Note to reader

This draft version of Chapter 6 in the Technical Background Report to the Global Mercury Assessment 2018 is made available for review by national representatives and experts. The draft version contains material that will be further refined and elaborated after the review process. Specific items where the content of this draft chapter will be further improved and modified are:

1. All graphics will be redrawn to a common appearance from the originals presented here, with their sources cited in the captions.
2. References will be completed and presented in a uniform style.
3. Conclusions and main messages will be formulated

GMA 2018 Draft Chapter 6. Relationships between Trends in Atmospheric Hg and Hg in Aquatic Biota.

Peter Outridge, Robert Mason, Feiyue Wang, Lars-Eric Heimburger, Milena Horvat, Xinbin Feng

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6.1 Relationships between Trends in Atmospheric Hg and Hg in Aquatic Biota

The goal of the Minamata Convention is to reduce Hg emissions mainly from atmospheric sources (see Chapter 1.1), with the ultimate aim of reducing the exposure and harmful effects of Hg in wildlife and humans. However, the pathway between Hg´s release into the atmosphere and its eventual accumulation in wildlife and humans is biochemically and geochemically complex. Mercury is emitted into the air from most low- and high-temperature anthropogenic and natural sources primarily as gaseous elemental Hg (GEM; Hg^0). In the atmosphere, GEM is ultimately oxidized to Hg^{II} and part of this airborne inorganic Hg is deposited into aquatic environments, where it joins other inorganic Hg^{II} that is present as a result of waterborne releases from other natural and anthropogenic sources. A small fraction of the inorganic Hg pool in aquatic environments is converted by natural microbial processes into more toxic methylated forms - monomethyl Hg (MeHg), and (less commonly) dimethyl Hg (DMeHg), with MeHg being the form that is bioaccumulated and biomagnified within foodwebs.

The aquatic geochemistry stage of the global Hg cycle is therefore an important transformative step in the sequence between anthropogenic GEM emissions, the atmospheric deposition of inorganic Hg^{II}, and MeHg accumulation in foodwebs. A number of environmental and ecological factors (including redox condition, pH, organic carbon and nutrient concentrations, food web trophic structure, temperature, and light intensity) have a strong influence on the rates of MeHg production and degradation, as well as the rate of uptake of MeHg by aquatic biota. Together, the complexity of the atmospheric Hg cycle, the limits of our understanding of the methylation/demethylation cycle, and the number of influential factors affecting MeHg bioaccumulation, mean that there is considerable uncertainty about how closely changes in the emissions and deposition of Hg into the environment brought about by regulatory action will be tracked by changes of Hg in aquatic food webs.

This chapter describes recent advances in our developing understanding of the aquatic geochemistry of Hg, particularly focussing on the connectivity between atmospheric Hg and Hg levels in aquatic biota. The chapter is divided into two sections which address the following issues: (1) recent advances in understanding of methylation and demethylation in marine systems (6.1); and (2) observed relationships between the trends in atmospheric Hg emissions and deposition and in aquatic biota, and the reasons for dichotomies between those trends (6.2). These topics were chosen because they are of the greatest

11069 importance with respect to advances in aquatic Hg geochemistry since AMAP/UNEP (2013), and because
11070 of their relevance to predicting the efficacy of the Minamata Convention in ultimately reducing Hg
11071 exposure in humans and wildlife.

11072 **6.1.1 How has our understanding of the marine methylation and demethylation cycle**
11073 **evolved since GMA 2013?**

11074 The concentration of methylated Hg species (MeHg, DMeHg) in an aquatic water column represents the
11075 culminating effect of various processes that influence the methylation of ionic Hg (Hg^{II}) to MeHg and
11076 DMeHg, their demethylation, as well as transport from the location of their formation to the water
11077 column. Generally, Hg is methylated by bacterial processes in sediments and the water column of large
11078 water bodies, such as the ocean and large lakes, but not in the water column of most freshwater
11079 ecosystems. While methylated Hg can be produced by abiotic reactions and processes, its formation is
11080 thought to be primarily biotic and microbially-mediated (Paranjape and Hall 2017). In contrast,
11081 demethylation of these compounds is thought to be by both abiotic and biotic pathways, with DMeHg
11082 being volatile and more unstable in the environment than MeHg. Overall, therefore, the concentration
11083 of methylated Hg is the net result of many competing processes of formation, transport, and
11084 destruction.

11085 Methylated Hg compounds constitute a small fraction of the total Hg present in some environments
11086 (e.g. < 1% in air and typically <5% in marine sediments, but with somewhat higher relative
11087 concentrations in freshwater sediments and wetland soils; Paranjape and Hall 2017). However, these
11088 compounds can be a much larger fraction of the total Hg in the water column, and can exceed 20% of
11089 total Hg in the open ocean. Additionally, in some marine waters such as in the Arctic Ocean, DMeHg can
11090 be as abundant as MeHg (Lehnherr 2014). In biota, the fraction as MeHg increases as a function of
11091 trophic level, from ~20% of total Hg in seston to >90% in high trophic level biota. As MeHg is the more
11092 toxic form of Hg, and poses the primary exposure risk to humans and other top predators, it is of prime
11093 importance to understand the production and fate of these compounds.

11094 As discussed further below, due to the complexity of methylation and demethylation, it is not possible
11095 to generalise these processes into either global or regional MeHg budgets, although some progress is
11096 being made in this regard. Furthermore, given the complexities that control the *net* formation of MeHg
11097 in the environment, it is clear that while reducing total Hg emissions to the environment can be
11098 expected to ultimately reduce MeHg in biota in general and over time, more detailed predictions of the

effects of regulatory actions on Hg in biota in a specific ecosystem requires further understanding of the methylation/demethylation processes in the ecosystem in focus. This conclusion is further outlined in the sections below. The following text focuses on methylation/demethylation in marine systems, because of the predominance of MeHg from marine foodwebs as the main exposure route in many human populations around the world (see Chapter X in this Report).

6.1.1.1 Methylation in Coastal Waters

Key points: 1) sediments are not always the most important source of MeHg to the estuarine water column; 2) water column methylation occurs in coastal waters; and 3)the factors controlling methylation in coastal environments (e.g. nutrient and carbon loading, redox) are complex.

Much of the earlier work concerning Hg methylation in coastal waters highlighted in the previous Technical Report (AMAP/UNEP 2013) was focused on the factors controlling methylation in the sediments and the flux from sediments to the water column. Overall, the consensus view at that time was that for many environments, sediments were the major source of MeHg to coastal waters, although there were indications that this was not always the case and that inputs from terrestrial watershed and/or from ocean exchange were important in many ecosystems. Nevertheless, the consensus was that any new (*in situ*) production of MeHg within the estuarine and coastal environment was due to the production of MeHg in sediments.

In the last few years, however, a number of studies have challenged this notion, and suggested that MeHg accumulation in coastal/estuarine biota is not exclusively from sediment inputs. Firstly, Chen et al. (2014) found that the concentrations of MeHg in forage fish across multiple estuaries on the US east coast did not track with the MeHg content of the sediments, but with the water column concentration, even though these fish are considered to forage at the sediment-water interface. Conversely, in the same study MeHg in benthic worms did track the sediment MeHg concentrations. Mercury stable isotope analyses also tended to confirm that the sediment may not have been the most important source of MeHg to the organisms in these ecosystems (Kwon et al., 2014). Li et al. (2016) used Hg isotope analyses to demonstrate that the source of MeHg in biota in Lake Melville, a large subarctic fjord, was from pelagic production. Similarly, sulphur (S) isotope analyses of plankton from Long Island Sound (LIS) did not support the idea that the accumulated MeHg had a substantial sediment component (Gosnell et al., 2017).

However, Buckman et al. (2017) showed that within the Delaware estuary, these patterns were more complex and it was less easy to discern the importance of sediment inputs of MeHg compared to riverine inputs. Gosnell et al. (2016) showed that for the Delaware River, sediment could be an important MeHg source at certain times of the year, suggesting that sediment sources should not be completely ignored. Jonsson et al. (2017) showed that it is not just the MeHg loading that was important, but that changes in the concentration of dissolved organic carbon (DOC) can influence MeHg bioaccumulation (see also Balcom et al., 2015, and Gosnell et al., 2016). Comparison of water column and sediment MeHg concentrations show that in some ecosystems, such as the Hudson River, there is a reasonably strong relationship between dissolved water column MeHg and porewater MeHg, and between sediment and suspended particulate MeHg, but there are many ecosystems where there is little correlation.

One important factor, which has received less attention, is the degree to which the MeHg levels are influenced by demethylation of MeHg rather than by its formation. Many studies have assumed that demethylation is not a strong control on MeHg levels in coastal ecosystems but this assumption needs to be tested further. Overall, current literature suggests that there are no clear-cut trends across coastal ecosystems and that both internal and external sources of MeHg are likely important contributors of MeHg to the food chain.

Recent studies have reached contrasting conclusions on the role of nutrient inputs impacting methylation rates in sediments, and MeHg levels is coastal waters and biota. In mesocosm studies, Liem-Nguyen et al. (2016) showed that the addition of nutrients could impact Hg methylation in sediments, and that inorganic Hg input to the water column was more efficiently methylated than Hg injected into sediment, as found in earlier studies (Jonsson et al., 2014), suggesting that the bioavailability of inorganic Hg for methylation may change with time. Oxygen status of the water column is also an important factor in methylation rates, with the consensus being that increased eutrophication leading to oxygen depletion (hypoxia) in bottom waters results in increased MeHg production. A recent example of this process was provided by the modelling of Soerensen et al. (2016) which suggested that increased MeHg in Baltic Sea plankton was associated with increasing eutrophication. However, contrary examples have also been reported recently, with no increase in sediment MeHg levels in some coastal regions with bottom water hypoxia (Chakraborty et al., 2016; Liu et al., 2015). In LIS, in the more eutrophic regions where bottom waters are seasonally hypoxic, plankton had lower MeHg than those from more oligotrophic regions, which was the opposite of the expected pattern (Gosnell et al., 2017). Again, these

11159 results suggest that the interaction between eutrophication and MeHg levels in biota is complex, and
11160 likely to differ in different locations.

