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

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2   **policy**

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List of abbreviations and definitions:

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BASS	Bioaccumulation and Aquatic System Simulator
Hg(0)	elemental mercury
Hg(II)	oxidized mercury
Hg(P)	particulate mercury
MeHg	Methylmercury
RfD	Reference Dose
SERAFM	Spreadsheet-based Ecological Risk Assessment for the Fate of Mercury model
U.S. EPA	United States Environmental Protection Agency
WASP	Water Quality Analysis Simulation Program
WCS-MLM	Watershed Characterization System Mercury Loading Model
WHO	World Health Organization

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54 **Abstract**

Background: Since the 1970s, policies attempting to reduce adverse effects of environmental

56 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

58 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

60 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

62 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

64 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

66 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

68 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

70 across various populations.

1. Introduction

72   Mercury is a global pollutant that is ubiquitous in the environment. Once deposited to ecosystems, inorganic mercury may be converted to methylmercury (MeHg), which

74   bioaccumulates in food webs. MeHg exposure causes severe human health effects including immune system suppression, neurodevelopmental delays in children, and compromised

76   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

78   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,

80   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).

82   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.

84   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

86   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

88   analysis to assess the expected impacts of current policies and to identify gaps in present understanding of the mercury exposure pathway.

90   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

92   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

94   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

96   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

98   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

100   and imported species (Carrington et al. 2004; Sunderland 2007). This means that the

relationships among anthropogenic, historical and natural mercury sources, MeHg levels in

102   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

104   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

106   different mercury control strategies. Environmental modeling combines disparate atmospheric,

aquatic and human health data with our best understanding of underlying processes to help assess

108   the future effects of possible policies and regulatory decisions (National Research Council 2007).

Previous studies have analyzed potential benefits from domestic mercury emissions

110   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

112   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

114   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

116   the U.S. by assuming a linear relationship between declines in atmospheric emissions and human

exposure levels.

118   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

120   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

122   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

124   chemistry transport model (GEOS-Chem). We explore the timescales required for each

component of atmospheric deposition to cycle through ecosystems by simulating mercury

126   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.

128   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

130   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

132   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

134   may be affected by changes in ecological concentrations resulting from emission controls.

        We attribute exposure to natural background, present-day anthropogenic emission

136   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*

138   *et al.* (2008). The natural component represents the pre-industrial steady state of roughly one

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third of present global emission and deposition. Direct anthropogenic emissions from North

140   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

142   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

144   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

146   American and international anthropogenic sources) and the timescales of ecosystem response to

historical contamination, which is not directly addressed by contemporary emissions reduction

148   policies.

2. Methods

150   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

152   *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

154   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

156   can transport globally. Oxidized and particulate mercury (Hg(II) and Hg(P)) are shorter-lived

and deposit on a local to regional scale.

158   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

160   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

162 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

164 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

166 *Selin et al* (Selin et al. 2008).

   GEOS-Chem simulates wet and dry deposition of Hg(II) and Hg(P) as well as dry

168 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

170 influencing aquatic ecosystems. Globally, the source attribution of deposition reflects the

attributions of emissions; however, these attributions vary spatially, as anthropogenic emissions

172 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

174 their relative areas (Selin 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

176 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

178 here on meteorological data for 2004-2005.

   Table 1 shows GEOS-Chem deposition and source attribution for two U.S. deposition

180 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

182 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

184 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

186 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

188 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

190 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-

192 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

194 2003). The remainder comes from international anthropogenic sources (23%), natural sources

(42%), and historical mercury (24%) (Selin and Jacob 2008).

196 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

198 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

200 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

202 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

204 consistent with our results reported here.

Across all ocean basins, the mean percentage of deposition attributed to present-day

206 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

208   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

210   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

212   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

214   desert regions, and thus predicts higher than expected deposition based on the observational data

(Kotnik et al. 2007; Zagar et al. 2007).

216   *Sunderland and Mason* (2007) compared atmospheric deposition estimates from three

models and the influence of such variability on ocean mercury concentrations over the next

218   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

220   increases in concentrations over the next several decades compared to decreases in

concentrations based on other atmospheric models. Resolving this uncertainty requires additional

222   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

224   uncertain, as it reflects the global attributions of emissions sources.

For the analysis presented here, we drive the aquatic mercury fate and transport models

226   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

228   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

230   when interpreting results for future exposure pathways. The approach presented here may

underestimate potential contributions from the Asian continent (which are increasing) and

232   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

234   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

236   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

238   and future mercury emissions and trends in environmental concentrations become available

(Mason et al. 2005; Streets and Zhang 2008).

