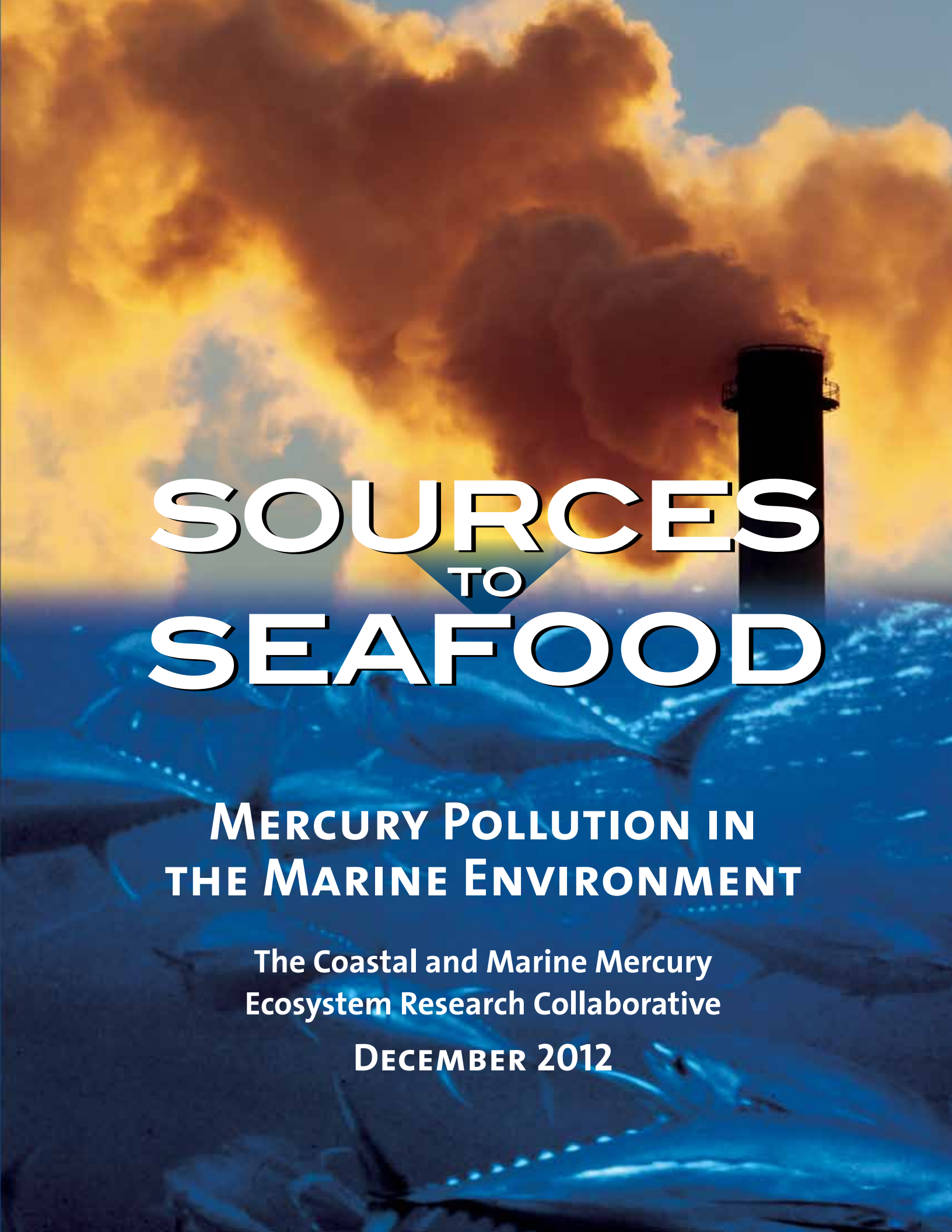


SOURCES TO SEAFOOD

Mercury Pollution in the Marine Environment

The Coastal and Marine Mercury Ecosystem Research Collaborative

December 2012

About the report

In 2010, the Toxic Metals Superfund Research Program at Dartmouth College brought together a group of 50 scientists and policy stakeholders to form C-MERC, the Coastal and Marine Mercury Ecosystem Research Collaborative. The goal was to review current knowledge—and knowledge gaps—relating to a global environmental health problem, mercury contamination of the world's marine fish. C-MERC participants attended two workshops over a two-year period, and in 2012 C-MERC authors published a series of peer-reviewed papers in the journals *Environmental Health Perspectives* and *Environmental Research* that elucidated key processes related to the inputs, cycling, and uptake of mercury in marine ecosystems, effects on human health, and policy implications. This report synthesizes the knowledge from these papers in an effort to summarize the science relevant to policies being considered at regional, national, and global levels.

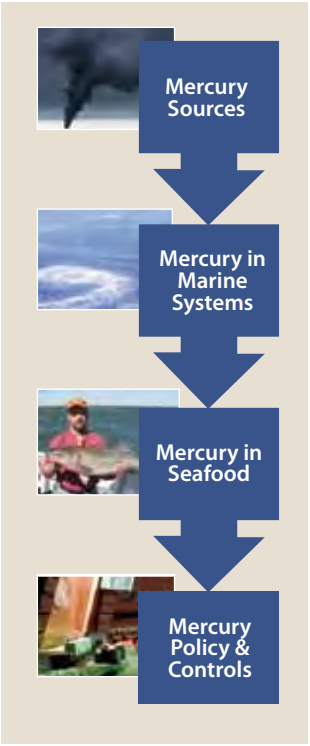
The Dartmouth Toxic Metals Superfund Research Program uses an interdisciplinary approach to investigate the ways that arsenic and mercury in the environment affect ecosystems and human health. Arsenic and mercury are commonly found in Superfund sites around the U.S. as well as other areas that result in exposures to certain communities. The Research Translation Core of the program communicates program science to government partners, non-governmental organizations, health care providers and associations, universities and the lay community, and facilitates the use of its research for the protection of public health. The Research Translation Core organized the C-MERC effort.

The Superfund Research Program of the National Institute of Environmental Health Sciences supports a network of university programs that investigate the complex health and environmental issues associated with contaminants found at the nation's hazardous waste sites. The Program coordinates with the Environmental Protection Agency and the Agency for Toxic Substances and Disease Registry of the Centers for Disease Control and Prevention, federal entities charged with management of environmental and human health hazards associated with toxic substances.

Suggested citation

Chen, C.Y., C.T. Driscoll, K.F. Lambert, R.P. Mason, L.R. Rardin, C.V. Schmitt, N.S. Serrell, and E.M. Sunderland. 2012. Sources to Seafood: Mercury Pollution in the Marine Environment. Hanover, NH: Toxic Metals Superfund Research Program, Dartmouth College.

Executive Summary



Mercury poses substantial threats to human health, and is ranked third on the U.S. Agency for Toxic Substances and Disease Registry's priority list of contaminants that are hazardous to the U.S. population (ATSDR 2011). Mercury pollution in the surface ocean has more than doubled over the past century, leading governments and organizations to take actions to protect humans from the harmful effects of this toxic element. The increase in mercury pollution comes from past and present human activities such as coal burning, mining, and industrial processes. Mercury released into the environment by these activities contaminates food webs in oceans and coastal ecosystems, accumulating to levels of concern in fish consumed by humans. More than 90 percent of methylmercury exposure from fish consumption in the U.S. and in many regions of the world comes from estuarine and marine fish.

The Coastal and Marine Mercury Ecosystem Research Collaborative (C-MERC) brought together 50 scientists and policy experts to analyze and synthesize the current science on mercury pollution in the marine environment from mercury sources to methylmercury in seafood. In 2012, C-MERC authors published a series of 11 peer-reviewed papers in the journals *Environmental Health Perspectives* (Chen 2012) and *Environmental Research* (Chen et al. 2012). In this report we synthesize information from the C-MERC manuscripts which include data from six marine systems (*Figure 1*), data from three additional coastal basins, and the scientific literature.

Major Findings

Four major findings emerge from the C-MERC synthesis. These statements represent a consensus of the coauthors of this report based on a review of existing scientific information.

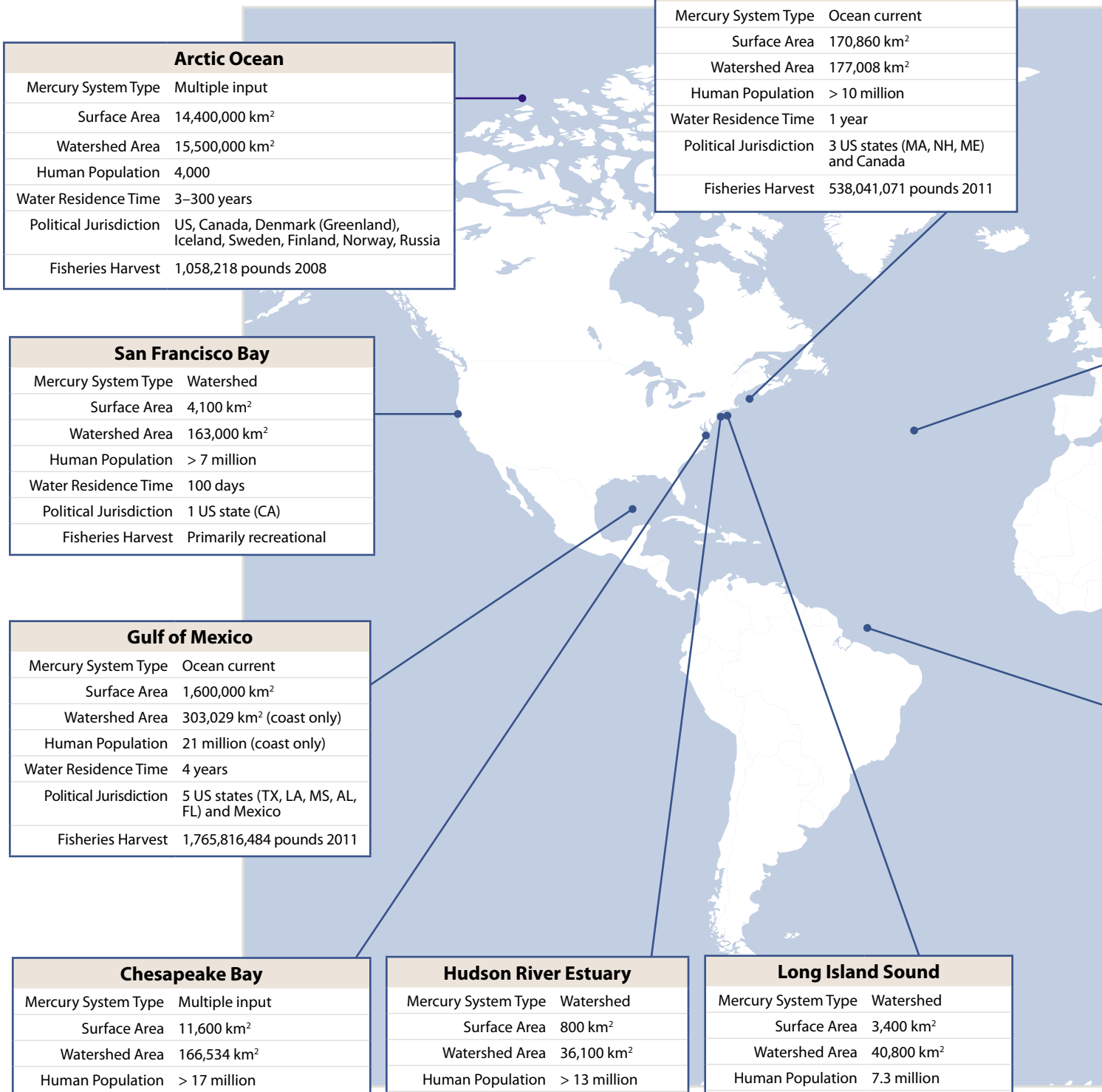
1. Mercury pollution is ubiquitous in the world's oceans and coastal waters. It contaminates fish and other seafoods that are important sources of protein and nutrition for people worldwide. Despite improvements in some regions, methylmercury in commonly consumed marine fish continues to exceed human health guidelines, and mercury pollution is on the rise.

