Source attribution of mercury exposure for U.S. seafood consumers: Implications for policy

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Source attribution of mercury exposure for U.S. seafood consumers: Implications for policy

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Abstract

Background: Since the 1970s, policies attempting to reduce adverse effects of environmental mercury exposure from fish consumption in the U.S. have targeted reductions in anthropogenic emissions from domestic sources. Objectives: To analyze the potential effectiveness of future domestic and international emissions controls, we assess the contributions of anthropogenic, historical and natural mercury to exposure trajectories in the U.S. population over a 40-year time horizon. Methods: Our analysis is conducted using models that simulate global atmospheric chemistry (GEOS-Chem), mercury cycling among different ocean basins, and the fate, transport and bioaccumulation of mercury in four types of freshwater ecosystems. We consider effects on mercury exposures in the U.S. population based on dietary survey information and consumption data from the sale of commercial market fisheries. Results: Our results show that while U.S. emissions controls may reduce mercury exposure by up to 50% for certain high-risk groups such as indigenous peoples in the Northeast, their potential effects on populations consuming marine fish from the commercial market are not well-constrained. Conclusions: Despite uncertainties in the exposure pathway, results clearly indicate that a combination of domestic and international emissions controls with adaptation strategies are necessary to manage methylmercury risks across various populations.
1. Introduction

Mercury is a global pollutant that is ubiquitous in the environment. Once deposited to ecosystems, inorganic mercury may be converted to methylmercury (MeHg), which bioaccumulates in food webs. MeHg exposure causes severe human health effects including immune system suppression, neurodevelopmental delays in children, and compromised cardiovascular health in adults (Mergler et al. 2007). National human health data from 1999-2002 suggest that 300,000-600,000 children are born each year with blood mercury levels that exceed the U.S. Environmental Protection Agency’s (U.S. EPA) reference dose (RfD) for MeHg (Mahaffey et al. 2004; Trasande et al. 2005). In an attempt to reduce MeHg exposures, regulatory efforts to date have focused on controlling anthropogenic sources such as waste incinerators and coal-fired power plants (U.S. Environmental Protection Agency 2005).

However, the global biogeochemical mercury cycle has been significantly altered by historical mercury releases, and the timescales required for recovery are largely unknown (Lamborg et al. 2002; Mason and Sheu 2002; Selin et al. 2008; Sunderland and Mason 2007; Swain et al. 2007). Here we apply atmospheric, oceanic and freshwater ecosystem fate and bioaccumulation models to illustrate the potential contributions of anthropogenic, historical and natural mercury to exposure trajectories in the U.S. population over a 40-year time horizon (to 2050). We use this analysis to assess the expected impacts of current policies and to identify gaps in present understanding of the mercury exposure pathway.

Relative contributions of present-day anthropogenic sources to deposition vary considerably among locations (Cohen et al. 2004; Seigneur et al. 2004; Selin et al. 2008). In addition to atmospheric loading rates, fish MeHg levels depend on ecosystem-specific properties and food-web structure (Benoit et al. 2003; Harris et al. 2007; Munthe et al. 2007). Individual
choices regarding the types and amounts of seafood consumed also play a large role in determining overall exposure levels and resulting risks (Burger and Gochfeld 2004; Carrington et al. 2004; Stern et al. 2001). Effectively managing MeHg risks therefore requires information on the exposure pathway at both the local and global scale. For example, while subsistence and recreational fishers may harvest and consume fish from local water bodies, most individuals obtain the majority of their fish from the commercial market, which combines locally harvested and imported species (Carrington et al. 2004; Sunderland 2007). This means that the relationships among anthropogenic, historical and natural mercury sources, MeHg levels in aquatic systems, bioaccumulation in food webs, and consumption patterns that result in human exposure must be analyzed at multiple spatial and temporal scales. Although many elements of this exposure pathway are uncertain, policy analysis requires synthesis of our best-available understanding to quantify these processes and to determine the potential effectiveness of different mercury control strategies. Environmental modeling combines disparate atmospheric, aquatic and human health data with our best understanding of underlying processes to help assess the future effects of possible policies and regulatory decisions (National Research Council 2007).