11161 Organic carbon (OC) is an additional important factor influencing both Hg methylation as well as MeHg
11162 retention in sediments. Mazrui et al. (2016), for example, found that the binding of Hg to DOC enhanced
11163 methylation compared to Hg bound to particulate (POC) and cinnabar. However, the origins and
11164 geochemical quality of the OC (terrestrial or marine) is at least as important as its quantity in terms of its
11165 effect on Hg bioavailability (Schartup et al., 2015b; Jonsson et al., 2012, 2017). Additionally, it has been
11166 shown in pure cultures and laboratory sediment studies that nanoparticulate Hg has higher
11167 bioavailability for methylation than microparticulate (Mazrui et al., 2016; Zhang et al., 2014). These
11168 studies reinforce the conclusions of prior studies (Schartup et al., 2013; 2014; Jonsson et al., 2012) that
11169 the factors controlling Hg methylation in sediments are extremely complex given the interactions
11170 between Hg (and MeHg) and sediment biogeochemistry (primarily, the levels of OC and reduced
11171 sulphur) which impact binding, bioavailability and sediment-water exchange. While speciation of the Hg
11172 is an important driver, desorption kinetics and microbial community activity are also important controls
11173 over the extent of Hg methylation in sediments.

11174 The weight of evidence for the importance of water column methylation in coastal waters has increased
11175 in recent years. A number of studies have followed up on earlier work in the Thau Lagoon, France
11176 (Monperrus et al., 2007), examining the potential for methylation of Hg within the water column of
11177 coastal environments. A number of other studies have now shown that there is the potential for
11178 methylation in the water column of estuaries and coastal waters, especially in locations of mixing and
11179 flocculation of particulate material (Schartup et al., 2015; Sharif et al., 2016; Ortiz et al., 2015). These
11180 studies point to the likely enhancement of methylation within aggregated particles where micro-anoxic
11181 conditions could exist, as demonstrated by the laboratory experiments of Ortiz et al. (2015). Overall,
11182 these studies do not suggest that Hg methylation is occurring through a different microbial biochemical
11183 pathway, but that it is occurring within the anoxic microzones within large particulates. Some of these
11184 studies have concluded that there is significant net methylation within the water column (Schartup et
11185 al., 2015; Ortiz et al., 2015) while in other cases, the extent of demethylation leads to a net decrease in
11186 MeHg (Sharif et al., 20016).

11187 In conclusion, there is not one specific source for the MeHg accumulating in biota in coastal systems,
11188 and the sources are likely to vary spatially and temporally. In examining, and understanding, the

dynamics of MeHg bioaccumulation in coastal environments it is necessary to examine both the potential external inputs (watershed and ocean inputs), and the internal production within the system (water column and sediment net Hg methylation). Furthermore, it is likely that their relative importance will change in the future due to climate and other human-caused alterations within these ecosystems.

6.1.1.2 Open Ocean Hg methylation

Like coastal seas, there is increasing evidence for active methylation in the oxygenated water column of open oceans. Early pioneering work by Mason and Fitzgerald (Mason et al. 1990) suggested the potential for high rates of *in situ* production of MeHg in the open ocean, however, the prevailing paradigm continued to favour a coastal sediment MeHg source with offshore transport to the open oceans. Since GMA 2013, additional studies have confirmed the suggestion that *in situ* MeHg formation takes place in open ocean waters (Monperrus et al. 2007, Cossa et al. 2009, Sunderland et al. 2009, Heimbürger et al. 2010, Cossa et al. 2011). There is now published evidence for water column methylation from almost all major ocean basins: the Atlantic Ocean (Bowman et al. 2015, Bratkič et al. 2016), Pacific Ocean (Hammerschmidt et al. 2012, Munson et al. 2015, Bowman et al. 2016, Kim et al. 2016), Arctic Ocean (Wang et al. 2012, Heimbürger et al. 2015), Southern Ocean (Gionfriddo et al. 2016), Mediterranean Sea (Cossa et al. 2012), Baltic Sea (Soerensen et al. 2016), and Black Sea (Rosati et al., GBC in review). No data has been published for the Indian Ocean thus far. Laboratory experiments confirm that net Hg methylation can occur in "marine snow" (settling organic particles), with similar rates compared to marine sediments (Ortiz et al. 2015). Furthermore, several papers point out open ocean methylation is required to balance the oceanic MeHg mass budget (Sunderland et al. 2009, Mason et al. 2012, Soerensen et al. 2016).

The relationships observed between MeHg concentrations and apparent oxygen utilization as well as organic carbon remineralization in the oceanic water column indicate that particulate organic matter remineralization controls the methylation of Hg by providing inorganic Hg as the substratum, and by stimulating the activity of methylating bacteria. A pioneering study explored for the first time the carbon isotope composition of the MeHg compound in tuna fish, and found similar $\delta^{13}C$ values to marine algal-derived organic matter, suggesting its role as the carbon substrate for Hg methylation (Masbou et al. 2015). Additional evidence comes from Hg isotopic analysis of marine biota. Fish that forage at different depths in the North Pacific Ocean show Hg isotope gradients that can only be explained if 60-80% of their MeHg is produced below the surface mixed layer and is not from a sediment source (Blum et al. 2013).

In general, the depth, shape and importance of the MeHg peak in ocean waters depend on physical forcing and biological productivity. Several independent studies found methylation hotspots at the density gradients of stratified systems (Wang et al. 2012, Heimbürger et al. 2015, Schartup et al. 2015, Soerensen et al. 2016). Two field studies (Baya et al. 2015, St. Pierre et al. 2015) and a modelling study (Soerensen et al. 2016) suggest important evasion of DMHg from the Arctic Ocean, where MeHg is produced at shallow depths (Heimbürger et al. 2015).

A major breakthrough has been made with the discovery of two key genes, hgcA and hgcB, that control anaerobic Hg methylation in sulphate-reducing bacteria (Parks et al. 2013). The hgcA and hgcB genes were found to be present in many anaerobic microorganisms. An analysis of publicly available microbial metagenomes found the hgcAB genes in nearly all anaerobic environments, but not in aerobic systems (Podar et al. 2015). A marine microaerophilic bacterium has been identified as a potential Hg methylator within sea ice, where anaerobic bacteria which are known to methylate Hg were absent (Gionfriddo et al. 2016). Surprisingly, laboratory experiments have not found a clear relationship between the expression level of the key genes and net MeHg production (Goni-Urriza et al. 2015).

6.2 How and why do Hg levels in aquatic biota respond to changes in atmospheric Hg?

As discussed, there are many processes that may affect the dissolved concentrations and biouptake of MeHg following its formation. Other factors and processes affect the transport and fate of inorganic Hg (GEM and ionic Hg^{II}) between their emission sources and aquatic environments (see earlier chapters of this GMA). The complexity of these processes raises the question of whether Hg emissions, especially those that are regulated under the Minamata Convention, are likely to result in immediate and proportional changes of Hg concentrations in aquatic food-chains. In this section, we review the evidence that atmospheric Hg and biotic Hg levels have changed synchronously in the recent past.

6.2.1 Does Hg in aquatic biota follow the trends in atmospheric Hg emissions and deposition?

Here a number of case studies which examined temporal trends of Hg in aquatic biota are compared against the trends of Hg in atmospheric concentrations and/or deposition fluxes in the same regions. These case studies come from North America, Europe, China and the Arctic; no other regions of the

world are represented in the literature, and are thus not discussed. First, we review the literature concerning the trends of atmospheric Hg from the three study regions.

Trends in Atmospheric Hg Emissions, Concentrations, and Wet Deposition

North America and Europe: North America and Europe are considered together here because their overall atmospheric Hg concentrations and deposition fluxes have trended together over the past few decades (Zhang et al., 2016). Measured near-surface gaseous elemental mercury (GEM) concentrations in North America and Europe have declined by 30–40% between 1990 and 2010 (Slemr et al., 2011; Cole et al., 2014), a pattern that has been matched by trends in wet deposition Hg fluxes (Prestbo et al., 2009; Cole et al., 2014). By contrast, global emission inventories for the same period have suggested flat or slightly increasing total Hg emissions, because declines in Hg emitted by the energy and other industrial sectors in North America and Europe were offset by rising coal-fired power generation in Asia and by emissions from a rapidly-growing global artisanal and small-scale gold mining (ASGM) sector (AMAP 2010; AMAP/UNEP 2013). Recently, however, Zhang et al. (2016) showed that the discrepancy between emission inventories and atmospheric measurements could be resolved mainly by accounting for the declining emissions from commercial Hg-containing products since 1990 which had not been previously counted in the inventories (Horowitz et al., 2014); additional corrections were made for shifts in the speciation of airborne Hg emissions related to air pollution control technology, and by reducing the putative importance of atmospheric Hg emissions from ASGM. Calculated atmospheric Hg concentrations and trends, based on GEOS-CHEM modelling of the revised emission inventories, then agreed within error with observations (Figure 6.1). In North America and Europe, the observed and modelled atmospheric GEM trends since 1990 were –1.5 and –2.0% per year, respectively, and the trends for Hg^{II} fluxes in wet deposition were –1.6 and –1.4% per year, respectively. The agreement between the new corrected emission history by Zhang et al. (2016) and empirical atmospheric data lends confidence that the modelled atmospheric trends presented by Zhang et al. (2016) conform to reality.

The Arctic: For the Arctic region (above 60°N), atmospheric GEM concentrations have also been declining, but at a markedly slower rate than elsewhere (see Figure 6.1). The observed and modelled trend regressions also disagreed more than in other regions, with observed GEM concentrations decreasing at –0.2±0.45% per year since 1994, and the modelled rate at –1.3±0.11% per year. There are no decade-long observational datasets of Hg trends in deposition available for the Arctic or sub-Arctic; existing depositional data are confined to 1-2 years of measurements only (e.g. Sanei et al., 2010).

China: China is the largest national emitter of atmospheric Hg worldwide (Fu et al., 2015b). In contrast to the global trend, anthropogenic Hg emissions in China increased rapidly from 1978 to as recently as 2007 at an average rate of ~5.5% per year, except for 1998-2000 when the emissions decreased due to the Asian financial crisis which led to a reduction in fuel consumption (Wu et al., 2016). Mercury emissions in China are reported to have plateaued around 2007 to 2010, and showed a declining trend in the past few years (Wu et al., 2016).

Available but limited data on atmospheric Hg concentrations in the past decade in China are in general agreement with this emissions trend. Direct measurements of GEM at Guiyang, an urban site in southwest China (Fu and Feng, 2015), revealed that annual mean GEM concentrations increased at a rate of ~2.5% per year between 2002 and 2010 (Fu and Feng, 2015); GEM concentrations also increased at Mt. Changbai, a remote site in north-eastern China, at about the same rate from 2009 to 2013 but then appeared to stabilize (Fu et al., 2015b, 2016; Fig. 6.2). Mercury passive sampling and plant biomonitoring on the Tibetan Plateau suggested that atmospheric Hg concentrations were stable during 2006 to 2009 and decreased during 2010 to 2015 (Tong et al., 2016).