240   **3. Results**

Temporal trends in freshwater fish mercury and associated exposure

242   We combine source-attributed deposition from GEOS-Chem with previously published

watershed, water body, and food web bioaccumulation models for mercury applied to four

244   different types of freshwater ecosystems across the contiguous U.S. (Knightes et al. 2009). Each

of these modeling frameworks is publicly available

246   (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

248   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)

250   (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

252   (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)

254   model (Barber 2003; Barber 2006).

        The four ecosystems considered here are systems having the biological and geochemical

256   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

258   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

260   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-

262   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)),

264   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

266   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

268   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.*

270   (2009) and *Barber* (2003).

        For each ecosystem, we apply the deposition scenarios typical of the Northeast and

272   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

274   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

276   affecting mercury speciation have changed over time, we cannot simulate the true pre-industrial

13

state of these ecosystems. Instead, we simulate the fate of combined historical anthropogenic and

278   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

280   simulation with total deposition and those with direct anthropogenic sources.

Figure 1 shows the temporal evolution of source attributions of MeHg in predatory fish

282   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

284   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

286   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

288   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

290   shows no trend (Butler et al. 2008).

The faster the aquatic system responds, the more rapidly fish methlymercury reflects the

292   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.

294   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

296   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

298   sources make up a larger fraction of MeHg in the Southeast than the Northeast, reflecting their

greater contribution to deposition.

300   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,

302   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

304   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

306   (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

308   in mercury loading affect emissions from land and ocean reservoirs (~100 years), which are at

present increasing in response to anthropogenic loadings; this would increase the magnitude of

310   deposition from historical sources.

As an illustrative example of the impacts of changes in freshwater fish mercury on human

312   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

314   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}).

316   Figure 2 shows the MeHg intake (μg 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

318   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)

320   maximum intake criteria of 0.47 μg Hg kg^{-1} bw d^{-1}, and the dashed line the U.S. EPA reference

dose for MeHg of 0.1 μg Hg kg^{-1} bw d^{-1}. Contributions to the consumer's intake from North

322   American and international deposition are shown in red and purple, respectively, and

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historical+natural mercury is shown in orange. Thus, the red represents the fraction of intake that

324   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.

326   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

328   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

330   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

332   the model Ecosystem A. International emissions controls, however, are as important as North

American controls in the Southeast.

334   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

336   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

338   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

340   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).

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

Although mercury exposure from freshwater ecosystems is important for some of the

344   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.

346   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

348   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

350   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

352   consumption data. Understanding mercury dynamics in open-ocean environments is especially

       important because pelagic marine species such as tuna and swordfish cumulatively account for

354   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

356   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

358   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

360   such an analysis.

           The *Sunderland and Mason* (2007) model uses a simplified physical framework for ocean

362   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

364   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

366   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.

368   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

370   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

372   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

374   difference between the total concentration and the sum of the natural and direct anthropogenic

component.

376   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

378   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

380   contributions to deposition. Present and future emission sources are divided between North

American (shown in red) and international anthropogenic contributions (shown in purple).

382   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

384   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

386   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

388   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

390   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

392   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

394   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

396   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;

398   Sunderland et al. 2009)

          At present, gaps in present understanding of the marine mercury exposure pathway limit

400   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

402   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

404   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

406   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

408   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

410   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

412   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

414   atmospheric deposition. Historically-emitted mercury continues to cycle through the land-atmosphere system and affect the atmosphere, and thus deposition. In addition, the large

416   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.

418   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

420   substantial global reductions in emissions (near-elimination of releases) are needed to prevent further increases in concentrations in marine organisms and associated human exposure.

422   **4. Discussion**

    Our results show that the potential impact of domestic and international emissions

424   reductions on mercury exposure varies considerably for different populations of consumers. Domestic emissions controls have the potential to substantially reduce exposure of freshwater

426   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

428   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

430   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

432   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

434   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.

436   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

438   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

440   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

442   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

444   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

446   would be avoided with emissions reductions.

Minimizing human exposures to mercury will require both mitigation and adaptation

448   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

450   change. Due to carbon cycle timescales, anthropogenic warming and sea level rise would

continue for centuries even if greenhouse gas concentrations stabilized (Intergovernmental Panel

452   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

454   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.

456   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 polices and approved for publication.

460   Mention of trade names or commercial products does not constitute endorsement or

recommendation for use.