Previous studies have analyzed potential benefits from domestic mercury emissions controls on exposure levels in parts of the U.S. population. Rice et al. (2005) analyzed both marine and freshwater exposure pathways, assuming a linear and instantaneous change in fish mercury levels with declines in atmospheric deposition. The U.S. EPA (2005) analyzed the effects of regulating emissions from coal-fired utilities on exposure of recreational fishers and their families, assuming a linear response of MeHg to emissions reductions. Trasande et al. (2005) analyzed the public health costs associated with mercury emissions from power plants in
the U.S. by assuming a linear relationship between declines in atmospheric emissions and human exposure levels.

To assess the prospects of various policy interventions for managing mercury exposure risks, we go beyond these previous studies by linking temporal trends in exposure to contributing source regions for atmospheric mercury emissions globally. We use physically meaningful simulations of mercury fate and transport in the environment for this analysis. We attribute the origin of atmospheric mercury deposition to present-day anthropogenic sources, natural sources, or historical mercury that continues to circulate in ecosystems, using a global 3-D atmospheric chemistry transport model (GEOS-Chem). We explore the timescales required for each component of atmospheric deposition to cycle through ecosystems by simulating mercury transport, speciation, and bioaccumulation. To do this, we combine source-attributed deposition from GEOS-Chem with ecosystem-scale fate and bioaccumulation models developed by the U.S. EPA (Knightes et al. 2009) and a multi-compartment global box model for mercury cycling in different ocean basins (Sunderland and Mason 2007). We use the results of these simulations to help constrain the likely source attribution of freshwater and marine fish MeHg changes over a 40-year time horizon. Using information on per-capita fish consumption rates (Sunderland and Mason 2007) and the consumption patterns of sensitive groups (Mahaffey et al. 2004; Moya 2004), we are able to analyze for both freshwater and marine pathways how changes in exposure may be affected by changes in ecological concentrations resulting from emission controls.

We attribute exposure to natural background, present-day anthropogenic emission sources in North America and internationally, or the legacy of past historical anthropogenic emissions that continue to cycle in the environment, similarly to the source attributions of Selin et al. (2008). The natural component represents the pre-industrial steady state of roughly one
third of present global emission and deposition. Direct anthropogenic emissions from North
American and international sources are also roughly a third of emission and deposition. The
historical mercury component includes not only the mercury already in aquatic systems, but also
the anthropogenic enhancement of surface emissions since industrialization that continues to
cycle through the surface and atmosphere, and comprises the remaining third of present-day
deposition. By distinguishing between exposure to mercury from these different sources, we are
able to assess both the prospects for both direct emissions reductions (decreases in the North
American and international anthropogenic sources) and the timescales of ecosystem response to
historical contamination, which is not directly addressed by contemporary emissions reduction
policies.

2. Methods

Mercury emissions, chemistry and atmospheric deposition are simulated here using the
GEOS-Chem model, described in detail by Selin et al. (2007; 2008) with updates as described by
Selin and Jacob (2008). The GEOS-Chem simulation has been extensively evaluated against
measurements of atmospheric mercury species and deposition and matches seasonal and spatial
trends (Selin et al. 2007; Selin et al. 2008). Elemental mercury (Hg(0)), the dominant (>95%)
atmospheric mercury species, has a relatively long lifetime (0.5-2 years) in the atmosphere and
can transport globally. Oxidized and particulate mercury (Hg(II) and Hg(P)) are shorter-lived
and deposit on a local to regional scale.

Direct anthropogenic releases are mainly from sources such as coal-fired power plants,
metal smelting, mining and waste incineration (Pacyna et al. 2006). Anthropogenic emissions in
GEOS-Chem are based on the global inventory of Pacyna et al. (2006) for the year 2000,
modified as described in Selin et al. (2008) to satisfy global observational constraints. The total
mercury emission in the model is 11,200 Mg y\(^{-1}\), of which 3400 Mg y\(^{-1}\) is from direct anthropogenic sources. Globally, 58% of direct anthropogenic emissions are as Hg(0), 33% are as Hg(II), and 9% are as Hg(P). Land and ocean emissions, all as Hg(0), of pre-industrial and historical origin are simulated using the coupled land-ocean-atmosphere simulation described by Selin et al. (Selin et al. 2008).