2. Mercury pollution enters the marine environment along distinct pathways that are linked to different mercury sources. Atmospheric inputs from global sources of mercury emissions dominate the "open ocean" and "ocean current" systems. Riverine mercury inputs dominate coastal waters that are "watershed systems." Some coastal waters are "multiple input" systems that reflect both atmospheric and riverine inputs.

3. Most seafood consumers are "general consumers" whose methylmercury intake comes from fish typically harvested from the open oceans which receive atmospheric inputs from global mercury emission sources. Methylmercury intake by "local consumers" comes from seafood caught from nearby coastal waters that receive riverine inputs from local, regional, and global sources.

4. Methylmercury concentrations in marine fish will decline roughly in proportion to decreases in mercury inputs, though the timing of the response will vary. Methylmercury in open ocean fish will begin to decrease within several years to decades after emissions controls. In contrast, methylmercury in fish from coastal systems may exhibit a range of response times over many decades to centuries, depending on the relative importance of atmospheric to other inputs.

Figure 1. C-MERC Study Regions



Arctic Ocean

Mercury System Type	Multiple input
Surface Area	14,400,000 km^2
Watershed Area	15,500,000 km^2
Human Population	4,000
Water Residence Time	3–300 years
Political Jurisdiction	US, Canada, Denmark (Greenland), Iceland, Sweden, Finland, Norway, Russia
Fisheries Harvest	1,058,218 pounds 2008

Gulf of Maine

Mercury System Type	Ocean current
Surface Area	170,860 km^2
Watershed Area	177,008 km^2
Human Population	> 10 million
Water Residence Time	1 year
Political Jurisdiction	3 US states (MA, NH, ME) and Canada
Fisheries Harvest	538,041,071 pounds 2011

San Francisco Bay

Mercury System Type	Watershed
Surface Area	4,100 km^2
Watershed Area	163,000 km^2
Human Population	> 7 million
Water Residence Time	100 days
Political Jurisdiction	1 US state (CA)
Fisheries Harvest	Primarily recreational

Gulf of Mexico

Mercury System Type	Ocean current
Surface Area	1,600,000 km^2
Watershed Area	303,029 km^2 (coast only)
Human Population	21 million (coast only)
Water Residence Time	4 years
Political Jurisdiction	5 US states (TX, LA, MS, AL, FL) and Mexico
Fisheries Harvest	1,765,816,484 pounds 2011

Chesapeake Bay

Mercury System Type	Multiple input
Surface Area	11,600 km^2
Watershed Area	166,534 km^2
Human Population	> 17 million
Water Residence Time	< 1 year
Political Jurisdiction	Six US states (DE, MD, NY, PA, VA, WV) and District of Columbia
Fisheries Harvest	572,224,898 pounds 2011

Hudson River Estuary

Mercury System Type	Watershed
Surface Area	800 km^2
Watershed Area	36,100 km^2
Human Population	> 13 million
Water Residence Time	4 months
Political Jurisdiction	Five US states (CT, MA, NJ, NY, VT)
Fisheries Harvest	Primarily recreational

Long Island Sound

Mercury System Type	Watershed
Surface Area	3,400 km^2
Watershed Area	40,800 km^2
Human Population	7.3 million
Water Residence Time	2-6 months
Political Jurisdiction	Two US states (CT, NY)
Fisheries Harvest	34,240,002 pounds 2011 (total for CT and NY)

Open Oceans	
Mercury System Type	Open ocean
Surface Area	361,000,000 km^2
Watershed Area	NA
Human Population	NA
Water Residence Time	500-1,000 years
Political Jurisdiction	All nations with coastlines
Fisheries Harvest	170,009,118,540 pounds 2009

Tropical Coastal Waters	
Mercury System Type	NA
Surface Area	Coastal and open ocean waters around the equator, 23.5 N and 23.5 S.
Watershed Area	NA
Human Population	>2.5 billion
Water Residence Time	Variable
Political Jurisdiction	Mexico, Central America, Caribbean, much of South America, Africa, and Asia, Australia, and Pacific Island nations
Fisheries Harvest	Primarily local consumption or aquaculture

Figure 1: Summary statistics for C-MERC study regions—Gulf of Maine (Thompson 2010), Gulf of Mexico (National Ocean Service 2011), Arctic (Raymond et al. 2007, AMAP 2009), San Francisco Bay (van Geen and Luoma 1999, US EPA 2012), Tropics, Chesapeake Bay (Chesapeake Bay Program), Hudson River (Howarth et al. 2000, Pace et al. 1992), and Long Island Sound (Turekian et al. 1996). Commercial fisheries landings based on FAO 2011 or NMFS 2012.

Policy Implications

Four major policy implications emerge from the C-MERC synthesis. These statements represent a consensus of the coauthors of this report based on a review of existing scientific information.

1. Given that most seafood consumers are general consumers, controlling sources of atmospheric mercury emissions will have substantial benefits for the largest fisheries that supply seafood to the most people.

2. Controlling direct discharges and managing legacy sources of mercury can have a substantial impact on coastal fisheries that supply seafood to local populations of recreational and subsistence consumers.

3. Expanded marine monitoring of mercury in air, sediments, water, wildlife, and fish is needed to evaluate the effectiveness of national and international policies. Monitoring could also provide insight on the effects of global environmental change on mercury pollution in marine systems.

4. Improved fish consumption advice could be beneficial, particularly for highly contaminated systems with large populations of local consumers. However, fish consumption advisories often have mixed results and are not a viable substitute for source controls.