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Tables

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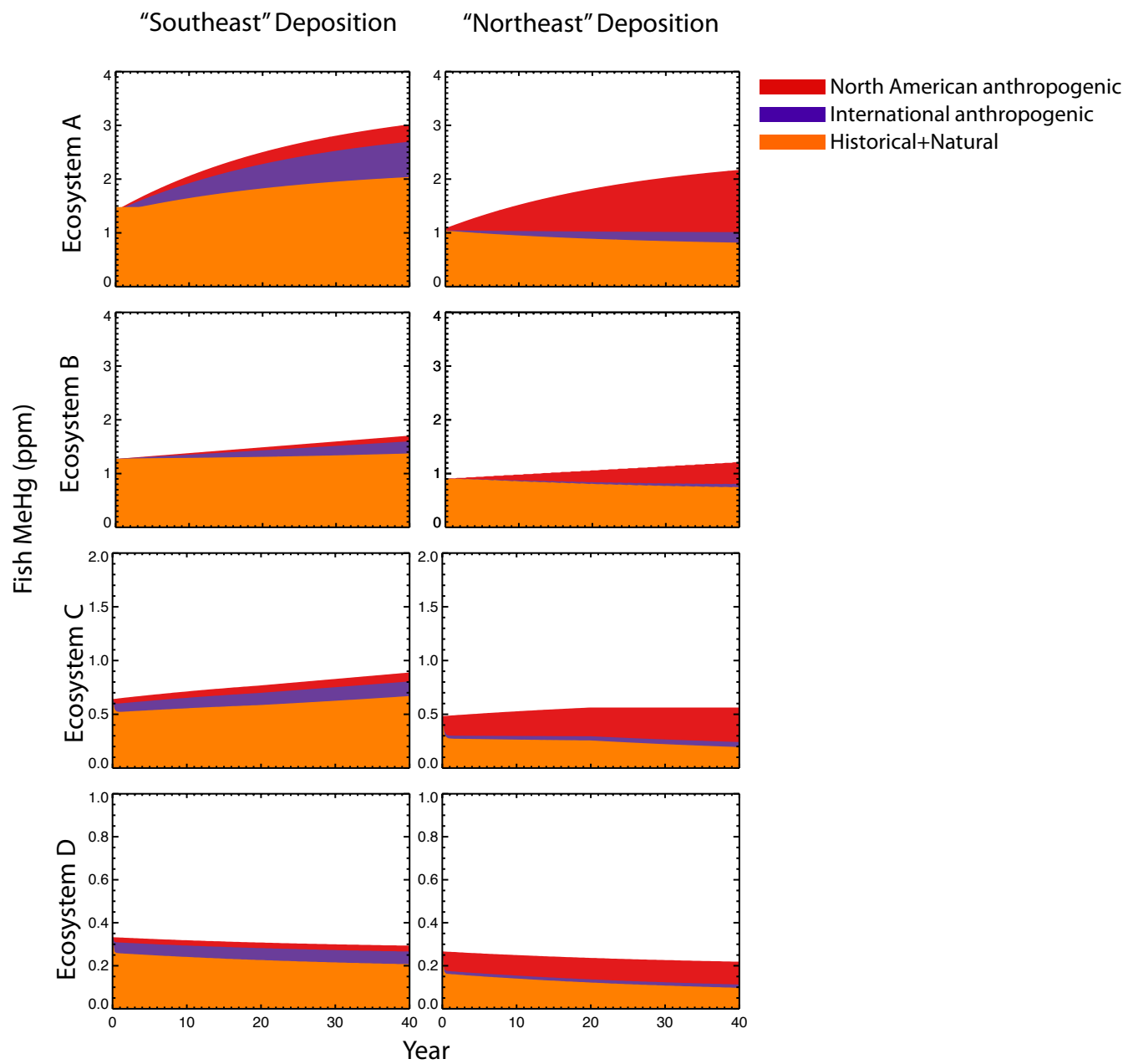
Region	GEOS-Chem				
	Total	*Natural*	*U.S.*	*International*	*Historical*
Northeast US (40-44º N, 77.5-82.5º W)	24.2	3.97 (16%)	14.3 (59%)	2.09 (9%)	3.87 (16%)
Southeast US (24-28º N, 77.5-82.5º W)	34.1	14.3 (42%)	3.69 (11%)	7.94 (23%)	8.19 (24%)
North Atlantic (>55º N)	9.73	3.37 (35%)	0.71 (7%)	2.71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

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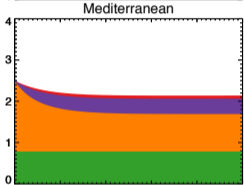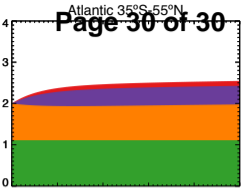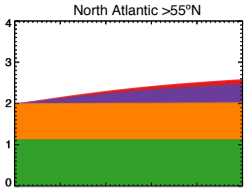
Figure 2: Methylmercury intake source attribution for a hypothetical subsistence fisher eating 0.7 g kg^{-1} bw d^{-1} for the four model ecosystems under Northeast and Southeast deposition scenarios shown in Figure 3. Solid line indicates WHO maximum intake recommendation; dashed line shows U.S. EPA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