For the purposes of this review, the reported trends in atmospheric Hg concentrations and wet deposition by Zhang et al. (2016) and Wu et al. (2016) are taken as the basis for our comparison with aquatic biota Hg trends over recent decades. The key test of agreement between atmospheric and biotic datasets will be whether the direction of trend (increasing, decreasing, or stable) is the same in both.

Biological Hg Trend Cases Studies

Major recent studies since GMA-2013 on biotic Hg trends over the last few decades are summarized in four case studies below. ***While biotic Hg trends often follow the concurrent pattern in atmospheric Hg concentrations, there is widespread evidence for non-matching trends between them, especially in the past decade.***

Case Study 1: Fish and Birds in Lakes and Coastal Waters of North America

In the Great Lakes, Blukacz-Richards et al. (2016) evaluated the temporal trends since the 1970s of Hg levels in eggs of a piscivorous bird (herring gull - *Larus argentatus*), in two piscivorous fish (trout - *Salvelinus namaycush*, and walleye - *Sander vitreus*), and in a planktivorous fish (rainbow smelt - *Osmerus mordax*). Lipid content in bird eggs and fish tissues, and length of fish, were used as covariates in temporal statistical models. The results present a mixed temporal pattern (Figure 6.3a), with declining biotic Hg trends in all species in the first few decades (up to about 1995–2000), which matched the

declining atmospheric Hg trend in North America (see Figure 6.1), but were followed by trend reversals in most (but not all) species at some sites. In the 2000s, Hg trend reversals occurred for herring gull eggs at two sites in Lake Erie and two sites in Lake Ontario, and for lake trout in Lake Superior and at a single station in Lake Ontario. Mercury levels in lake trout continued to slowly decline at all of the remaining stations, except for Lake Huron, where the levels remained stable. Similar trends were reported by Eagles-Smith et al. (2016) when examining over 96,000 fish muscle samples from 206 species in over 4,200 lakes in western Canada and the USA. They found a significant, rapid decline in length-adjusted tissue Hg concentrations during the 1970s (from 1969 to 1977), with no subsequent significant trend up to 2012. In both of these studies, ***the authors attributed the early decline in biotic Hg to regional declines in atmospheric Hg concentration and deposition. They suggested that the subsequent trend reversal, or lack of a significant trend, could be explained by shifts in trophic dynamics resulting from invasive species, and/or geochemical changes in Hg cycling and methylation rates possibly driven by climate change.***

A more complex temporal pattern in hundreds of small Ontario lakes was reported by Gandhi et al. (2014), who found a general decline in length-adjusted fish muscle Hg concentrations from the 1970s to 1990s for northern pike (*Esox lucius*), walleye and lake trout. This decline was followed by relatively small increases in some lakes starting about 1995–2000. The initial declines in the 1970s and 1980s were more rapid in most lakes than during the 1990s, and were more pronounced in northern Ontario lakes than in southern Ontario lakes at that time. In fact, northern Ontario boreal forest lakes displayed significant overall muscle Hg declines from 1974 up until 2012 for walleye and northern pike, but not for lake trout which were relatively constant over time. In contrast to the Great Lakes studies discussed above, Gandhi et al. (2014) found that southern Ontario lakes displayed non-significant changes between 1974–2012 in walleye, pike and lake trout. Furthermore, the recent increasing trends were also more pronounced in northern Ontario lakes than in southern Ontario lakes which were nearly constant or weakly increasing, and more so in northern pike and walleye than in lake trout.

Different patterns were, however, reported by Tang et al. (2013), which examined changes in muscle Hg in 5 piscivorous fish (walleye, northern pike lake trout, burbot (*Lota lota*), and smallmouth bass (*Micropterus dolomieu*) and 2 benthivorous species (lake whitefish (*Coregonus clupeaformis*) and white sucker (*Catostomus commersonii*)) from 873 Ontario lakes based on data collected from the Ontario Sport Fish Contaminant Monitoring Program. In contrast to the declining patterns in walleye and northern pike in northern Ontario lakes reported by Gandhi et al. (2014), no significant decreases over

recent decades were observed in any of the 7 species in this study; instead, mean concentrations were found to be slightly higher in 2005-2010 than in 1974-1981, and were significantly so in northern pike. The reason for the difference between these two studies is unknown.

Substantial reductions in muscle Hg were reported between 1972–1974 and 2011 in a marine fish species, the bluefish (*Pomatomus saltatrix*), caught off the northeast coast of the USA (Figure 6.3b). Although no data were available for the period 1974 to 1993, it is clear that a ~30–40% decline in bluefish Hg concentrations in New York and New Jersey waters occurred at some period between 1972–1974 and the mid-1990s (Cross et al., 2015). Subsequently, however, the New York regional data suggest no further change in fish Hg levels up to 2007.

Most of the above studies did not include stable C and N isotopic data, making it impossible to investigate whether changes in feeding behaviour (prey trophic level and feeding location) influenced the Hg trends. The value of including trophic dynamic information based on stable C and N isotopic data in the interpretation of Hg temporal trends was clearly demonstrated by Burgess et al. (2013) in a study of Hg in herring gull eggs on the eastern Canadian seaboard. Between 1972 and 2008, two sites displayed a trend of significantly declining egg Hg, which is consistent with the declining atmospheric Hg deposition occurring at that time (see Figure 6.1). However, when trophic level changes over time were factored into the analysis using $\delta^{15}N$ isotope data, it was found that the Hg declines were due to feeding behaviour shifts. $\delta^{15}N$ is a widely-used indictor of the trophic level of species' prey selection, and was highly correlated with egg Hg in the birds. ***The authors concluded that Hg in coastal waters in that region had remained relatively constant over the last few decades despite the reduction in airborne Hg fluxes.***

Case Study 2: Fish in Swedish Lakes

Åkerblom et al. (2014) assessed the Hg temporal trends in 15 species of fish (mainly northern pike, Eurasian perch (*Perca fluviatilis*), and Arctic char (*Salvelinus alpinus*)) during the past 50 years based on almost 45,000 observations from 2881 lakes throughout Sweden. To allow for trend analysis, individual Hg concentrations of fish from any species were normalized to a standard 1-kg pike in the same lake. The average Hg concentrations in such 1-kg pike equivalent fish were found to have increased during the 1970s and peaked at the end of the 1980s before decreasing sharply between 1990 and 1996. During the late1990s, Hg levels increased again and, after peaking by 2003, they appeared to have decreased up to present (Figure 6.4A). Overall Hg levels decreased approximately 1% per year since 1970,

corresponding to a decrease of about 30% over 40 years. ***This trend matches well with the general declining atmospheric Hg trend over Northern Europe*** (see Figure 6.1). Also of note is that fish Hg levels in "limed" lakes across Sweden were consistently higher than in the lakes that were never-limed (Åkerblom et al. 2014), pointing to a significant effect on fish Hg from pH or other indirect ecosystem effects caused by the recovery of the limed ecosystems. However, the mechanism responsible for this pattern was not investigated. The temporal trends in both limed and non-limed lakes were similar.

Further analysis of data from the latest decade (2003–2012; Figure 6.4B) revealed that while there was an overall significant decreasing trend in southwestern Sweden (up to 10% per year), the trends were weaker, mostly not significant, and in a few cases even increasing in northern Sweden. In one lake (Spjutsjön), fish Hg concentrations increased steeply at a rate of about 20% per year. ***The authors noted that the more prominent decrease in fish mercury in the south matches with a larger decrease in atmospheric Hg loads in the south compared to the north of Sweden, and attributed the significant increase in Spjutsjön to possible local anthropogenic sources of Hg.***

Case Study 3: Fish in Reservoirs: North America and Europe vs China

Some of the longest time series of aquatic Hg data exist for man-made reservoirs due to concerns about the effects of impoundment on Hg methylation rates and thus on fish Hg levels. Although these reservoirs are not natural habitats for aquatic life, they contain abundant fish and invertebrate communities, and support important recreational fisheries in some areas and large aquaculture operations in others.

Studies in North America and Europe have shown that following the impoundment, the large influx of flooded vegetation and organic matter in submerged soil stimulates microbial methylation of Hg, resulting in sharp increase in fish Hg due to biomagnification of methylmercury (St. Louis et al. 2004; Hall et al. 2005; Lucotte et al., 1999; Bodaly et al., 2007). Hg methylation rates and hence fish Hg levels typically decrease as the reservoir ages and the organic matter further decomposes (Bodaly et al., 2007). This was clearly demonstrated in a recent analysis of the temporal trends of Hg in a range of fish species from 883 reservoirs across western North America (Willacker et al. 2016). Temporal patterns (normalized for confounding variables such as species and body length) were clearly related to the time elapsed since reservoir impoundment, with maximum fish Hg concentrations being reached on average three years after the impoundment (Figure 6.5). Fish Hg levels thereafter declined relatively rapidly for 4–12 years, followed by a monotonic slow decline that last many decades. Because the reservoirs were

built at different dates over the last century and a half, it may be concluded that the fish Hg pattern is not related to changing atmospheric Hg deposition over the last few decades. Instead, water storage management is shown to be a key factor influencing this temporal pattern. Fish in reservoirs that experienced maximum drawdown during summer months (May–July) exhibited significantly (up to 11-fold) higher concentrations than fish in reservoirs in which drawdown occurred during other times of the year (Willacker et al., 2016).

Reservoirs in China, however, present a different story. Different from reservoirs in North America and Europe which are typically inhabited by native fish populations used for recreational purposes, reservoirs in much of China support important aquaculture activities with fish harvested for human consumption. The fish in Chinese reservoirs thus tend to grow faster and be harvested while young. Therefore, the Hg concentrations in fish from these reservoirs are typically low due to biodilution. Unfortunately, monitoring of fish Hg concentrations in most of the Chinese reservoirs only started recently, making it impossible to deduct long-term temporal trends. One exception to this is the reservoirs in the Wujiang Basin in southwest China, where extensive studies have been carried out in the past decade. Since these reservoirs vary greatly in their ages (time since their initial impoundment), an interesting evolution scheme in fish Hg concentrations starts to emerge when the data from all the reservoirs are pooled together.