Figure 2. **Mercury in Fish and Marine Mammals**

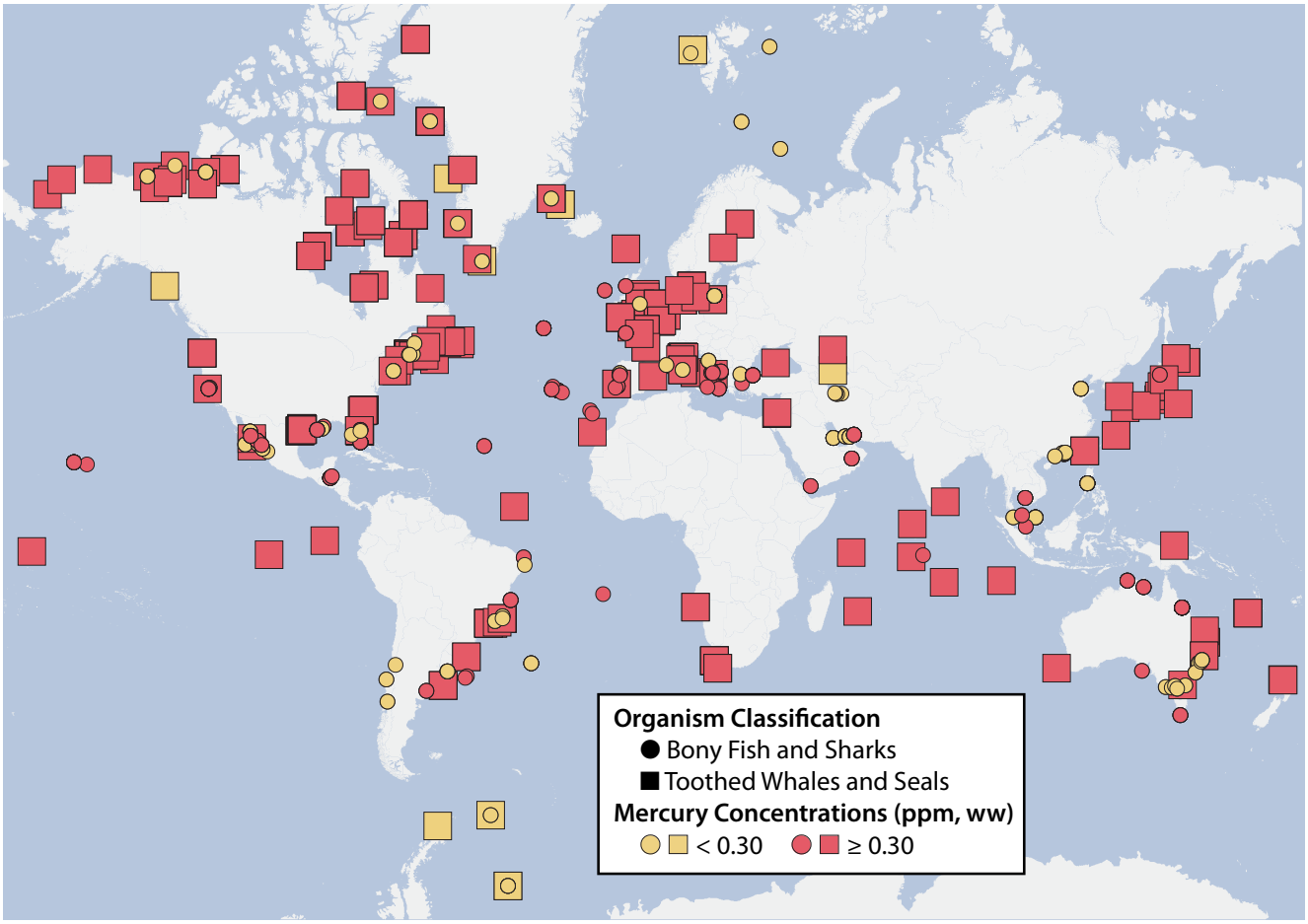


Figure 2: The global distribution of average mercury concentrations (ppm, wet weight) in sharks and rays, bony fish, seals, and toothed whales from muscle tissue. Most samples exceed 0.3 ppm, the U.S. EPA human health criterion. Map from Biodiversity Research Institute, Gorham, ME, based on data summarized from published literature.

I. Why is mercury pollution in the marine environment a concern?

Mercury pollution is ubiquitous in the world's oceans and coastal waters. It contaminates fish and other seafoods that are important sources of protein and nutrition for people worldwide. Despite improvements in some regions, methylmercury in commonly consumed marine fish continues to exceed human health guidelines, and mercury pollution is on the rise.

Mercury, particularly in the chemical form methylmercury, is a toxic pollutant that can adversely affect the health of people. Mercury concentrations in the surface ocean (upper 100 m) have increased four-fold over the past 500 years, with a two-fold increase over the last century concurrent with increasing industrialization and energy production (Streets et al. 2011, Mason et al. 2012). Fish consumption is the main source of methylmercury exposure for people worldwide (Sunderland 2007), and marine fish constitute 92% of the global fish harvest for human consumption (Carrington et al. 2004). Methylmercury concentrations in commonly consumed marine fish, such as tuna, mackerel, and swordfish, exceed the U.S. Environmental Protection Agency's human health criterion of 0.3 parts per million (ppm) in most marine systems studied (*Figure 2*). Most of the mercury (about 90%) in fish consumed by humans occurs as methylmercury.

Human health risks from methylmercury exposure have been widely documented, and include neurological effects, impaired fetal and infant growth, and possible contributions to cardiovascular disease (Grandjean et al. 2005, Mergler et al. 2007, Karagas et al. 2012). Since the early 1970s, government agencies have lowered the recommended daily intake of mercury, from nearly 100 micrograms to 0.1 micrograms per kilogram of body weight per day, reflecting improved understanding of the harmful effects that even low levels of methylmercury can have on human health (Stein et al. 2002). Since the developing brain is particularly sensitive to methylmercury, women of childbearing age, pregnant and breastfeeding women, developing fetuses, and children under the age of 12 are among the most vulnerable. All people who frequently eat fish high in methylmercury by choice or for subsistence face an elevated risk for adverse effects (Mahaffey et al. 2004). However, consuming lower-methylmercury fish provides important nutritional benefits for early neurodevelopment and cardiovascular health (Mozaffarian and Rimm 2006).

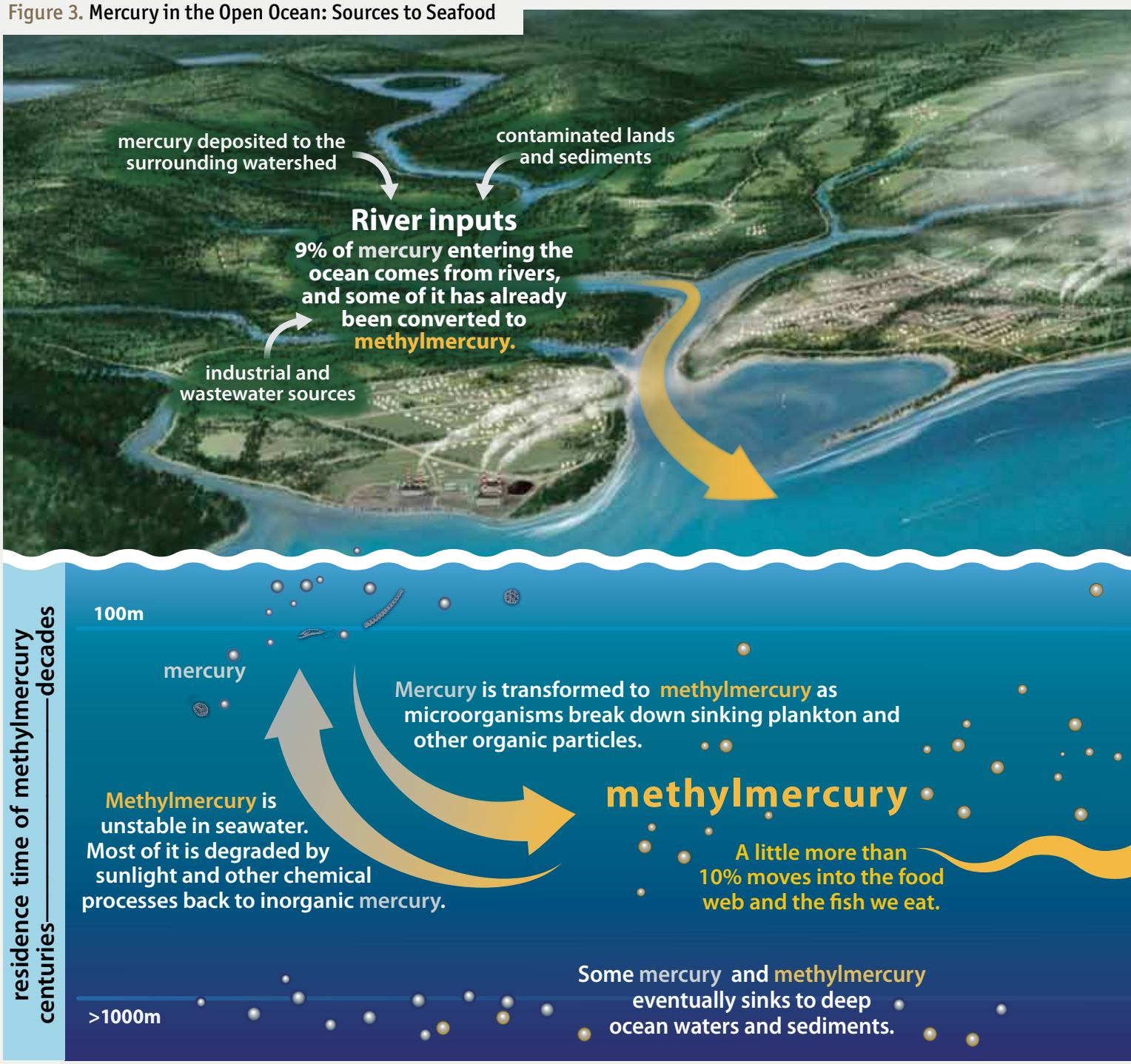
Across the United States, more fish consumption advisories exist for mercury than for any other pollutant (U.S. EPA 2011). Unfortunately, fish consumption advisories aimed at curbing human exposure to methylmercury have had mixed results (Oken et al. 2012) and are not a viable substitute for controlling mercury pollution sources. Without additional policy interventions, global emissions of mercury to the atmosphere are projected to increase by roughly 25% by 2020 from 2005 levels (Pacyna et al. 2010) and could double by 2050 under a business-as-usual scenario (Streets et al. 2009). Fortunately, strategies for decreasing mercury pollution are available, and there is evidence they have been effective. In the United States, atmospheric emissions of mercury have been cut 60% since 1990 by controls in a number of sectors (Schmeltz et al. 2011). These cuts have contributed to regional decreases in atmospheric mercury emissions and deposition as well as mercury concentrations in freshwater fish in those regions (Evers et al. 2011). Limits on direct discharges of mercury to surface waters in some rivers and estuaries also have resulted in decreased mercury in marine animals (see, for example, Sunderland et al. 2012).

While these local and national policies have been effective in mitigating local and regional contamination, mercury transcends political borders and moves with air and water. Addressing the transboundary and multi-media nature of mercury pollution will require global action (Lambert et al. 2012). At the international level, the United Nations Environment Programme has convened 140 countries, including the United States, to negotiate a global legally-binding mercury treaty. To craft effective mercury policy and management decisions that will decrease human exposure to mercury through seafood consumption, it is critical that policies are based on current science that integrates information on mercury from sources to seafood.