GEOS-Chem simulates wet and dry deposition of Hg(II) and Hg(P) as well as dry deposition of Hg(0) (Liu et al. 2001; Selin and Jacob 2008). For this application, we archive wet and dry deposition of Hg(II) and Hg(P), the predominant forms of atmospheric deposition influencing aquatic ecosystems. Globally, the source attribution of deposition reflects the attributions of emissions; however, these attributions vary spatially, as anthropogenic emissions have caused enrichment of mercury deposition in various regions by a factor of 2-10 (Selin et al. 2008). In GEOS-Chem, 34% of Hg(II)/(P) deposits to land and 67% to oceans, consistent with their relative areas (Selín et al. 2008). Contributions of various emissions sources to deposition in a particular location depend on both the form of mercury emitted as well as atmospheric chemical processes, transport and circulation patterns. Annual mean source attribution results are relatively insensitive to the choice of particular meteorological year. We base results reported here on meteorological data for 2004-2005.

Table 1 shows GEOS-Chem deposition and source attribution for two U.S. deposition scenarios and the global ocean basins. Contributions to deposition from natural sources are diagnosed by the pre-industrial simulation described in Selin et al. (2008). Deposition from North American and international emissions are determined from simulations with those sources shut off. We calculate historical sources by the difference between summed natural and direct anthropogenic deposition and total deposition. For freshwater ecosystems across the U.S., we use
two deposition scenarios representative of the Northeast and Southeast regions. Previous analyses have shown that mercury deposition in these regions comes from different source combinations. In the Northeast/Midwest U.S., the majority of deposition comes from North American anthropogenic sources. For the region used in this analysis (40-44°N, 72.5-77.5°W), as shown in Table 1, GEOS-Chem attributes 59% of deposition to North American anthropogenic sources, 9% to anthropogenic sources outside North America, 16% to natural sources, and 16% to historical mercury (Selin and Jacob 2008). In contrast, in the Southeast U.S. (24-28°N, 77.5-82.5°W), GEOS-Chem attributes only 11% to North American anthropogenic sources, though total measured deposition is the highest in the U.S. (National Atmospheric Deposition Program 2003). The remainder comes from international anthropogenic sources (23%), natural sources (42%), and historical mercury (24%) (Selin and Jacob 2008).

Uncertainties are inherent in any effort to model the fate and transport of mercury on a global scale, including specifying the atmospheric redox chemistry of Hg(0)/Hg(II) and quantifying the fluxes of mercury between the atmosphere and surface reservoir (Lin et al. 2006; Pongprueksa et al. 2008). While it is difficult to quantify the influence that these uncertainties have on our results, the major features of source attribution in GEOS-Chem are consistent across global mercury models. For example, Seigneur et al. (2004) report contributions from North American emissions to U.S. deposition at 25-32% on average for the U.S., with maximum values near sources in the Midwest and the lows in the Southeast in the Gulf of Mexico, which is consistent with our results reported here.

Across all ocean basins, the mean percentage of deposition attributed to present-day anthropogenic sources varies between 23-35%, though this percentage may be higher locally. For each ocean basin, we archive deposition over the geographical area corresponding to the model
compartments of Sunderland and Mason (2007), described further below, with the exception of
the Mediterranean Sea. For the Mediterranean region, we scale the source attributions from
GEOS-Chem to a concentration trajectory based on empirical estimates described in Sunderland
and Mason (2007) to account for differences in spatial resolution between the atmospheric and
oceanic models. We do this because the 4°x5° resolution of GEOS-Chem does not allow for the
Mediterranean Sea to be distinguished from the large-scale subtropical downwelling seen in
desert regions, and thus predicts higher than expected deposition based on the observational data
(Kotnik et al. 2007; Zagar et al. 2007).

Sunderland and Mason (2007) compared atmospheric deposition estimates from three
models and the influence of such variability on ocean mercury concentrations over the next
several decades. Differences between atmospheric deposition rates used here and other models
are most apparent for the Atlantic Ocean, where GEOS-Chem deposition results in slight
increases in concentrations over the next several decades compared to decreases in
concentrations based on other atmospheric models. Resolving this uncertainty requires additional
data on mercury concentrations trends in different ocean basins, which are presently extremely
limited. The relative contribution of various sources to the global oceans, however, is less
uncertain, as it reflects the global attributions of emissions sources.

