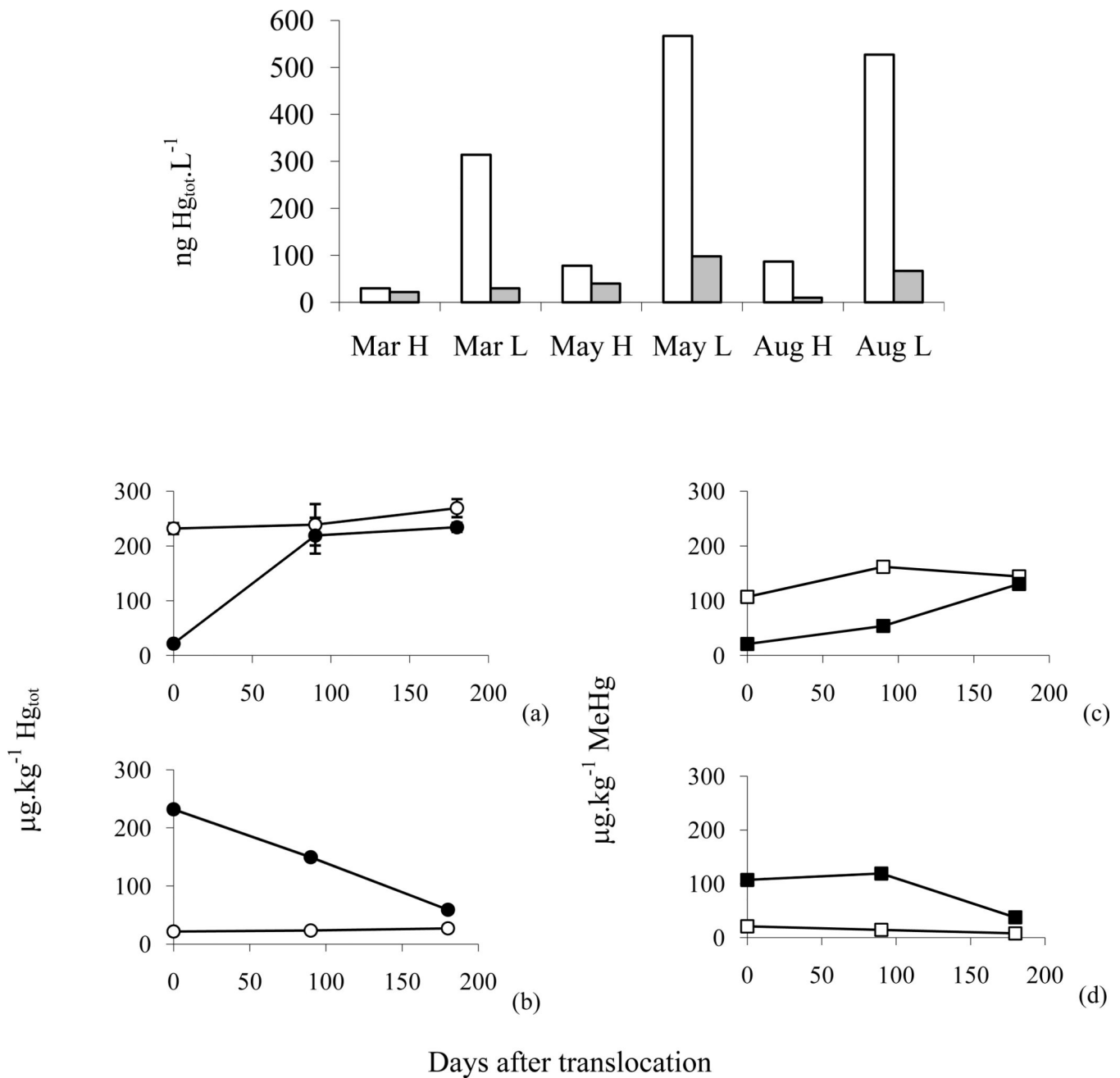
Annual mercury deposition (wet plus dry, including  $\text{Hg}(0)$ ;  $\mu\text{g m}^{-2}\text{h yr}^{-1}$ ) over the tropical oceans simulated using GEOS-Chem. Deposition shown here is averaged over tropical latitudes ( $24^\circ\text{N}$  to  $24^\circ\text{S}$ ).



**Figure 3.** The seasonal variability in total mercury deposition (wet plus dry including Hg(0);  $\mu\text{g m}^{-2} \text{ month}^{-1}$ ) across latitude simulated using GEOS-Chem.



**Figure 4.** Microbial loop and trophic transfer of methylmercury (MeHg) through a tropical estuarine food web. From Kehrig, 2011.

**Figure 5.**

Total mercury ( $Hg_{tot}$ ) and methylmercury (MeHg) in *Crassostrea rhizophorae* (mangrove oyster) transplanted between two estuaries of the Brazilian Northeast. Top panel: Mercury in suspended particulate matter ( $ng\ Hg_{tot}.L^{-1}$ ) at experimental sites. March (end dry season), May (early rainy season) and August (late rainy season) 2000. High tide (H); Low tide (L). White bars contaminated estuary; gray bars non-contaminated estuary. Other panels: White symbols local oysters; Black symbols transplanted oysters. (a) and (c) contaminated estuary;

(b) and (d) non-contaminated estuary. M. F. Costa, N. Sant'Anna Jr. and H.A. Kehrig, unpublished data.