Marine mercury primer: sources to seafood

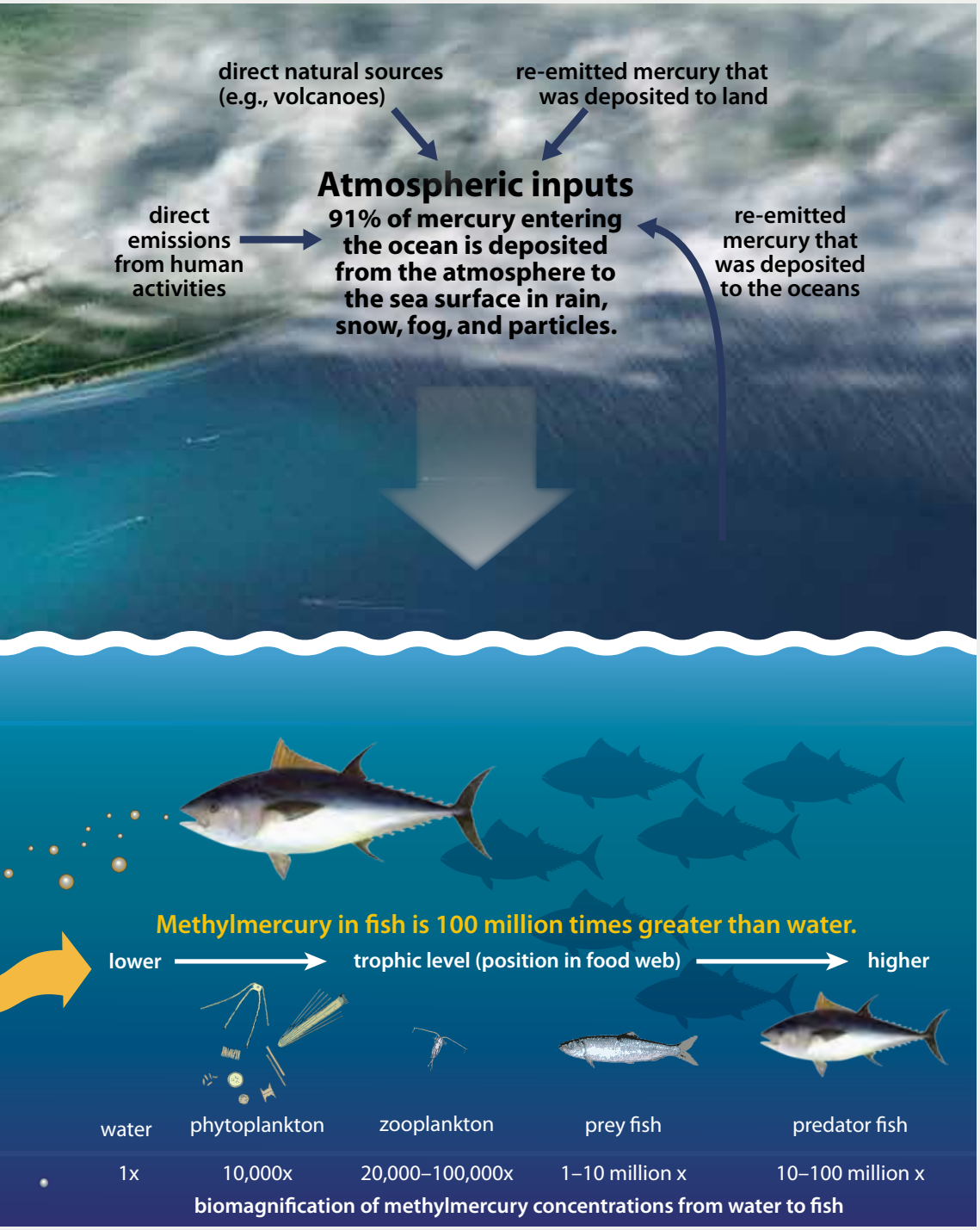
Mercury is a naturally occurring element, but human activities have greatly accelerated its release into the environment leading to widespread mercury pollution. The pathways and consequences of mercury pollution vary across marine systems and are influenced by three main factors: 1) the magnitude of sources and the transport of this mercury; 2) differences in the amount of methylmercury produced within marine systems; and 3) variation in methylmercury bioaccumulation and biomagnification in food webs that ultimately provide fish and other seafood for human consumption (Figure 3).

Figure 3. Mercury in the Open Ocean: Sources to Seafood



Mercury Sources and Transport

Mercury is released to the environment in several ways, but the dominant pathway for long-range transport is atmospheric emissions and deposition. Total mercury emissions are made up of primary sources that transfer mercury from geologic reservoirs to the atmosphere and can be natural or human-generated, and secondary (re-emission) sources. Today, roughly two-thirds of the mercury emitted to the atmosphere annually originates from current and past human activities since industrialization (Corbitt et al. 2011). The two largest primary anthropogenic sources of atmospheric mercury emissions globally are stationary fossil fuel sources, primarily coal-fired power plants; and artisanal gold mining, a source that appears to be widely underestimated (FIGURE 4).



After being emitted to the atmosphere, mercury is deposited to the Earth in rain and snow, and as gases and particles. It can deposit directly onto the ocean surface, or deposit to upland watersheds and eventually enter coastal waters via rivers. The timing and location of mercury deposition is determined by its chemical form, the location of emission sources, and environmental conditions. Atmospheric mercury occurs in different chemical forms. Oxidized gaseous and particulate forms (i.e., ionic mercury) are very reactive and deposit within hours

Figure 3: A conceptual diagram showing mercury sources and cycling in the open ocean, and methylmercury bioaccumulation and biomagnification in the ocean food web (adapted from Mason et al. 2012). Top background illustration by William W. Scavone.

to days after emission, typically within several hundred miles of the source. In contrast, elemental mercury can remain in the atmosphere for months to a year, and may deposit locally, regionally, or disperse globally before being deposited.

Mercury enters marine systems largely by direct deposition from the atmosphere to the ocean surface, but also through ocean currents that transport atmospherically-deposited mercury from the open ocean to nearshore areas, and from rivers draining the upstream watershed. Once inorganic mercury enters the marine environment it can accumulate in the deep ocean, be buried in sediments, be converted to elemental mercury and re-emitted back to the atmosphere, or be converted to methylmercury.

Figure 4. **Global Mercury Emissions from Human Activities**

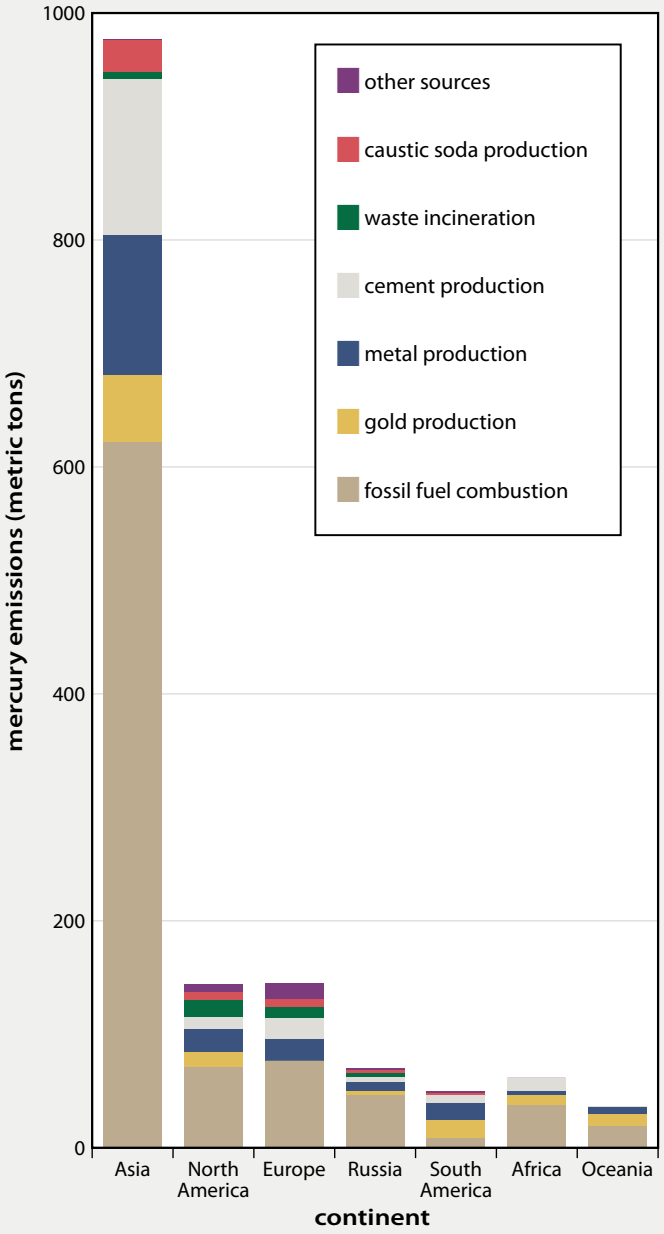
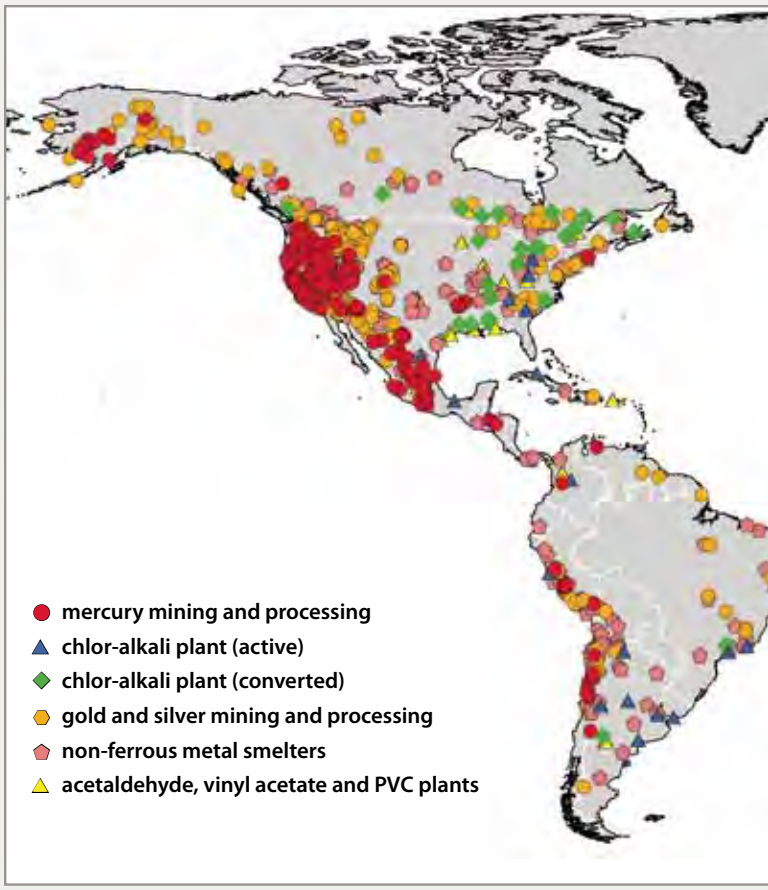


Figure 4: Globally, Asia emits more mercury to the atmosphere than any other major geographic region. Emissions from fossil fuel combustion, primarily coal-burning power plants, are the major anthropogenic source of mercury emissions (AMAP/UNEP 2008); however, emissions from artisanal gold mining are an important and underestimated source at present (AMAP/UNEP 2012).