For the analysis presented here, we drive the aquatic mercury fate and transport models
using contemporary atmospheric deposition rates and present-day attribution by source regions.
We use present-day emissions and deposition because future emissions scenarios are not
presently available for global-scale mercury cycling applications. This analysis helps to inform
policies about the potential magnitude and timing of ecosystem responses, but we advise caution
when interpreting results for future exposure pathways. The approach presented here may
underestimate potential contributions from the Asian continent (which are increasing) and
overestimate contributions from North America and Europe due to ongoing declines in
emissions. Global emissions have been relatively constant for the past decade, and current
projections are that the global total will remain within ±20% until 2020 (Pacyna et al. 2006). We
focus our analysis on trajectories of mercury concentrations in freshwater and marine ecosystems
over a 40-year time frame (roughly 2050), which is a medium to long time horizon in policy-
making. It will be possible to further refine these analyses as additional information on historical
and future mercury emissions and trends in environmental concentrations become available
(Mason et al. 2005; Streets and Zhang 2008).

3. Results

Temporal trends in freshwater fish mercury and associated exposure

We combine source-attributed deposition from GEOS-Chem with previously published
watershed, water body, and food web bioaccumulation models for mercury applied to four
different types of freshwater ecosystems across the contiguous U.S. (Knightes et al. 2009). Each
of these modeling frameworks is publicly available

(http://www.epa.gov/ceampubl/products.html) and has been evaluated extensively for previous
applications (Ambrose et al. 2005; Brown et al. 2007; Knightes 2008). We simulated mercury
dynamics in water bodies using the Spreadsheet-based Ecological Risk Assessment for the Fate
of Mercury (SERAFM) model and the Water Quality Analysis Simulation Program (WASP)
(Knightes 2008; U.S. Environmental Protection Agency 1997). Watershed-mercury dynamics are
based on the U.S. EPA Region 4 Watershed Characterization System Mercury Loading Model
(WCS-MLM) and land-cover characteristics (Greenfield et al. 2002; Knightes et al. 2009).

Bioaccumulation is simulated using the Bioaccumulation and Aquatic System Simulator (BASS)
The four ecosystems considered here are systems having the biological and geochemical properties consistent with a seepage lake, coastal plain river, drainage lake, and stratified lake. Here and below, we refer to these ecosystem types as Ecosystems A-D, respectively, recognizing that responses within each class of freshwater ecosystems can vary substantially as well depending on a variety of biogeochemical attributes. Simulations driven with source attributed atmospheric deposition from GEOS-Chem were initially calibrated to the specific ecological characteristics described by Knightes et al. (2009). The models use empirically constrained, first-order and pseudo-first order rate constants to simulate the rate of methylation (Hg(II) conversion to MeHg), demethylation (MeHg degradation to Hg(II)), oxidation (Hg(0) conversion to Hg(II)), reduction (Hg(II) conversion to Hg(0)), and photo-degradation (MeHg conversion to Hg(0)). The BASS model is used to simulate the trophic dynamics of MeHg beginning with uptake at the base of the food web to the top predator fish species in each ecosystem. For consistency, we use a trophic level 4 fish species to compare the temporal responses across all ecosystems. Further details of algorithms used to describe mercury speciation, transport, and bioaccumulation in these models as well as model evaluation are described by Knightes (2008), Knightes et al. (2009) and Barber (2003).

For each ecosystem, we apply the deposition scenarios typical of the Northeast and Southeast U.S. detailed in Table 1. The model is initialized for all ecosystems with empirically-constrained concentrations (Knightes et al. 2009), and run with total present-day deposition from GEOS-Chem. For the watershed-dominated Ecosystem B, an initialization period (50 years) is needed due to slower response time (Knightes et al. 2009). Because geochemical characteristics affecting mercury speciation have changed over time, we cannot simulate the true pre-industrial
state of these ecosystems. Instead, we simulate the fate of combined historical anthropogenic and
natural components of deposition, which comprise all mercury prior to the present day regardless
of source. The change in this fraction over time is determined by the difference between a
simulation with total deposition and those with direct anthropogenic sources.