The Wujiang (Wu River) is the largest tributary of the upper Changjiang (Yangtze River). Since the 1960s, numerous large cascade reservoirs have been or are being constructed in the Wujiang Basin, including Wujiangdu (built in 1979), Dongfeng (1994), Puding (1994), Yingzidu (2003), Suofengying (2003), Hongjiadu (2004), and Pengshui (2008) on the main stream, and Aha (1960), Baihua (1966), and Hongfeng (1966) on its tributaries (Figure 6.6). Although impoundment was found to have significantly increased fish Hg concentrations in a newly constructed reservoir (Pengshui) (Li et al., 2013), fish Hg concentrations in this and another newly constructed reservoir (Hongjiadu) (Yao et al., 2011) were much lower than those in newly built reservoirs in North America and Europe (Yao et al., 2011; Li et al., 2013). For the much older Baihua Reservoir, no statistically significant differences were observed in Hg concentrations in common carp (*Cyprinidae*) among the four sampling campaigns from 2003 to 2011, more than 40 years after the impoundment (Liu et al., 2012). In general, Hg concentrations in various fish species studied, including carnivorous, omnivorous, planktivorous, and herbivorous fish, are remarkably low in all these reservoirs regardless of the age of the reservoir (Yao et al., 2011; Li et al.,

2009; Yan et al. 2010; Liu et al. 2012; Li et al., 2013), often an order of magnitude lower than the World Health Organization (WHO) guideline of 0.5 μg/g (wet weight) (WHO, 1990).

While biodilution and simple (short) food web structures clearly contribute to the generally low fish Hg concentrations (Yao et al., 2011; Meng et al., 2010, 2016; Feng et al., 2009a, 2009b; Larssen, 2010; Liu et al., 2012; Yan et al., 2010), comparisons of fish Hg concentrations in reservoirs with different ages in the same basin reveal three distinct stages of evolution due to changes in the source and concentration of organic matter in the submerged soil/sediment as the reservoir ages and cage aquaculture activities increase (Figure 6.7). As much of the Wujiang Basin is located in a karst environment, the organic matter contents in the submersed soils (typical range: 1.9 – 4.1%) are much lower than those in submersed soil (typically 30 – 50%) from the boreal forest or wetlands in North America and Europe (Yao et al., 2011; St. Louis et al. 2004; Hall et al. 2005; Lucotte et al. 1999). In addition, the water was lightly alkaline in most of the reservoir water as the result of the karstic geology of the Wujiang River, which could restrain the Hg methylation (Meng et al., 2010; Yao et al., 2011). Primary productivity in the newly constructed reservoirs in the Wujiang Basin is also low (oligotrophic–mesotrophic) due to the absence of cage aquaculture fishing (Yao et al., 2011; Meng et al., 2010, 2016), and thus autochthonous contribution to organic matter is also very limited (Jiang 2005; Yao et al., 2011). Therefore, in contrast to their counterparts in Europe and North America, the newly constructed reservoirs in the Wujiang Basin are not active sites of net Hg methylation due to the low organic carbon content in the submersed soils and/or low primary productivity (Yao et al., 2011; Meng et al., 2010). Consequently, the newly constructed reservoirs, such as Suofengying, Hongjiadu, and Yingzidu in the Wujiang River, are not a net source of MeHg and instead represent a net sink (Guo, 2008) (Figure 6.7a).

As these reservoirs become more productive (mesotrophic to eutrophic) with time, the organic matter content in the sediment increased due to continuous increases in autochthonous productivity due to the cage aquaculture activities. This would tend to promote in-situ Hg methylation, and as such reservoirs at this stage (e.g., Dongfeng and Puding) have transited from a net MeHg sink to MeHg source (Guo, 2008; Feng et al., 2009a,b; Zhang et al., 2009) (Figure 6.7B).Over the long-term evolution of the reservoir, primary productivity continues to increase and the reservoir will eventually become eutrophic. Phytoplankton-derived organic matter, and the fish feed and faeces, become significant sources of organic matter input to the surface sediments, as shown in Wujingdu (Meng et al., 2010, 2016; Zhang et al., 2009; Feng et al., 2009a). The increased oxygen consumption during fresh organic matter degradation causes progressively more anoxic conditions at the sediment-water interface (Meng et al.,

2010, 2016), which promotes microbial Hg methylation processes (Figure 6.7C), as shown in Wujingdu (Guo, 2008) where both the surface sediment and the hypolimnetic water are sites of net MeHg production (Meng et al., 2010, 2016; Feng et al., 2009a). Thus, in contrast to fish in North American reservoirs, and in spite of the relatively high atmospheric Hg loading across much of China, fish Hg levels in Chinese impoundments reflect within-impoundment processes, especially organic matter loadings to sediments, water/soil quality, food web structure, and biodilution, rather than atmospheric inputs.

Case Study 4: The Arctic

Rigét et al. (2011) summarized all available temporal Hg datasets on Arctic biota up to about 2009, and found that some species in some locations had shown significant increases over recent decades, whereas others with closely adjacent or overlapping distributions exhibited non- significant changes. Most of the increasing biotic Hg trends occurred in marine species in the North American and west Greenland sector of the Arctic, whereas declining trends were mostly observed in east Greenland and European Arctic biota. This regional dichotomy is clearly seen in the hair of polar bears (*Urus maritimus*), and has been suggested to be due to increased emissions from Asia entering the western Arctic coincident with decreasing emissions from North America and Europe in the eastern Arctic (Dietz et al., 2006).

A few additional studies have been published since then. Rigét et al. (2012) analysed temporal trends of Hg in livers of ringed seals collected from the early 1980s to 2010 from Greenland. Increasing levels of Hg were found in ringed seals in two out of three Greenlandic seal populations (Central East and Northwest Greenland), rising at a rate of 10.3% per year and 2% per year, respectively. In addition to age and trophic positions, the study showed that the Atlantic Oscillation Index, a parameter related to climate change, was positively associated with Hg concentrations in seals although the specific mechanism involved was not clear.

By analysing Hg in the teeth of polar bear from Svalbard in the Norwegian Arctic, Aubail et al. (2012) reported a decreasing trend in Hg concentrations over the period 1964–2003 (Fig. 6.8A). Since no temporal changes were found in tooth $\delta^{15}N$ and $\delta^{13}C$, they concluded that the decrease of Hg was not due to changes in trophic dynamics; instead, it was more likely due to a lower environmental Hg exposure in the region. McKinney et al. (2017) also reported a significant declining trend in hair Hg of the southern Beaufort Sea (SBS) polar bear population, at an average rate of -13% per year, between 2004 and 2011. This dataset differs from the general west-east pattern in Arctic biota Hg trends noted

above. However, only males in the SBS area exhibited significant decreases; females from the same area showed no significant trend. Mercury levels in the bears' main prey (ringed seal) also did not change up to 2007 (Gaden et al., 2009), which argues against changes in Hg inputs or the biogeochemical Hg cycle as contributing to the decline. Analyses of body condition and diet led to the conclusion that the bears' Hg trend was due to changing foraging patterns over time and not to alteration in environmental Hg levels (McKinney et al., 2017).

Braune et al. (2014) reported the temporal trend of Hg in thick-billed murre (*Uria lomvia*) eggs from Coats Island, northern Hudson Bay, and Prince Leopold Island in Lancaster Sound, Nunvut. Although there was no significant change in Hg concentrations in murre eggs from Coats Island from 1993 to 2013, $\delta^{15}N$ values for the eggs were found to be decreasingly significantly, suggesting a decline in trophic position for the bird due to the switch of its diet from Arctic cod to capelin. After adjusting egg Hg concentrations for the decline in trophic position, time trends in Hg concentrations at Coats Island changed from non-significant to significantly increasing. In contrast, at Prince Leopold Island, after adjustment for trophic position the egg Hg time trends changed from nonsignificant to significantly decreasing over the same period. These results suggest that in addition to trophic change in diet, there may have been other geographic factors at play that influenced Hg concentrations at the base of the marine food web, such as differences in Hg deposition, or in Hg bioavailability related to climate change.

Subsequently, Braune et al. (2016) updated the Hg trends in High Arctic seabird eggs at Prince Leopold Island to 2014 for five species: thick-billed murres, northern fulmars (*Fulmarus glacialis*), black-legged kittiwakes (*Rissa tridactyla*), black guillemots (*Cepphus grylle*), and glaucous gull (*Larus hyperboreus*). The first three species' eggs had been collected from the Island as early as 1975, while the guillemots and gulls were sampled from 1993 to 2013. Egg Hg trends were adjusted for possible shifts in tropic position of the birds using $\delta^{15}N$ data. Adjusted Hg concentrations in eggs of murres, fulmars and kittiwakes increased from 1975 to the 1990s, followed by a plateauing or slight decline of levels from the 1990s to 2014 (Figure 6.8B). However, the kittiwake trend was strongly influenced by the 1975 samples; when these were excluded, kittiwake eggs actually displayed a significant decreasing trend from 1976 to 2013. Trends in the eggs of murres, fulmars, kittiwakes, and guillemots had negative slopes between 1993 and 2013. The pattern in glaucous gull eggs was unique: decreasing by 50% from 1993 to 2003 before starting to increase again.

Braune et al. (2016) concluded that the general increasing trends in egg Hg during the 1970s and 1980s were consistent with atmospheric Hg increases over the Arctic *during that period.* They noted that the migratory habits of the five bird species, which overwinter in different southern regions away from Lancaster Sound, complicated interpretation of the reasons for the temporal trends. Environmental Hg changes in their wintering areas could have been different to those in the Arctic. Interpretation is also complicated by significant differences in the findings from glacier archives of atmospheric Hg on the western and eastern edges of the North American Arctic. Greenland glacial snow/firn (Faïn et al., 2009) showed a monotonic decline in atmospheric GEM concentrations during the 1970s and 1980s, following peak levels in the 1950s to 1960s. Glacial snow and ice core reconstructions of atmospheric Hg deposition from Mt. Logan (Yukon) showed increases in deposition through the 1990s, which could be an indication of increasing trans-Pacific contamination from Asia (Beal et al., 2015). Overall, these data, especially the declining GEM trend on Greenland through the 1970s and 1980s, are inconsistent with Braune et al.'s (2016) conclusions. However, the flat or slightly declining egg Hg data from about 1990 onwards is consistent with the recent remodelling of atmospheric GEM in the Arctic (see Fig. 6.1). Zheng (2015), on the other hand, reported that 20th century total Hg accumulation in a Greenland ice core was relatively constant until it increased during the 1970s to 2000s, a pattern similar to those in most of the bird species but not in agreement with the Zhang et al. (2016) modelling. Thus, uncertainty about the actual trends in Arctic atmospheric Hg deposition is a limiting factor in assessing agreement between environmental and biological Hg trends in this region.