Mercury discharges to land and water from active mining, industrial processes (e.g., chlorine production), runoff from developed lands, and wastewater discharges can be substantial sources to downstream coastal waters via river inputs. Releases of mercury within a watershed can also occur from legacy sources, such as closed mines, de-commissioned chemical facilities, and contaminated soils and sediments. These legacy sources generate a continual supply of mercury for decades or even centuries. A recent global inventory identified more than 2,700 contaminated

Figure 5. **Mercury-Contaminated Sites**



sites associated with mercury mining, metal production and processing, and other industries (FIGURE 5; Kocman et al. submitted). In specific local areas, mercury releases from contaminated sites can be orders of magnitude greater than mercury from atmospheric deposition.

Methylmercury Production

After mercury is released to the environment, it undergoes important transformations that drive human exposure and effects. Most mercury is released to the environment as inorganic mercury, but can be converted by bacteria to the organic form, methylmercury, usually in aquatic environments. Methylmercury can either be produced within an upland watershed and transported to downstream coastal waters, or can be produced internally within the marine system. Internal production can occur in coastal wetlands or sediments or in the water column of the open ocean. Methylmercury combined with inorganic forms of mercury constitute "total mercury."

Bioaccumulation and Trophic Transfer

Methylmercury is the form that is readily absorbed by organisms and that bioaccumulates in living tissues. After methylmercury is produced, it is taken up by microscopic algae that are eaten by zooplankton (such as small crustaceans), which are consumed by small fish, which are in turn eaten by large fish. Methylmercury *bioaccumulates* within organisms and is stored in muscle tissue where it is not easily eliminated. This results in *biomagnification* along the food chain as predator eats prey. The transfer of methylmercury through the food chain is influenced by factors such as the number of steps and nature of the food chain, organism growth rate, and supply of nutrients. Organisms at the top of the food chain that are exposed to high levels of contamination can have high methylmercury concentrations. Long-lived, predatory fish, such as swordfish and tuna, can have methylmercury levels as much as ten to 100 million times higher than methylmercury concentrations in the surrounding ocean water (FIGURE 3; Mason et al. 2012).

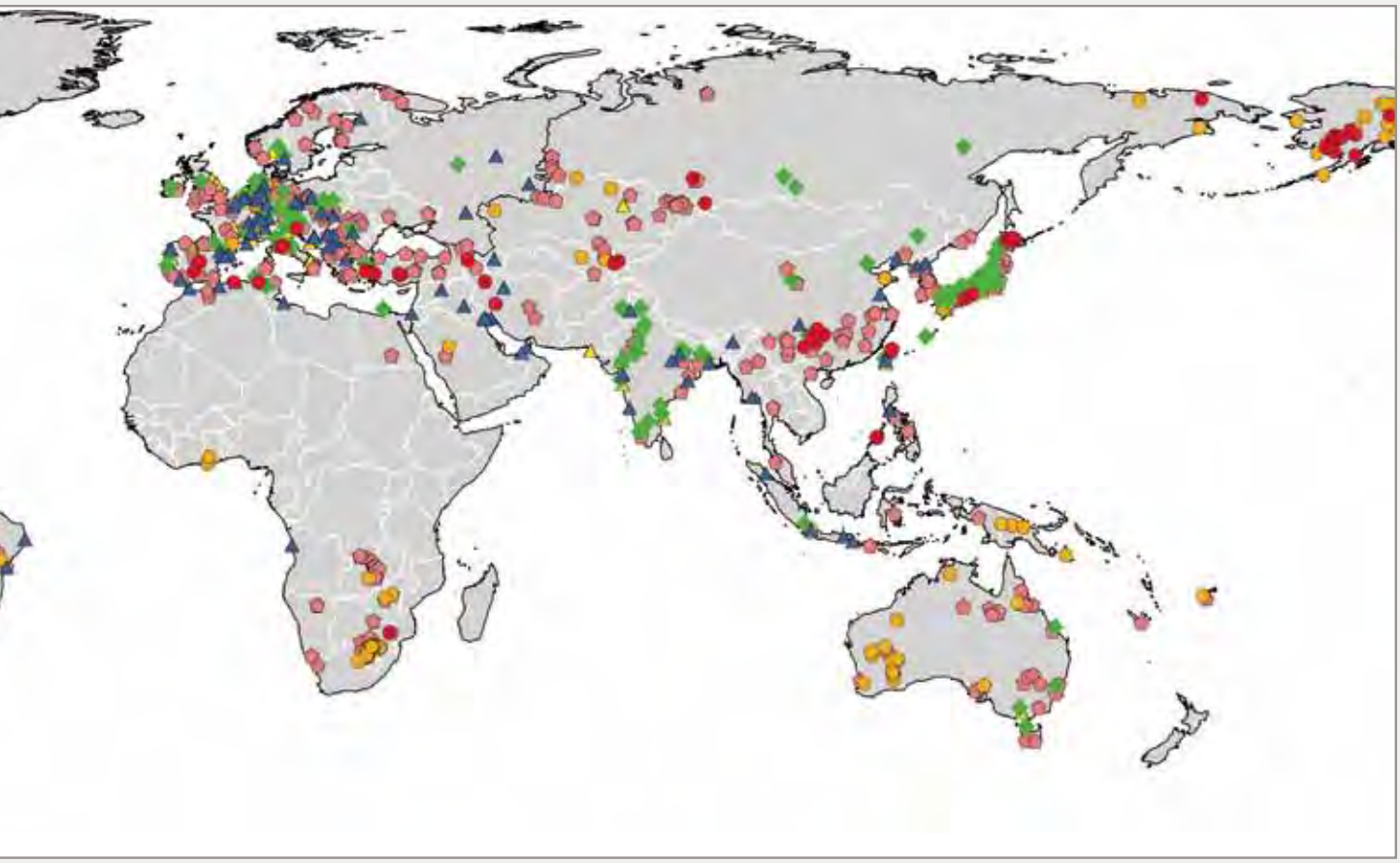
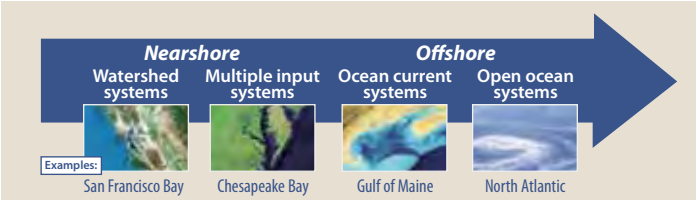


Figure 5: Global distribution of known sites contaminated with mercury from active and legacy mercury mining and processing (953 sites), gold and silver mining and processing (764), non-ferrous metal smelters (577), chlor-alkali plants (256 sites including active and converted), and factories which used or may have used mercury as a catalyst to produce acetaldehyde, polyvinylchloride (PVC) and vinyl acetate (156 sites). Map by D. Kocman, Jožef Stefan Institute (Kocman et al. submitted).

II. How are oceans and coastal waters polluted by mercury?

Mercury pollution enters the marine environment along distinct pathways that are linked to different mercury sources. Atmospheric inputs from global sources of mercury emissions dominate the "open ocean" and "ocean current" systems. Riverine mercury inputs dominate coastal waters that are "watershed systems." Some coastal waters are "multiple input" systems that reflect both atmospheric and river inputs.



The pathways of mercury pollution and the magnitude of total mercury and methylmercury supply vary across marine systems. Understanding and quantifying these contrasting pathways is useful in developing effective policy and management strategies for decreasing human exposure to methylmercury from seafood consumption.

Open Ocean Systems

Open ocean systems are deep basins that cover most of the Earth's surface. They include the North and South Pacific oceans, the North and South Atlantic oceans, and the Indian Ocean. Due to their size and distance from land, open ocean systems are not influenced substantially by watershed sources of mercury pollution. The C-MERC effort focused primarily on the Atlantic and Pacific oceans, because they are relatively well-studied and constitute important global fisheries.

In open ocean systems, it is estimated that atmospheric deposition dominates inputs of total mercury (about 90%; *Figure 6a, 6b*). Mercury deposited on the ocean's surface originates from atmospheric emissions such as fossil fuel combustion (including coal-fired power plants), artisanal gold mining, natural sources, and secondary emissions. This input of mercury is noteworthy for two reasons. First, atmospheric deposition should respond relatively rapidly to emission controls. Second, inorganic mercury from atmospheric deposition is more readily converted to the more toxic form of methylmercury than is inorganic mercury from watershed sources (*Figure 7*; Harris et al. 2007, Munthe et al. 2007).

Atmospheric deposition is considered a source of "new" mercury which is more easily transferred to biota than existing mercury in water, sediments, and soil. In the open oceans, mercury from the atmosphere is converted to methylmercury *within* the water column at intermediate depths (between 100 and 1,000 meters) as microorganisms break down sinking organic particles. Scientists have yet to identify the exact organisms and mechanisms involved, although it seems clear that the process is different from what occurs in freshwater systems, wetlands, and coastal areas (Lehnherr et al. 2011, Mason et al. 2012). Methylmercury that builds up, or bioaccumulates, in open ocean fish, such as tuna, is likely produced in the open ocean rather than transferred from estuaries and coastal waters (Blum et al. 2008, Senn et al. 2010).

The open ocean and the Arctic Ocean are relatively efficient in converting inorganic mercury inputs to methylmercury (*Figure 7*). Although the external input of total mercury to the open ocean is low on a "per unit of surface area per year" basis, compared to other marine systems studied, the sheer mass of the oceans and its high methylation efficiency make the internal production of methylmercury in the oceans a globally important process. Moreover, while only a small fraction (about 10%) of the methylmercury produced in the ocean moves into the food web and accumulates in fish (*Figure 3*), this relatively small fraction of a very large pool of mercury is the source of contamination for important ocean fisheries that supply seafood to most of the world's population (the Pacific and the Northeast Atlantic fisheries together supply 63% of the global marine catch; FAO 2011).