Figure 1 shows the temporal evolution of source attributions of MeHg in predatory fish
for the four model ecosystems for both the Northeast and Southeast deposition scenarios. Our
results show the fraction of fish MeHg attributable to North American anthropogenic sources
varies considerably both among systems and between the two deposition scenarios after 40 years
of constant atmospheric loading. In the model ecosystems, initial empirically-constrained
concentrations are not at steady state with respect to deposition inputs. In cases where
concentrations are increasing (decreasing) this suggests that historical loadings to our
hypothetical ecosystems were less than (greater than) simulated deposition. Recent analysis
suggests that deposition in the Northeast U.S. has been declining, while Southeast deposition
shows no trend (Butler et al. 2008).

The faster the aquatic system responds, the more rapidly fish methlymercury reflects the
attribution of deposition. Differences in response times are due to ecosystem-specific factors
such as evasion rates, sediment burial rates, and active sediment layer depths (Knightes et al.
2009). While North American sources contribute over half of deposition to all of the ecosystem
types for the Northeast deposition scenario, their contribution at year 40 to fish MeHg ranges
from 40% (Ecosystem B) to 60% (Ecosystem C). In the Southeast deposition scenario, the
contribution from North American sources ranges from 7-11% after 40 years. International
sources make up a larger fraction of MeHg in the Southeast than the Northeast, reflecting their
greater contribution to deposition.
The change in historical+natural loading over time is more complex. In some systems, such as Ecosystem A in the Southeast, the historical+natural contribution increases over time, while in others, such as Ecosystem D, it decreases. Decreases in the relative historical contribution reflect the rapid turnover of mercury deposited in the few years prior to $t=0$, while increases reflect both the influence of watersheds (Ecosystem B) and the increases of initial concentrations to reflect a steady-state relative to the deposition from historical sources (Ecosystem A). In all systems, in the very long term, source attributions in fish will approach those in deposition. These timescales, however, may approach the timescales in which changes in mercury loading affect emissions from land and ocean reservoirs ($\sim$100 years), which are at present increasing in response to anthropogenic loadings; this would increase the magnitude of deposition from historical sources.

As an illustrative example of the impacts of changes in freshwater fish mercury on human exposure, we assess the range of potential benefits of decreases in emissions for hypothetical consumers of fish from these ecosystems, using the variation among source contributions to regions and lakes shown in Figure 3. We apply these source contributions to the lower bound mean adult Native American fish intake in the survey of Moya (2004) (0.7 g kg$^{-1}$ bw d$^{-1}$).

Figure 2 shows the MeHg intake ($\mu$g kg$^{-1}$ bw d$^{-1}$) of a hypothetical consumer eating trophic level 4 fish from each of the four ecosystems shown in Figure 1 at year 40, for the two deposition scenarios. The upper panel shows a consumer eating 0.7 g fish kg$^{-1}$ bw d$^{-1}$ and the lower 3.3 g fish kg$^{-1}$ bw d$^{-1}$. The solid line represents the World Health Organization (WHO) maximum intake criteria of 0.47 $\mu$g Hg kg$^{-1}$ bw d$^{-1}$, and the dashed line the U.S. EPA reference dose for MeHg of 0.1 $\mu$g Hg kg$^{-1}$ bw d$^{-1}$. Contributions to the consumer’s intake from North American and international deposition are shown in red and purple, respectively, and
historical+natural mercury is shown in orange. Thus, the red represents the fraction of intake that can be affected by present and future domestic policy alone; the purple by international policies, and the orange intake that cannot be altered by mercury-specific emission control policies.

As is shown in Figure 2, the potential effectiveness of North American regulations alone varies substantially across the different ecosystems and the two deposition scenarios. MeHg intake exceeds WHO guidelines for five of the eight ecosystems in the lower consumption scenario and all ecosystems in the high consumption scenario. The EPA RfD is exceeded for all ecosystems in both scenarios. Reductions in North American deposition will result in significant decreases in MeHg intake, especially in the Northeast and in faster-responding ecosystems like the model Ecosystem A. International emissions controls, however, are as important as North American controls in the Southeast.