In Great Slave Lake in the western Canadian Arctic, temporal trends of Hg in lake trout, burbot, and northern pike were monitored irregularly between the late 1980s or early 1990s and 2012 (Evans et al., 2013; Fig. 6.8C). Muscle Hg data were adjusted for fish length, but not for trophic shifts over time. Mercury concentrations generally increased over time in lake trout and burbot, but not in northern pike, with considerable inter-annual variation. These increasing or flat patterns are inconsistent with atmospheric GEM concentrations and wet deposition fluxes that were declining at the time (see Figure 6.1), and with the Mt. Logan atmospheric deposition record of Beal et al. (2015). Statistical analysis of climate factors suggested that varying annual mean air temperatures, and particularly cold season temperatures, were related to the fish Hg patterns although a precise mechanism linking temperature to fish Hg could not be elucidated (Evans et al., 2013).

6.2.2. What Causes Decoupling between Aquatic Biota and Atmospheric Hg Trends?

In contrast to the recent decadal datasets described above, the available century-scale biotic Hg trends (from the Arctic; Dietz et al., 2009) generally matched remote glacial ice core archives of atmospheric Hg deposition and GEM concentrations (Zheng, 2015; Beal et al., 2015; Kang et al., 2016). In both cases, starting about the late-19th century, shortly after major anthropogenic uses and emissions of Hg became more common, Hg concentrations in the atmosphere and in aquatic biota increased steadily up to about the 1970s to 80s. Subsequently, as atmospheric and biological monitoring became more widespread and frequent, it became increasingly apparent that decoupling between the aquatic biotic and atmospheric Hg trends has been occurring in some areas and in some species within specific areas, especially in the past decade. Fundamentally, this decoupling can be generally attributed to the exceptional sensitivity of the Hg biogeochemical cycling to changes in climatic (e.g., temperature, light, hydrology), geochemical (e.g., pH, redox status, complexing ligands), biological (e.g., feeding behaviour of an organism) and ecological (e.g., organic carbon flux, microbial processes, and food web structure and dynamics) conditions (Table 6.1). Some of the major processes that trigger changes in these conditions and thus the decoupling between biotic and environmental Hg include:

Landscape changes: Major changes in landscape, such as flooding, damming, and deforestation, not only increase Hg flux from the terrestrial system to the aquatic system, but more importantly they change the organic carbon flux and redox conditions that directly control the Hg methylation process and mobilize Hg stored in soil organic matter This process is clearly demonstrated by the construction of reservoirs where biotic Hg concentrations are almost exclusively controlled by organic carbon dynamics and bear no relationship with Hg trends in the atmosphere (see Case Study 3).

Ecosystem changes: As methylmercury biomagnifies in the food web (i.e., methylmercury concentration increases from prey to predator), any changes in ecosystem structure, function and dynamics would result in major changes in Hg concentrations within the ecosystem. Processes such as acidification (Case Study 2) and eutrophication (see Case Study 3) affect not only methylmercury production by altering Hg speciation and bioavailability, but also Hg food-chain transfer and thus biotic Hg concentrations by altering species composition, biomass and growth rates (e.g., Clayton et al., 2013; Jardin et al., 2013). Aquaculture, overfishing, and invasion of non-native species can change not only the nutrient status of an aquatic ecosystem, but also change directly the structure, function, and dynamics of food webs, and thus could result in major changes in biotic Hg.

Climate change: On the global scale, climate change is the most prevalent contributor to the decoupling between biotic and environmental Hg. The impact of climate change on biotic Hg is perhaps most profoundly felt in the Arctic, where rapid climate warming has resulted in dramatic changes in many biogeochemical and ecological processes that drive Hg cycling (Wang et al., 2010; Stern et al., 2012). For instance, the rapid decline in the aerial coverage and thickness of Arctic sea ice and the replacement of multi-year sea ice with first-year ice have been shown to influence Hg distribution and transport across the ocean–sea ice–atmosphere interface, alter Hg methylation and demethylation rates, promote changes in primary productivity, and shift food web structures (bottom-up processes). In addition, changes in animal social behaviour associated with changing sea-ice regimes can affect dietary exposure to Hg (top-down processes) (Stern et al. 2012). As shown in Case Study 4, thick-billed murre from Coats Island in northern Hudson Bay has been shown to have moved down in its trophic position in the food web, presumably due to feeding increasingly on capelin instead of Arctic cod (Braune et al., 2014). However, the population's egg Hg concentrations did not change significantly from 1993 to 2013; thus, to explain this stable trend the availability of methylmercury in the environment and efficiency of Hg food web transfer must have increased. It has also been suggested that climate warming may cause a shift in energy flow from benthic to pelagic food webs as aquatic productivity increases in High Arctic lakes. Since zooplankton species such as Daphnia contain higher methylmercury than benthic organisms, this shift could increase Hg transfer in the food web (Chetelat and Amyot, 2008). The impact of climate change on biotic Hg has also been observed in lower latitude regions (e.g., Pinkney et al., 2014).

6.2.3. What are the implications for the Minamata Convention?

Recent reports about widespread biotic Hg trends not following the atmospheric Hg trends is not discouraging news when it comes to implementation of the Minamata Convention. The fact that the effectiveness of Hg emission control is expected to be followed by long delays before an ensuing reduction is seen in food-web Hg levels makes it all the more pressing to control and reduce mercury emissions as early as possible (Wang et al. 2010).

Wang et al. (2010) and Wang and Zhang (2013) proposed that the decoupling between biotic and environmental Hg is an indication that an aquatic ecosystem has entered a new "paradigm" in which the key controls on Hg bioaccumulation have switched from being "emissions-driven" to "processes-driven" (Figure 6.9). This switch occurs because the level of Hg in an aquatic ecosystem is determined not only by Hg influx (natural or anthropogenic) to the system, but also by the processes in the ecosystem that control the recycling, speciation, bioavailability, methylation and biological uptake of Hg. As the

accumulated mass of Hg in a water body becomes large enough relative to the emission-driven loading rate, the internal biogeochemical processes that control its permanent removal (e.g., burial), re-emission, or uptake into the biosphere would increasingly become the determining steps in bioaccumulation.

The changing relationship over time between atmospheric Hg concentrations or deposition, and biotic Hg, is shown in Figure 6.9. During the Holocene, when Hg emissions were at their natural level, the flux of Hg to the aquatic system was generally low, and so were its biotic concentrations (Phase I – "Holocene background"). At the onset of the Anthropocene in the 19th century, however, as industrialization resulted in a sharp increase in anthropogenic Hg emissions, aquatic biota Hg concentrations responded rapidly due to increasing Hg deposition, exposure and uptake of Hg from a small but growing environmental Hg inventory (Phase II – "Emissions-driven"). Once an aquatic ecosystem has accumulated sufficient Hg, additional increases in Hg influx become secondary to the amount that has been stored in the system accumulated by years of loading ("legacy" Hg). Bioaccumulation then draws predominantly on this "legacy" Hg, which is operated on by the internal biogeochemical processes (Phase III –"Internal Processes-driven"). Throughout all these three phases, biogeochemical processes (shown as sine-wave ''noise" in Figure 6.9) determines the transport of Hg from the abiotic part of the ecosystem to biota, but it is in Phase III that these processes emerge to create a variability that is large enough to obscure the external Hg emission trends, and hence the mismatch between biotic and atmospheric Hg trends (Wang et al., 2010).

In the context of the Minamata Convention to control Hg emissions, a new phase, Phase IV, can be envisioned (see Figure 6.9). As anthropogenic Hg emissions decrease, atmospheric Hg concentrations will decrease and eventually stabilize at a new steady state. However, recycling of the large quantities of legacy anthropogenic Hg presently contained in the world's oceans and soils, and revolatilization between oceans, soils and the atmosphere, means that atmospheric and aquatic Hg concentrations are likely to decrease much more slowly than changes in current emissions (see Chapter 1.2). While biotic Hg concentrations are also projected to decrease over the long term, the current phase of "processes-driven" bioaccumulation dictates that it will take much longer to establish a new steady-state in biotic Hg. The biotic Hg concentrations at the new steady-state are also likely to remain above the Holocene background levels. In the shorter term, however, aquatic biotic Hg concentrations, especially in marine ecosystems, are likely to continue to increase despite recent emission controls (Sunderland and Selin,

2013). Biota in smaller waterbodies such as lakes and coastal marine systems with restricted water mass turnover are more likely to respond relatively rapidly to emissions controls.

Examples of this long and "bumpy" recovery in biotic Hg can be found following the impoundment of a river, or following "de-acidification" of a lake. As shown in Case Study 2, fish Hg in reservoirs decreases a few years after the impoundment, but remains above the pre-impoundment level even after more than a century (see Figure 6.5). In the 1970s, liming was applied to many Swedish lakes that were acidified due to atmospheric acid deposition to help restore the lake ecosystem. Following the liming, fish Hg in those lakes declined 10-20% by the 1990s (Meili, 1995) and continued to decline to the present day (Åkerblom et al., 2014). Yet, more than 30 years after the liming, fish Hg concentrations in these lakes remained considerably higher (twice as high on average) than those in lakes that were not impacted by acidification (and not subjected to liming) (Åkerblom et al., 2014) (see Figure 6.4a).

Therefore, as anthropogenic Hg emissions are being placed under control due to the Minamata Convention, research and management emphasis should focus on the fate and effect of legacy Hg that is already stored in environmental reservoirs, and on the factors and processes that affect the recovery time of biotic Hg. Given the long and bumpy recovery road ahead, effective remediation and adaptation strategies are needed to assist the local communities that facing Hg contamination in their ecosystems and food sources.

11659 **Table 6.1** Unique properties of mercury and implications for its biogeochemistry (Wang and Zhang, 2013)

Property	Implications
Redox between Hg(0) and Hg(II)	Sensitive to changes in pe and pH; Sensitive to photochemical and microbial processes.
High vapor pressure of Hg(0)	Sensitive to changes in temperature; Long range atmospheric transport; A global problem needing global solutions.
Hg^{2+} ions being one of the softest Lewis acids	Strong affinity to ligands (e.g., reduced sulphides, halogens); Sensitive to changes in organic carbon.
Methylation is primarily microbial, with MeHg being the most bioavailable and toxic	Sensitive to changes in organic carbon, nutrients, redox and microbial processes; Direct source control of MeHg difficult.
MeHg biomagnifies in the food chain	Sensitive to changes in food web structure and dynamics.