In addition to open oceans, other marine systems receive a sizeable fraction of their total mercury inputs from direct atmospheric emissions and deposition (*Figures 6a, 6b*). These include the Arctic Ocean (32%), Chesapeake Bay (38%), and the Gulf of Maine (26%). However, direct atmospheric inputs are not the largest fraction of the total mercury inputs in these systems.

Ocean Current Systems

Ocean current systems refer to gulfs, bays, and estuaries that receive most of their mercury from ocean currents

Figure 6. Mercury and Methylmercury Inputs to Marine Systems

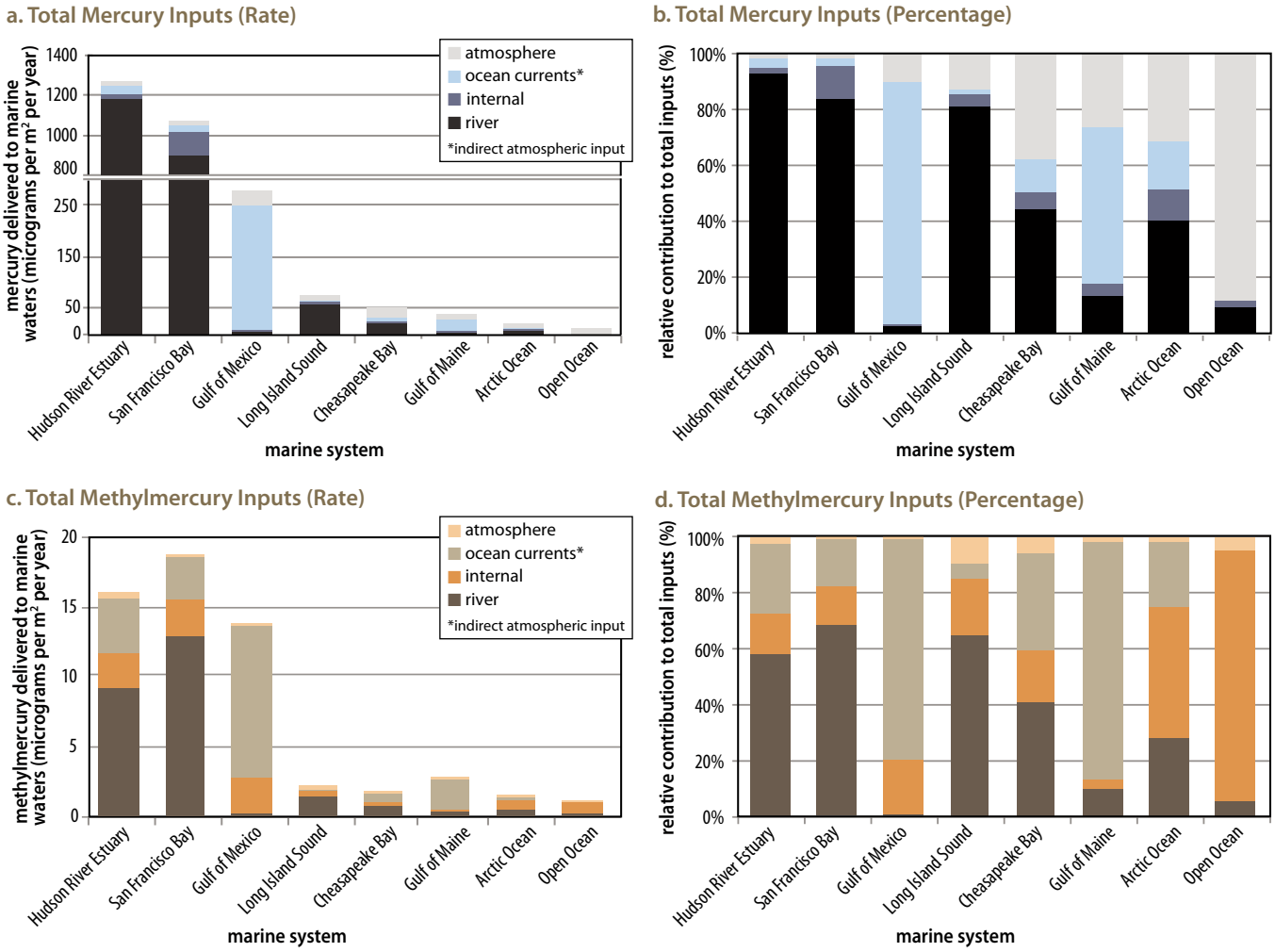


Figure 6: The rate and pathways of mercury and methylmercury inputs to marine systems vary widely. For example, Hudson River Estuary receives high annual inputs over a small basin area from river inflows linked to mercury sources in the watershed. The open ocean receives lower annual inputs over a very large area, mostly from atmospheric inputs linked to mercury emissions sources. Note that while the annual inputs to the open ocean are relatively low, the basin surface area is very large resulting in high absolute inputs of mercury. The inputs are based on information from the literature when possible (e.g., Balcom et al. 2004, 2008, 2010, Davis et al. 2012, Harris et al. 2012a, 2012b, Kirk et al. 2012, Mason et al. 1999, 2012, Sunderland et al. 2012). However, due to differences in the methodologies of estimation, the final values used in these figures may differ from the published estimates, but are mostly within their relative error (a factor of two to three).

that transport mercury from the open ocean into coastal areas. Since atmospheric deposition is the dominant source of mercury to the open ocean, ocean current systems receive atmospheric deposition of mercury indirectly. Like the open ocean, these systems are expected to respond relatively rapidly to controls on mercury emissions. The Gulf of Maine and the Gulf of Mexico are two examples of ocean current systems dominated indirectly by atmospheric deposition (*Figures 6a, 6b*).

Gulf of Maine – Based on model estimates, 56% percent of the total mercury inputs and 85% of methylmercury inputs to the Gulf of Maine come from mercury emissions that are deposited to the Atlantic Ocean and then transferred to the Gulf by currents (*Figures 6b, 6d*). Mercury pollution in this region affects some of the world's most productive fisheries and populations of whales, porpoises, seals, and many bird species (Pesch and Wells 2004, Thompson 2010, Sunderland et al. 2012). As in many coastal regions of the United States

and Canada, high mercury concentrations have been observed in fish and wildlife from the Gulf of Maine since measurements were first made in the 1970s. For example, harbor porpoises in the Gulf of Maine region had mercury muscle tissue concentrations of 1.12 ppm (Gaskin et al. 1979). Moreover, mercury concentrations in marine birds in the region are elevated and for some species exceed reproductive effect thresholds (Goodale et al. 2008).

Gulf of Mexico – Based on model estimates, nearly 90% of the total mercury and 80% of the methylmercury originate from atmospherically-deposited mercury that is transported from the Atlantic Ocean via the Loop Current entering from the Yucatan Channel (*Figures 6b, 6d*). However, recent model estimates suggest that mercury pathways in the Gulf vary geographically: mercury inputs in the central Gulf are dominated by delivery via ocean currents; mercury inputs in the coastal delta waters are dominated by waters draining the Mississippi and Atchafalaya rivers; and the major input along the coasts of Florida and Louisiana is direct atmospheric deposition (Harris et al. 2012a, 2012b). Fisheries of the Gulf of Mexico are important to the entire United States, accounting for 41% of the marine recreational fish catch and 16% of the marine commercial fish landings (Harris et al. 2012a). A recent data compilation suggests that fish such as tunas, mackerels, ribbonfishes, and bonitos in the Gulf of Mexico have average total mercury concentrations of approximately 0.7 ppm (Evers 2011). Moreover, average per-capita fish consumption in the Gulf of Mexico region is twice the U.S. national average, and recreational fishers in the region have a potential for elevated exposure to methylmercury (Harris et al. 2012a).

Figure 7. Methylmercury Production in Marine Systems

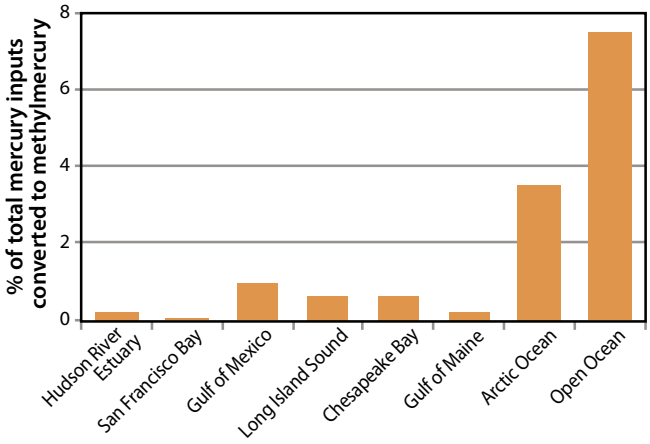


Figure 7: The fraction of total mercury inputs that is converted in the marine basin to methylmercury is an important controller of the ultimate uptake of methylmercury in fish and other seafood. This figure shows that the open ocean is the most efficient marine system at producing methylmercury from total mercury inputs. This pattern suggests that methylmercury in the open ocean also may be highly responsive to decreased inputs of mercury from external sources.

Watershed Systems

Watershed systems are coastal waters where mercury is primarily delivered by rivers that drain upland watersheds. Mercury supplied from the watershed can originate from ongoing industrial sources, urban runoff, and wastewater discharges; from the legacy of mercury left by past industry such as mining, felt making, and chlorine production; and from atmospheric deposition to the watershed. Many of these coastal watershed systems, such as the Hudson River Estuary, San Francisco Bay, and Long Island Sound, have highly contaminated bays due to historic and ongoing industrial sources of mercury.

As the freshwater from the river mixes with salt water from the ocean, mercury dissolved in river water and attached to particles settles to coastal sediments (Fitzgerald et al. 2007, Rice et al. 2009) where it can be transformed to methylmercury. In such cases, sediments can become an "internal source" of methylmercury production to the coastal system.