However, North American reductions alone will in most cases not result in decreases below guideline levels. Especially in the Southeast, our results suggest that historical emissions will continue in the long term to contribute MeHg in excess of exposure guidelines. The only long-term sink for mercury in the global biogeochemical cycle is burial in the sediments. It is important to note here that historical mercury continues to be emitted to the global atmosphere from surface reservoirs that have been enriched over time, and thus historical mercury will continue to deposit to these ecosystems until the global reservoirs return to their steady-state levels (on timescales that can range from centuries to millennia).

**Temporal trends in oceanic mercury and exposure from marine fish**

Although mercury exposure from freshwater ecosystems is important for some of the most highly exposed groups (e.g., recreational and subsistence fishers), for the average individual in the U.S. the majority of fish consumed comes from the commercial market.
Previous studies have shown that >90% of the edible species sold in the commercial market are from marine and estuarine systems (Carrington et al. 2004; Sunderland 2007). Thus, on the population-wide level, dietary intake of MeHg from estuarine and marine seafood accounts for most exposure. To analyze trends in exposure from marine and estuarine fish and shellfish for high-risk groups, we need information that links atmospheric deposition, concentrations of MeHg in coastal and open ocean environments, marine fish mercury levels and local scale consumption data. Understanding mercury dynamics in open-ocean environments is especially important because pelagic marine species such as tuna and swordfish cumulatively account for more than half of the population wide mercury intake in the U.S. (Sunderland 2007).

Presently there is no modeling framework that links atmospheric fate and transport of inorganic mercury to MeHg concentrations in the oceans and subsequent bioaccumulation in marine fish. This represents a major gap in our capability to analyze the effects of emissions reductions on human exposures from marine systems. However, existing models allow us to link source-attributed deposition to total mercury concentrations in seawater as a first step toward such an analysis.

The Sunderland and Mason (2007) model uses a simplified physical framework for ocean circulation that approximately matches the limited availability of oceanic mercury data that can be used to constrain the simulation of transport and accumulation of mercury in different ocean basins. The model is driven by atmospheric deposition but also includes mercury transport associated with lateral and vertical circulation and settling particulate matter, evasion of Hg(0) in the surface ocean, and freshwater discharges. In this study, we focus on water column depths that are most relevant for MeHg production, transport and biological exposures.
To assess the contributions of different emission sources to ocean concentrations, we drive the ocean model with source-attributed present-day deposition. The evolution over time of concentrations in ocean basins from direct North American and international anthropogenic emissions is diagnosed by running the model with deposition specified by GEOS-Chem with those sources shut off. We calculate the natural component by driving the ocean model with pre-industrial deposition from GEOS-Chem. The historical component is determined by the difference between the total concentration and the sum of the natural and direct anthropogenic component.

Figure 3 shows the evolution over time of concentrations in eight surface and intermediate ocean basins attributed to various sources. Present and future emissions begin to accumulate in each ocean basin at time $t=0$. Over time, the mercury concentration in surface ocean boxes approaches steady state. At steady state, source attributions are equal to their contributions to deposition. Present and future emission sources are divided between North American (shown in red) and international anthropogenic contributions (shown in purple).

As shown in Figure 3, present and future contributions to ocean concentrations after 40 years are still lower than atmospheric deposition enrichment levels (Table 1). Total mercury concentrations in the top 300-1500 m of the water column simulated here require decades to centuries to reach steady state with contemporary atmospheric deposition. The fraction of seawater mercury attributed to 40 years of present-day deposition ranges from 14-23%. The highest values are in the Pacific Ocean at mid-latitudes (below 30°N), and the lowest in the Southern Ocean. Historical contributions after 40 years (shown in orange in Figure 1) range from a low of 26% in the Southern Ocean to a high of 42% in the Mediterranean Sea. The growth of the historical contribution to concentrations occurs in part because historical mercury is a
continuing fraction of mercury deposition into the future, due to its continued cycling in the
environment. MeHg dynamics in oceans may vary substantially from total mercury because
water column methylation processes appear to occur primarily in the low-oxygen regions below
the well mixed surface layer of the ocean (Mason and Fitzgerald 1990; Mason et al. 1998; Mason
and Sullivan 1999; Sunderland et al. 2009). The temporal response of total mercury
concentrations in the surface ocean will be relatively rapid (several years) compared to those of
intermediate (decades to centuries) and deep waters (many centuries) (Sunderland 2007;
Sunderland et al. 2009)