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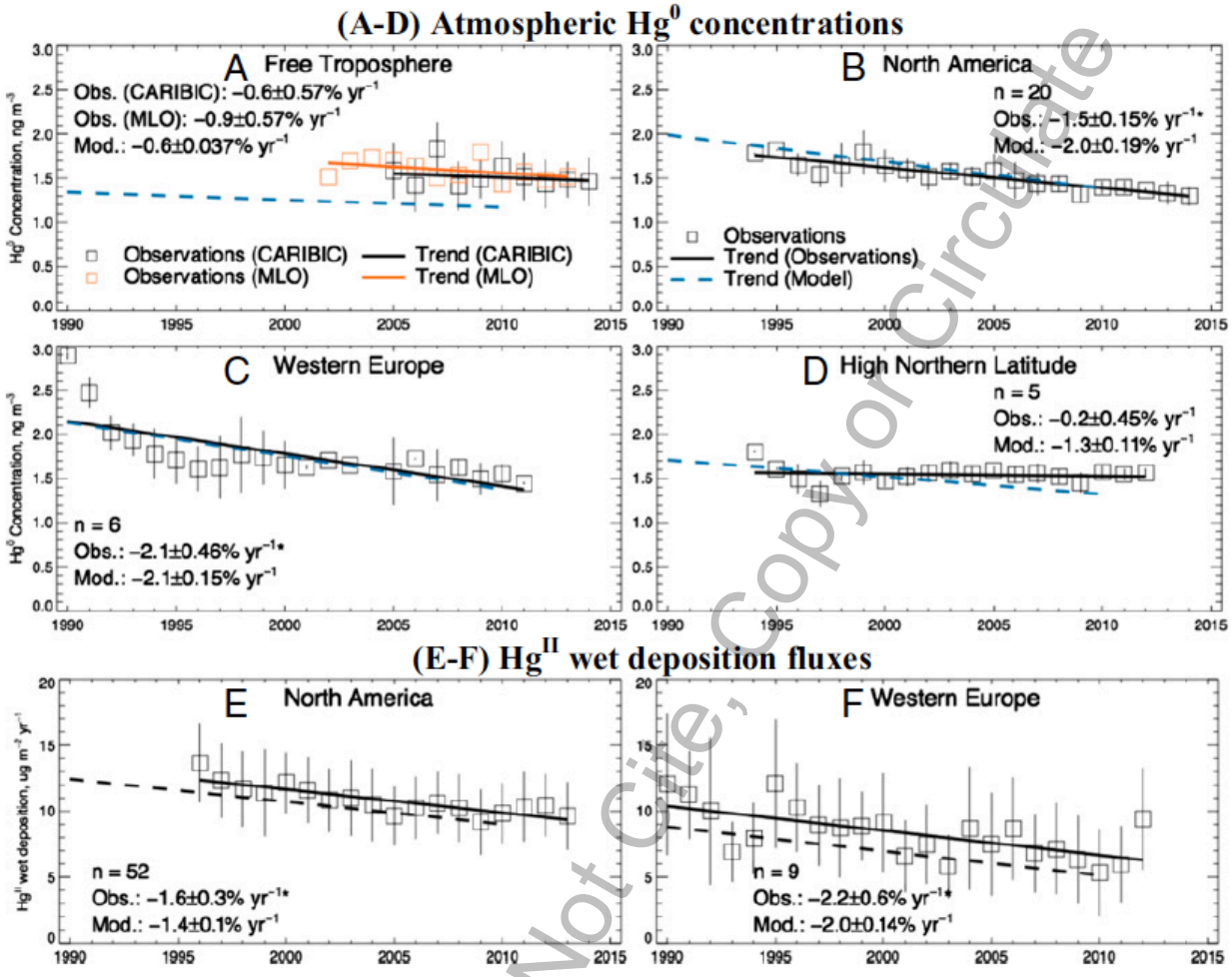

Figure 6.1. Observed and modelled trends for 1990 to 2013 in atmospheric Hg(0) concentrations (**A-D**) and Hg (II) wet deposition fluxes (**E** and **F**) in different regions of the northern hemisphere.Observations for individual years are shown as squares with linear regression as solid line. The dashed line is the trend from the GEOS-CHEMsimulation using the revised anthropogenic emissions inventory for 1990 and 2010. The data are averaged regionally across for the free troposphere (**A**), North America (**B**and **E**), Western Europe (**C** and **F**), and high northern latitude regions (**D**) (vertical bars show the SDs).Regression coefficients (slope ± SE) and number of sites (**n**) are given (**Insets**). From Zhang et al. (2016)

N.B., All captions will be recast, and figures redrawn by UNEP copy editors and graphic designers.

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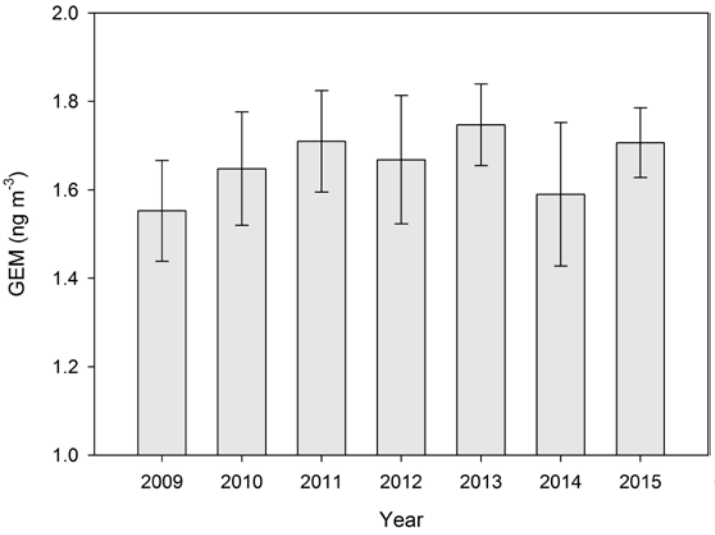

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11678 **Figure 6.2**. Annual mean gaseous elemental mercury concentrations measured at Mt. Changbai in

11679 northeastern China (Fu et al. 2015b)

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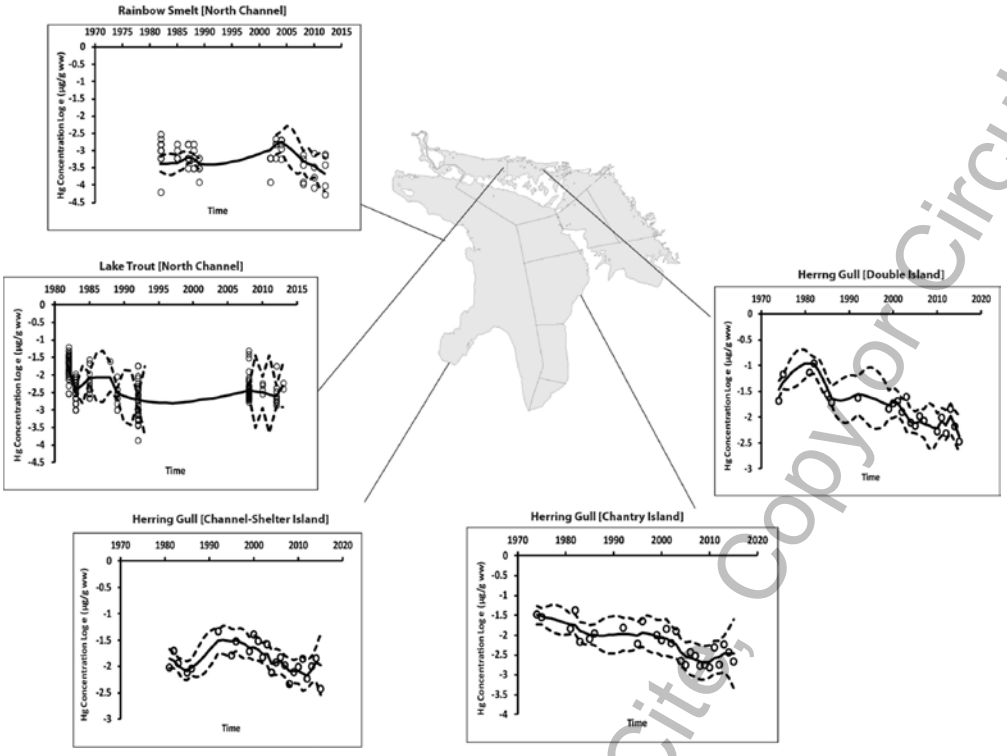

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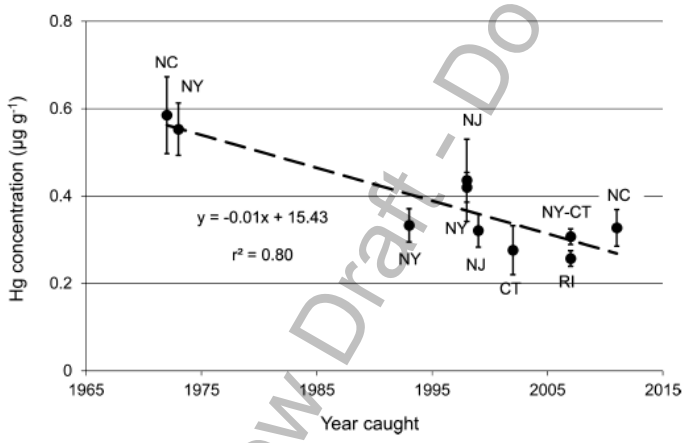