Hudson River Estuary – Balcom et al. (2008) investigated mercury in the Hudson River Estuary and found that river inputs account for 93% of total mercury inputs (*Figures 6a, 6b*). Likewise, river inflows supply 58% of the methylmercury to the estuary (*Figures 6c, 6d*), with smaller contributions from open ocean currents (25%) and internal sediment supply (15%). The watershed area that supplies mercury is large relative to the size of the estuary, which explains in part why riverine inputs dominate over direct atmospheric inputs. The large annual inputs of mercury to the Hudson River estuary (about 1,200 micrograms per square meter per year; $\mu g/m^2$-yr; *Figure 6a*) originate from runoff draining large urbanized areas carrying atmospherically-deposited mercury and mercury from current and legacy industrial sources. Legacy mercury sources

include the manufacturing of fluorescent and high-intensity discharge lamps, thermometers, measurements and control instruments, thermostats, switches and relays, catalysts in production of organic chemicals, and dental amalgams (Fitzgerald and O'Connor 2001). The resulting methylmercury bioaccumulation in the estuary occurs in important estuarine species including striped bass and white perch (Levinton and Pochron 2008, Goto and Wallace 2009).

San Francisco Bay – Like the Hudson River Estuary, annual total mercury inputs on an area basis to San Francisco Bay are more than one hundred times greater than rates of atmospheric deposition of mercury to the open ocean (*Figure 6a*). Mercury that enters San Francisco Bay largely derives from historic mining in the New Almaden mercury mining district in the Guadalupe River watershed and in the Sierra Nevada Mountains (Davis et al. 2012). Contaminated soils and sediments persist in the watershed and Bay, fueling production of methylmercury and contamination of local food webs. This legacy mercury is slowly eroded from soil and is associated with particles and dissolved organic matter that wash down into the Bay. Sediments currently entering the Bay from many local watersheds (both with and without historical mining) have comparable or higher mercury concentrations than those already in the Bay. Unless inputs of mercury from these watershed pathways are controlled or the methylation process interrupted, the problem of mercury contamination will likely continue indefinitely. Exposure to methylmercury in San Francisco Bay anglers is predominantly through harvest of local fish, including striped bass, which have some of the highest mercury concentrations measured in the United States.

Long Island Sound – Annual total mercury inputs to Long Island Sound are relatively high and dominated by river inputs (81%), with a small contribution from direct atmospheric deposition to the water surface (13%; *Figures 6a, 6b*). The rate of mercury input on an area basis for Long Island Sound, while much lower than either the Hudson River Estuary or San Francisco Bay, is still high and reflects the developed nature of the watershed with both atmospheric mercury deposition to the watershed and industrial and municipal sources of mercury. Methylmercury also is supplied largely by rivers (65%), with some produced internally (21%), and less supplied by exchange with the open ocean (5%; *Figures 6c, 6d*). Long Island Sound contains valuable natural resources including fish and shellfish.

Multiple Input Systems

Many marine systems receive a substantial amount of mercury from multiple inputs: direct and indirect atmospheric deposition as well as rivers. In some cases, the sources of the river inputs are largely atmospheric mercury deposition to the watershed. In highly urbanized watersheds, the rivers also carry mercury from ongoing or legacy sources to local rivers and coastal waters. The Arctic Ocean and Chesapeake Bay provide contrasting examples of multiple input systems.

Arctic Ocean – Total mercury inputs to the Arctic Ocean reflect a mix of direct atmospheric deposition (32%), indirect atmospheric deposition via ocean currents (17%), and river inputs (40%; *Figure 6b*). Unlike more developed systems, river mercury inputs to the Arctic Ocean derive almost entirely from atmospheric mercury emissions and deposition to the watershed. Moreover, the supply of mercury inputs on an annual area basis to the Arctic Ocean is relatively low (20.4 µg/m^2-yr) in contrast to more industrialized regions such as Hudson River Estuary or Long Island Sound (*Figure 6a*). However, the rate of mercury input is high compared to the open ocean (13 µg/m^2-yr). Mercury pollution contaminates the Arctic Ocean food web that supplies protein to local indigenous communities, where harp seals, narwhal, and halibut account for the majority of methylmercury exposure to humans *(Figure 8d*).

Chesapeake Bay – Chesapeake Bay drains a large watershed with a wide range of human land uses, and mercury originates from watershed sources and both indirect atmospheric deposition transported to the Bay in ocean currents and direct atmospheric deposition to the surface of the Bay. As a result, total mercury inputs to the Bay reflect a mix of river inflows (44%), direct atmospheric deposition (38%), and indirect atmospheric deposition transported from the open ocean (11%; *Figure 6b*). Methylmercury inputs are mixed, originating from river inflows (41%), internal production (19%), and exchange with the open ocean (35%; *Figure 6d*). Like the Arctic Ocean, the Chesapeake Bay is a locally and regionally important source of fish and seafood (*Figure 8e*).

III. Who is exposed to mercury in seafood?

Most seafood consumers are "general consumers" whose methylmercury intake comes from fish typically harvested from the open oceans which receive atmospheric inputs from global mercury emission sources. Methylmercury intake by "local consumers" comes from seafood caught from nearby coastal waters that receive riverine inputs from local, regional, and global sources.

General Seafood Consumers

Studies show that for the average American and many global "general consumers," a few types of seafood account for the majority of their methylmercury intake through fish consumption (*Figure 8a*). One of the most commonly consumed species is tuna, mostly in the form of canned "light" tuna and canned "white" tuna, but also fresh and frozen tuna (Sunderland 2007, Groth 2010). More than 60% of the global tuna harvest comes from the Pacific Ocean and the rest from the Atlantic Ocean, Indian Ocean, and Mediterranean Sea (FAO 2010). In the United States, more than 75% of the methylmercury exposure from the seafood eaten comes from fish caught and consumed from the open oceans (Pirrone and Keating 2010; *Figure 8b*). Even in coastal regions such as the Atlantic seaboard and Gulf of Mexico, the most popular seafood species are not local but rather store-bought shrimp, tuna, or salmon (Mahaffey et al. 2009). Therefore, most people who consume seafood are exposed to methylmercury from fish that are harvested from the surface waters of the open oceans—the areas of the ocean where mercury contamination comes directly from sources of atmospheric emissions and deposition.

Local Seafood Consumers

In contrast to general consumers, "local consumers" of fish and other seafood are more likely to live in coastal areas and include recreational anglers who eat their catch; people who rely on local marine fish and marine mammals for a majority of their protein and nutrition; immigrant communities who catch their own fish and may have different eating habits, such as consuming whole fish instead of fillets; and consumers who prefer to eat local seafood. Local consumers are prevalent in all coastal areas, including contrasting C-MERC study regions of San Francisco Bay, the Arctic Ocean, and Chesapeake Bay.

San Francisco Bay – San Francisco Bay is a popular fishing area for both recreational and subsistence anglers of diverse socioeconomic and ethnic backgrounds (Gassel et al. 2011, Davis et al. 2012). One survey reported that 66.6% of fish eaten by San Francisco Bay anglers were caught in the Bay, and 33.3% were from other sources, largely fish purchased from grocery stores (SFEI 2000). An estimate of mercury intake by these fish consumers suggests that 85% of their total mercury intake is from consumption of locally caught fish, mostly striped bass (*Figure 8c*). This profile of a "local" seafood consumer highlights how controls on local watershed sources of contamination (e.g., mitigation of legacy sources) are likely to have the strongest influence on mercury intake for this local consumer population.

The Arctic – The data from Greenland offer a contrasting example of the local consumer in the Arctic (*Figure 8d*). Northern peoples living in the Arctic harvest and rely on marine mammals and fish for subsistence and cultural survival. Methylmercury is present in numerous Arctic marine mammals, such as ringed seals and beluga whales, at concentrations high enough to pose health risks to Northern peoples consuming these animals as traditional foods (AMAP 2011, Kirk et al. 2012). Although Arctic populations are local consumers, their mercury intake is likely to be affected by controls on global sources of mercury emissions, since the majority of the mercury in the Arctic originates from atmospheric emissions and deposition.

Chesapeake Bay – Consumers who live in regions with commercial fisheries, such as the Chesapeake Bay, have methylmercury intake from both local and non-local sources (*Figure 8e*). Even though most fish species caught in Chesapeake Bay are consumed locally, a high percentage of the total fish consumed (~30%) is canned tuna

that is harvested from the open ocean. Therefore, methylmercury intake by local consumer populations in this and similar regions would be limited by both controls on local sources in the watershed that decrease riverine inputs, and by global controls on atmospheric emissions of mercury that curb atmospheric inputs (see page 18).