At present, gaps in present understanding of the marine mercury exposure pathway limit
our ability to assess the potential source contributions to U.S. commercial market mercury
exposure. As a first-order approximation relating oceanic mercury concentrations to potential
exposures from pelagic marine fish, we can assume that over long timescales changes in
inorganic mercury concentrations will be reflected by differences in MeHg and subsequent
bioaccumulation in biota. Although this represents an improvement over previous exposure
studies that have not considered how ocean circulation and other loss processes affect mercury
accumulation, such an assumption is still highly uncertain for the reasons detailed above.
However, we can postulate based on our results that given the relatively limited fraction of total
mercury in open ocean environments from North American sources over the next 40 years
(Figure 3) and the large fraction of population-wide exposure from marine species sold in the
commercial market (13), domestic emission reductions alone are not likely to substantially
reduce marine fish mercury levels. This analysis suggests that mercury exposure to consumers
from commercial market fish may continue to rise over the next several decades. This is because
mercury levels in much of the ocean have not yet reached steady-state with contemporary
atmospheric deposition. Historically-emitted mercury continues to cycle through the land-
atmosphere system and affect the atmosphere, and thus deposition. In addition, the large
contribution from sources outside North America means that domestic regulatory action may not
substantially affect mercury concentrations in pelagic marine species such as tuna and swordfish.
Furthermore, global mercury sources are likely to increase, especially in Asia (50% of present-
day global emissions) as growing economies use more coal-based energy. We therefore posit that
substantial global reductions in emissions (near-elimination of releases) are needed to prevent
further increases in concentrations in marine organisms and associated human exposure.

4. Discussion

Our results show that the potential impact of domestic and international emissions
reductions on mercury exposure varies considerably for different populations of consumers.
Domestic emissions controls have the potential to substantially reduce exposure of freshwater
fish consumers in the Northeast U.S. This is true especially for those who consume fish from
rapidly responding lakes dominated by direct atmospheric deposition to the water surface and
with low watershed to water surface area ratios. In the Southeast U.S., atmospheric deposition
from international sources is as important as domestic mercury sources. Thus, for freshwater
fish consumers in this region, international emission reductions are likely very important for
reducing fish mercury burdens and human exposure. International sources are also likely to
dominate future trends in mercury exposure from fish sold in the U.S. commercial market. Our
analysis highlights that historical mercury will have a continuing, long-term impact on exposure
regardless of mercury emissions. Thus, reducing present-day emissions has the two-fold benefit
of reducing future exposures from direct and re-emitted mercury in the environment.
The potential for international emissions reductions to reduce exposure, as presented here, should be viewed as a lower bound, since emissions under a “business-as-usual” scenario are likely to increase (particularly in Asia) rather than stay constant. Our results indicate that international emissions reductions can dramatically slow the pace of future exposure increases from ocean fish. However, past emissions may have already committed the ocean system to a rate of increase that cannot be addressed by present-day emissions reductions alone, and to continuing deposition to local ecosystems. As mentioned above, a substantial long-term benefit of global-scale anthropogenic emissions reductions, which is not directly considered in our analysis, is that continuing emissions add to the pool of historical mercury, which cycles over very long timescales in the environment and can further contribute to exposure; this increase would be avoided with emissions reductions.

Minimizing human exposures to mercury will require both mitigation and adaptation actions because of the long-term impacts of historical mercury illustrated here. In this way, the mercury cycle’s effect on continuing exposure is similar to the carbon cycle influence on climate change. Due to carbon cycle timescales, anthropogenic warming and sea level rise would continue for centuries even if greenhouse gas concentrations stabilized (Intergovernmental Panel on Climate Change 2007). In response to this, policy actions have begun to focus both on mitigation and adaptation (Pielke 1998). In the case of mercury, policy makers have already initiated some adaptation measures, such as dietary advice for pregnant women and children on ways to limit their mercury intake (U.S. Department of Health and Human Services and U.S. Environmental Protection Agency 2004). The long-term nature of the mercury problem suggests that a variety of approaches, at different scales, is necessary (Selin and Selin 2006).

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Protection Agency's peer and administrative review policies and approved for publication.