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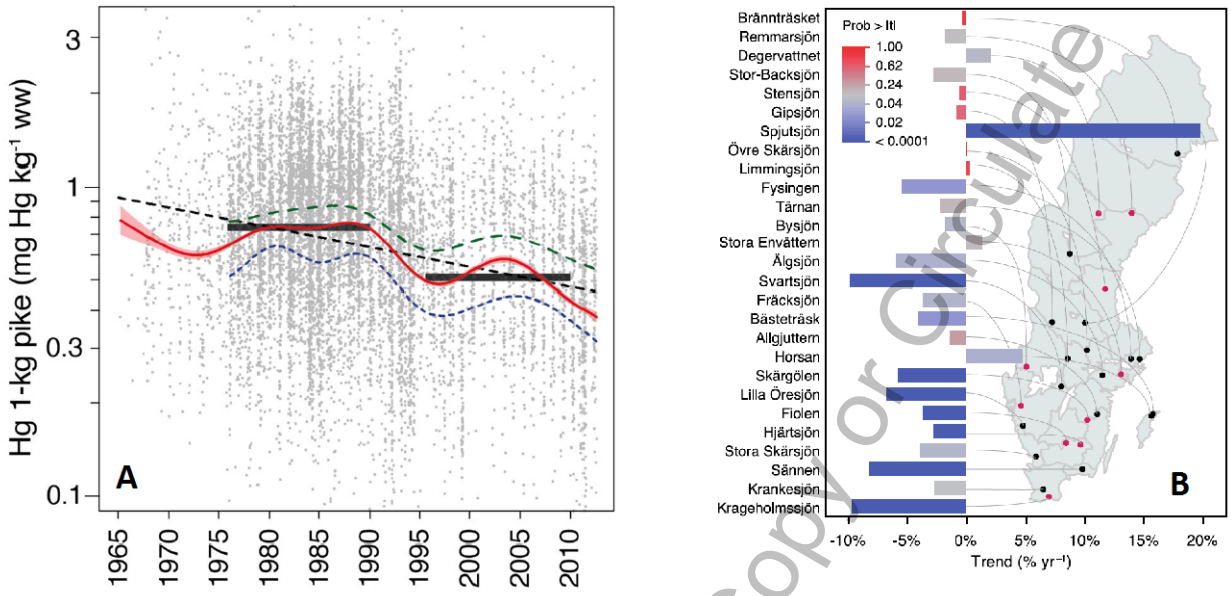
11685 Figure 6.3. Mercury trends in fish and waterfowl of North America. A) Hg concentrations (Ln-

11686 transformed mg/g wet weight for Herring Gulls, Lake Trout and Rainbow Smelt) in Lake Huron. The

solid and dashed lines correspond to the median and the 95% credible intervals of the predicted mercury concentrations (Blukacz-Richards et al. 2016). B) Mean and two SEM of the estimated mercury concentrations for bluefish from 1972 to 2011 (Cross et al. (2015). NC = North Carolina; NY = New York; NJ = New Jersey; CT = Connecticutt; RI = Rhode Island.

NOTE TO GRAPHICS: draw a basemap of North America and then show each of these (and perhaps others cited in the text) as an inset



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11697 **Figure 6.4.** Total Hg concentrations in Swedish fish 1965–2012. A) Normalized (1-kg pike) Hg
11698 concentrations of 10,176 catches; each point represents the mean from a single date and site. A linear
11699 regression model (black dashed line) and a generalized additive model (GAM; red line ± SE) were
11700 applied to all data to visualize temporal patterns. The parallel dashed lines are separate GAMs fitted either
11701 to limed lakes (upper green dashed line) or to never-limed lakes (lower blue dashed line). The black step
11702 lines indicate the geometric mean Hg concentrations between 1976–1990 (0.74 mg kg^{-1} ww) and1996–
11703 2010 (0.52 mg kg^{-1} ww). B) Recent trends of total ww Hg concentrations in medium-sized perch (total
11704 length 140–220 mm) from 27 national monitoring lakes during the period 2003 (red dots) or
11705 2005/2006/2007 (black dots) to 2012. Trends were estimated by linear regression on log-transformed Hg
11706 concentration normalized site specifically for fish age and body length. The bar colour represents the
11707 probability of individual trends being equal to zero (t test). Blue bars highly significant, red bars not
11708 significant. From Åkerblom et al. (2014).

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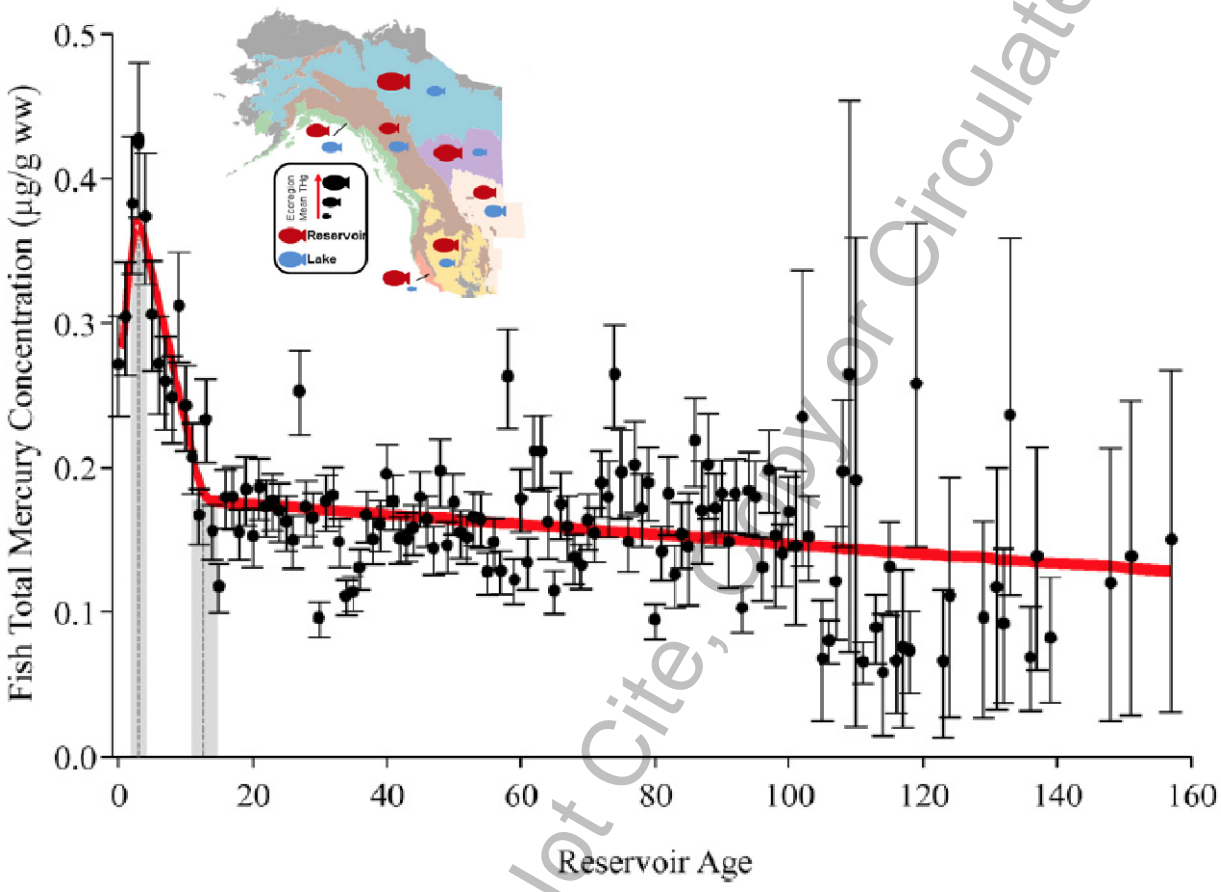


Figure 6.5. Fish tissue Hg trends from reservoirs across western North America. The data show least squares mean total mercury concentrations (µg/g ww±standard error) in size-standardized fish. Least squares mean account for the effects of ecoregion, waterbody, species, and sampling year. Vertical grey dashed lines and shaded regions indicate estimated breakpoints (±standard error) from segmented linear regression (solid line) on fish mercury concentration when accounting for the effects of ecoregion, waterbody, species, and sampling year. (From Willacker et al 2016). NOTE TO GRAPHICS: REMOVE RED LINE DURIGN REDRAW.