Mention of trade names or commercial products does not constitute endorsement or recommendation for use.
References


### Table 1: Source-attributed total deposition (µg m\(^{-2}\) y\(^{-1}\)) and percentage source contribution to deposition for ocean basins and U.S. regions. GEOS-Chem deposition results are from Selin and Jacob (2008).

<table>
<thead>
<tr>
<th>Region</th>
<th>Total</th>
<th>Natural</th>
<th>U.S.</th>
<th>International</th>
<th>Historical</th>
</tr>
</thead>
<tbody>
<tr>
<td>Northeast US (40-44° N, 77.5-82.5° W)</td>
<td>24.2</td>
<td>3.97 (16%)</td>
<td>14.3 (59%)</td>
<td>2.09 (9%)</td>
<td>3.87 (16%)</td>
</tr>
<tr>
<td>Southeast US (24-28° N, 77.5-82.5° W)</td>
<td>34.1</td>
<td>14.3 (42%)</td>
<td>3.69 (11%)</td>
<td>7.94 (23%)</td>
<td>8.19 (24%)</td>
</tr>
<tr>
<td>North Atlantic (&gt;55° N)</td>
<td>9.73</td>
<td>3.37 (35%)</td>
<td>0.71 (7%)</td>
<td>2.71 (28%)</td>
<td>2.95 (30%)</td>
</tr>
<tr>
<td>Antarctic (&gt;65° S)</td>
<td>1.39</td>
<td>0.41 (29%)</td>
<td>0.09 (6%)</td>
<td>0.24 (17%)</td>
<td>0.65 (47%)</td>
</tr>
<tr>
<td>Surface Pacific/Indian (40° S - 30° N)</td>
<td>19.7</td>
<td>6.16 (31%)</td>
<td>0.87 (4%)</td>
<td>3.97 (20%)</td>
<td>8.74 (44%)</td>
</tr>
<tr>
<td>North Pacific (&gt;30° N)</td>
<td>15.4</td>
<td>3.81 (25%)</td>
<td>0.73 (5%)</td>
<td>3.20 (21%)</td>
<td>7.63 (50%)</td>
</tr>
<tr>
<td>Surface Atlantic (35° S - 55° N)</td>
<td>19.9</td>
<td>6.68 (34%)</td>
<td>1.10 (6%)</td>
<td>4.18 (21%)</td>
<td>7.92 (40%)</td>
</tr>
<tr>
<td>Mediterranean</td>
<td>27.1</td>
<td>8.05 (30%)</td>
<td>1.12 (4%)</td>
<td>5.71 (21%)</td>
<td>12.2 (45%)</td>
</tr>
</tbody>
</table>
**Figure Legends**

**Figure 1:** Temporal evolution of fish MeHg source attributions for various model lake ecosystems to deposition scenarios for the Northeast (40-44 N, 77.5-82.5 W) and Southeast (28-32N, 77.5-82.5 W) regions of the United States, as specified by the GEOS-Chem model (Selin and Jacob 2008; Selin et al. 2008) and the ecosystem models of Knightes *et al.* Ecosystems A-D refer to the seepage lake, coastal plain river, drainage lake, and stratified lake respectively from *Knightes et al.* Note difference in scale for Ecosystems C and D. The model is run for 50 years; the first 10 years are treated as initialization.

**Figure 2:** Methylmercury intake source attribution for a hypothetical subsistence fisher eating 0.7 g kg$^{-1}$ bw d$^{-1}$ for the four model ecosystems under Northeast and Southeast deposition scenarios shown in Figure 3. Solid line indicates WHO maximum intake recommendation; dashed line shows U.S. EPA RfD.

**Figure 3:** Evolution of relative contributions of anthropogenic (North American and international), historical, and natural sources to mercury concentrations in six surface ocean basins.
The graphs illustrate the deposition of fish mercury (ppm) in different ecosystems labeled A, B, C, and D over a period of 40 years, categorized into "Southeast" and "Northeast" Deposition. Four primary types of deposition are depicted:

- **North American anthropogenic**
- **International anthropogenic**
- **Historical+Natural**

The graphs show a consistent trend of increasing mercury deposition over the years in all ecosystems and categories, indicating the impact of anthropogenic and natural factors.