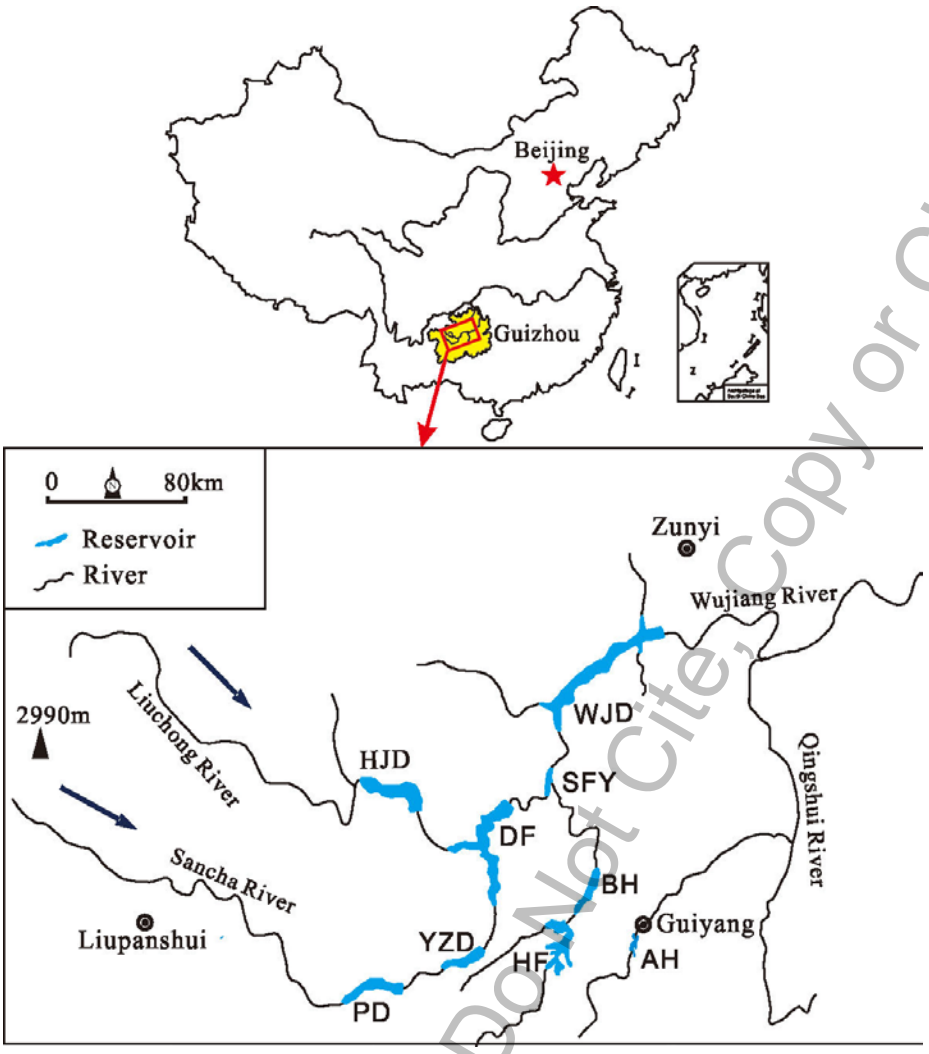
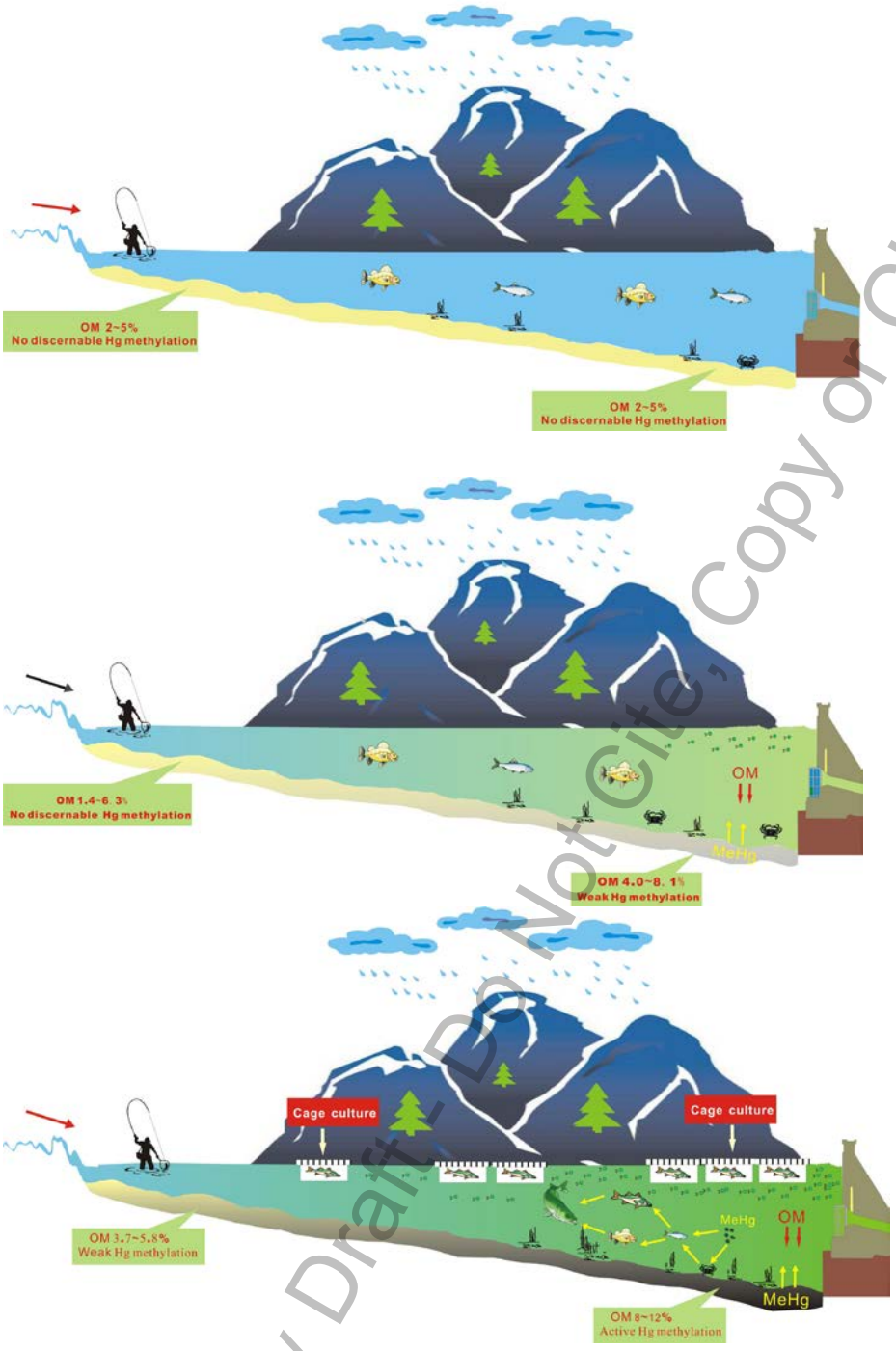


Figure 6.6. Reservoirs along the Wujiang River



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11729 Figure 6.7. Conceptual models of the Hg cycling in A) primary, B) intermediate, and C) advanced

11730 evolutionary stage reservoirs in the Wujiang River Basin, Southwest China

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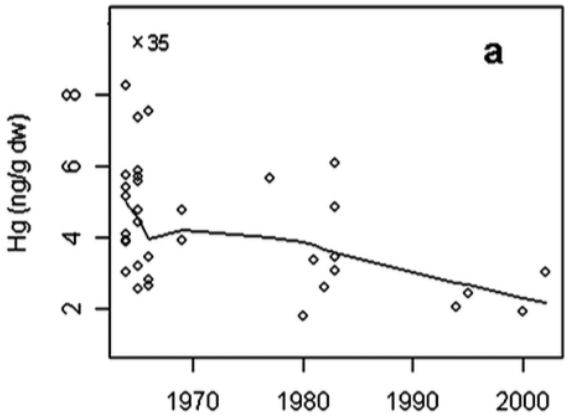

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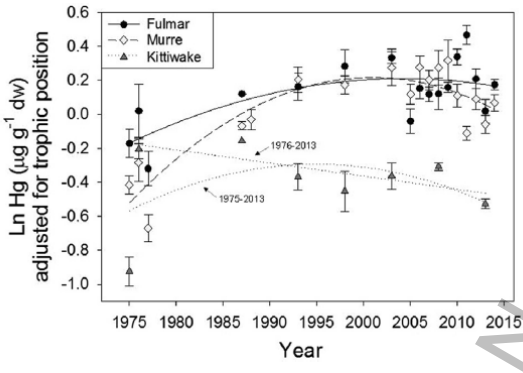

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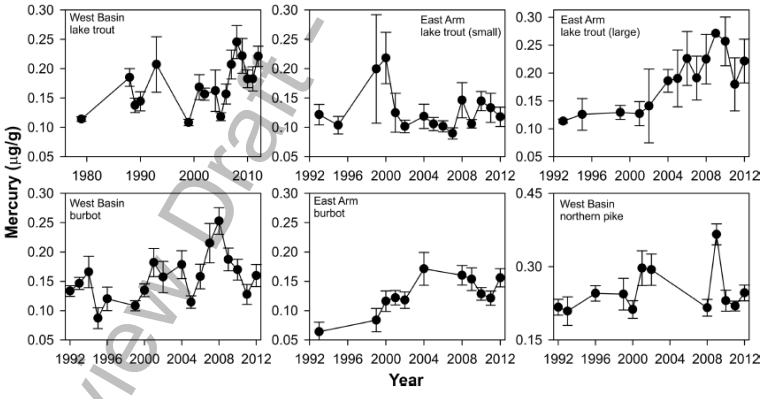

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Figure 6.8. Mercury trends in the Arctic. A) Year vs. dental Hg concentrations (ng/g dw) in polar bears from Svalbard, aged from 3 to 10 years. Smoothing lines (robust, locally weighted scatter plot smoothing system based on the LOWESS algorithm) represent the fitted non-linear trend of the values. From Aubail et al. (2012). B) Annual mean Hg concentrations (ug/g dry weight; ln-transformed) adjusted for trophic position in eggs of thick-billed murres, northern fulmars, and black-legged kittiwakes from 1975 to 2014. from Braune et al. (2016). C) Hg concentrations in burbot and lake trout collected from the West basin and east Arm of Great Slave Lake. from Evans et al. (2013). NOTE TO GRAPHICS: draw a basemap of the Arctic and then show each of these (and perhaps others cited in the text) as an inset

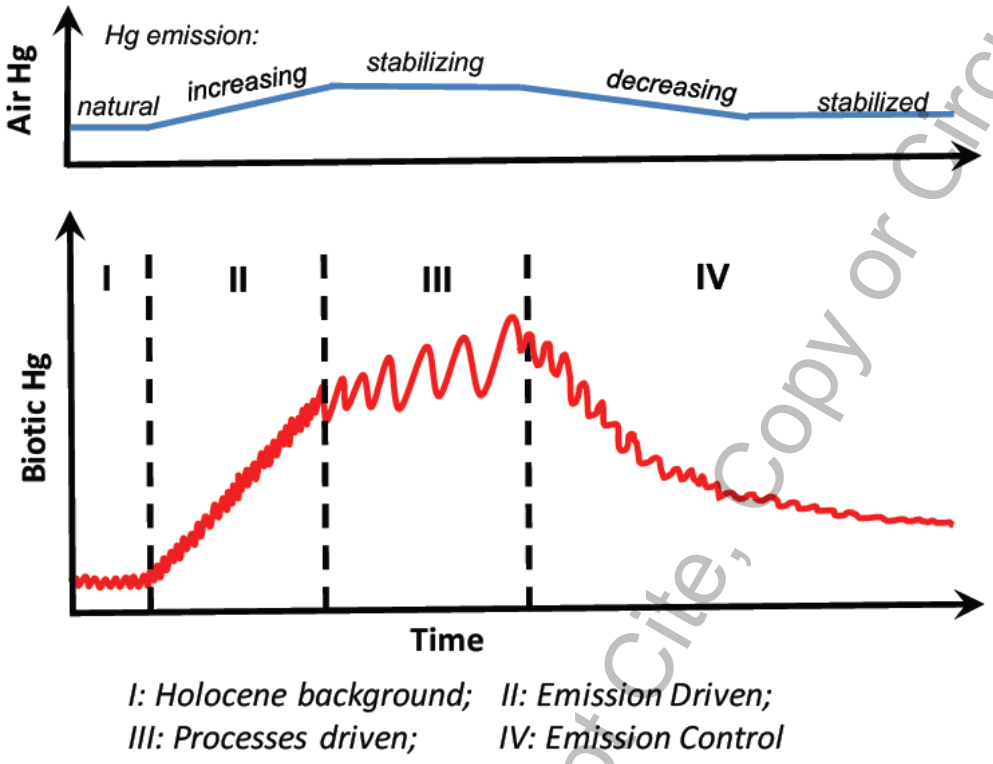

Figure 6.9. General stages in the principal drivers of mercury bioaccumulation (bottom panel), following increasing and decreasing trends in anthropogenic emissions (top panel). Modified from Wang et al. (2010).

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