Figure 8. Mercury Intake by Seafood Type for General and Local Consumers

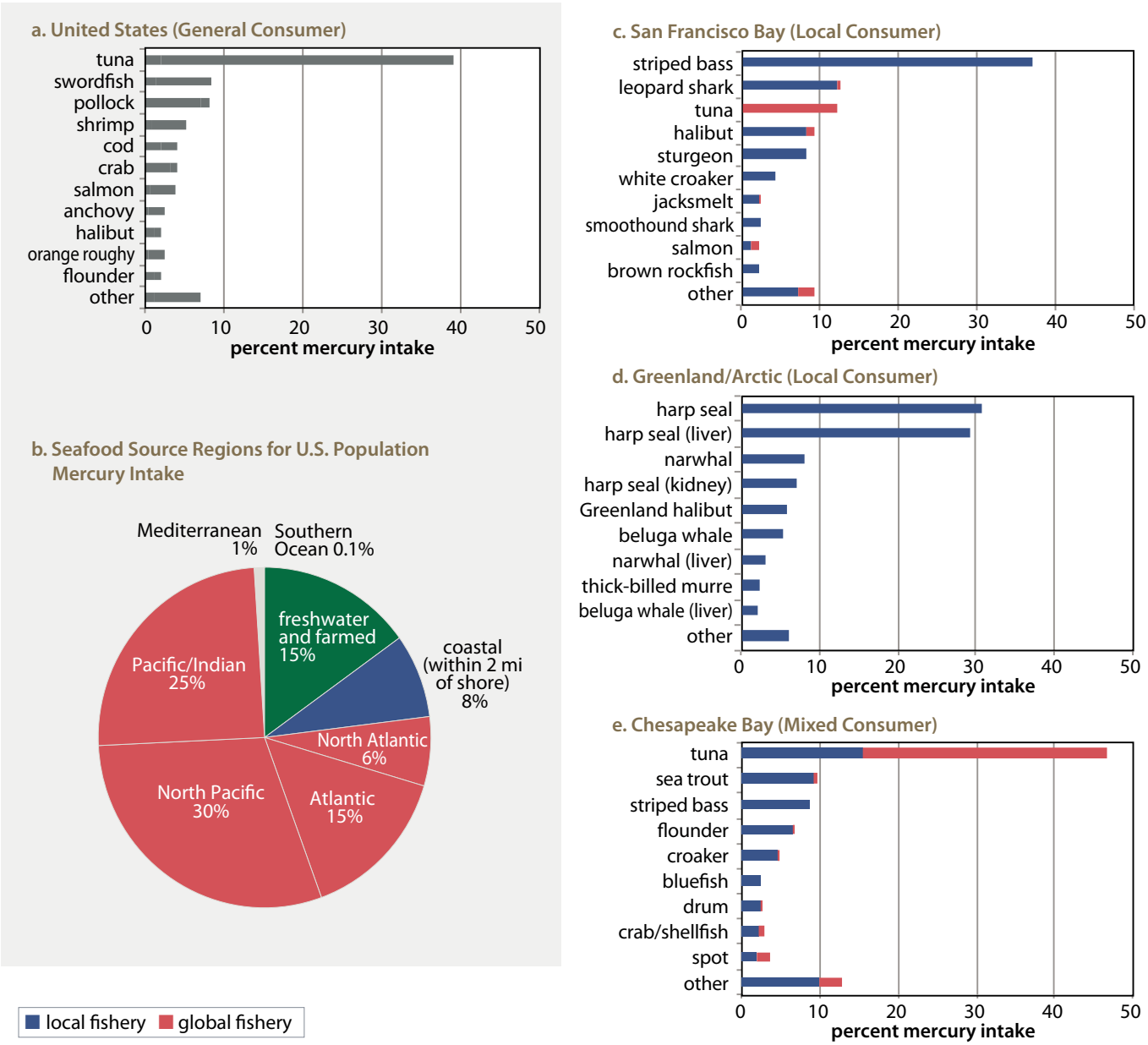


Figure 8: Estimated contribution of locally and globally sourced seafood species to total mercury intake of (a) the "general consumer" as represented by population-wide estimates for the United States (Sunderland 2007) and (b) the relative contribution of seafood source regions to mercury exposure of the U.S. general consumer (Pirrone and Keating 2010); (c) a "local consumer" as represented by recent consumers of recreationally caught fish in the legacy-contaminated San Francisco Bay (estimated using data from Sunderland 2007, Davis et al. 2011, Gassel et al. 2011); (d) a "local consumer" from the Arctic as represented by population-wide estimates for Greenland (Johansen et al. 2004); and (e) consumers exposed to mercury from both local and global seafood sources in the Chesapeake Bay region (Mason 2012).

IV. How will marine systems respond to mercury controls?

Methylmercury concentrations in marine fish will decline roughly in proportion to decreases in mercury inputs, though the timing of the response will vary. Methylmercury in open ocean fish will begin to decrease within several years to decades after emissions controls. In contrast, methylmercury in fish from coastal systems may exhibit a range of response times over many decades to centuries, depending on the relative importance of atmospheric to other inputs.

For centuries, the open oceans have been responding to changes in atmospheric emissions and deposition of mercury associated with human activities. Model calculations suggest that mercury in the ocean surface (above 100 meters) has doubled over the past 100 years in response to three-fold increases in emissions (Mason et al. 2012). Although long-term data for fish mercury are limited, analyses of archived samples of bird feathers and eggs show increases in mercury that parallel estimated increases in methylmercury concentrations in surface waters in the open ocean during this period. Mercury concentrations in birds appear to have increased by a factor of four in the North Atlantic over the past century (Monteiro and Furness 1997, Monteiro et al. 1998). Methylmercury has also increased by a factor of two to three in the black-footed albatross of the North Pacific over the last century (Vo et al. 2011).

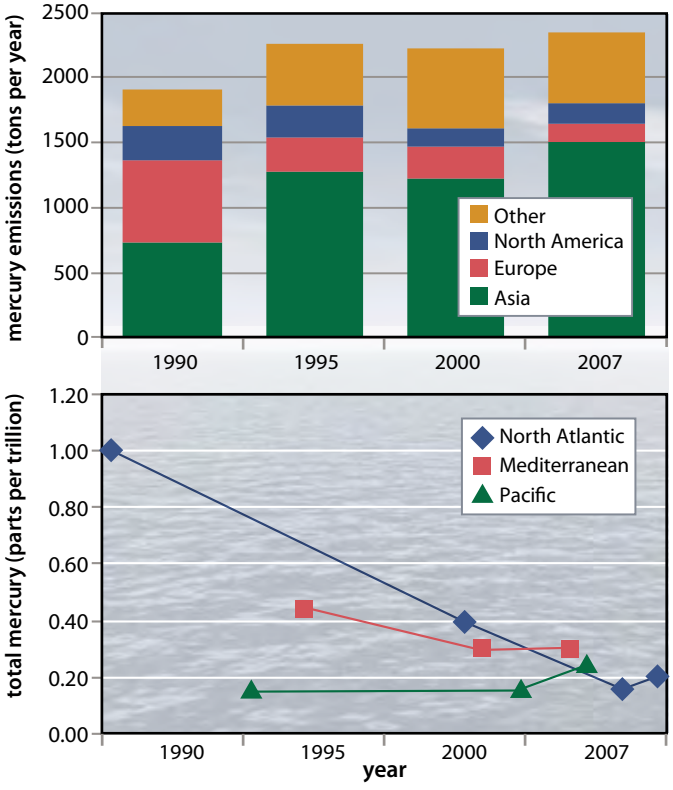
Since 1990, mercury emissions from Europe and North America have decreased while total global mercury emissions have increased 17%, largely due to a marked increase in emissions from Asia (*Figure 9a*; Pacyna et al. 2006, Streets et al. 2009, Pirrone et al. 2010). Recent measurements of mercury concentrations in the surface waters of the open oceans appear to reflect this regional shift in mercury emissions. Specifically, there has been an increase in mercury in the Pacific Ocean and a decrease in the Atlantic Ocean as well as the Mediterranean Sea over the last two decades (*Figure 9b*; Mason et al. 2012).

Recent model estimates suggest that changes in mercury concentrations in the upper waters of the oceans will produce changes in methylmercury concentrations in fish within decades (Sunderland and Mason 2007). This relatively rapid response is because methylmercury production as well as the foraging of fish consumed by humans occur largely in the surface and subsurface regions of the open ocean that are responsive to cuts in atmospheric emissions and deposition.

In the United States, there have been significant declines in atmospheric mercury emissions (60% from 1990 levels; U.S. EPA 2005, Schmeltz et al. 2011). Emission controls have been accompanied by proportional decreases in mercury concentrations in marine and

Figure 9. Trends in Global Mercury Emissions and Ocean Mercury

a. Trends in Atmospheric Emissions



b. Trends in Ocean Mercury Concentrations

Figure 9a: Mercury emissions to the atmosphere from human activities have decreased in Europe and North America since 1990 and have increased in Asia (Pacyna et al. 2006, Streets et al. 2009, Pirrone et al. 2010). **Figure 9b:** Concentrations of mercury in the surface and subsurface ocean are increasing in the Pacific Ocean and decreasing in the Atlantic Ocean and Mediterranean Sea, reflecting regional shifts in total global mercury emissions (Mason et al. 2012).

freshwater systems as observed in sediments, fish, and wildlife (e.g., Balcom et al. 2010, Evers et al. 2011, Monson et al. 2011, Drevnick et al. 2012).

A marked decline has also occurred in direct discharges of mercury to waters from wastewater and industrial sources in some U.S. waters. For example, in the Hudson River Estuary, there have been substantial decreases in mercury loading from the Hudson River since the 1960s (Fitzgerald and O'Connor 2001, Balcom et al. 2010). During this same period, there were large and rapid decreases in mercury in four fish species observed for the Hudson River and its estuary (Levinton and Pochron 2008). In the Gulf of Maine, sediment and mussel mercury concentrations are elevated in embayments near large historical sources (Sunderland et al. 2012), yet in some cases concentrations of mercury in mussels have declined where there have been controls on local industrial sources (*Figure 10*). These examples suggest additional future improvements in fish mercury levels will reflect the extent to which sources are controlled and the magnitude of those controls.

Figure 10. Mercury in Blue Mussels – Gulf of Maine, 1990-2007

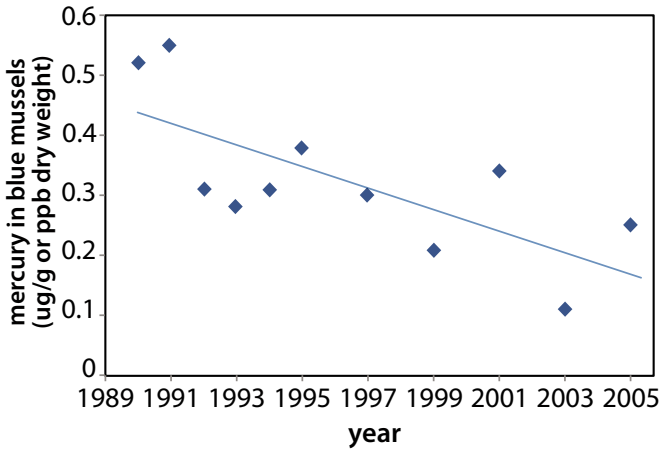


Figure 10: Mercury concentrations measured in blue mussels (ppb dry weight) near Brewster Island, Massachusetts, as part of the Gulfwatch monitoring program in the Gulf of Maine. Decreasing trends reflect improvements in response to controls on industrial releases of mercury from wastewater treatment plants and chlor-alkali plants in the watershed. A similar pattern is seen in mussels from Gaphead, Mass., and Sears Island, Maine (Sunderland et al. 2012).

How might future mercury controls affect mercury in fish?

Model calculations were made to examine changes in fish mercury concentrations in contrasting marine basins in response to hypothetical decreases in atmospheric mercury deposition (20% decrease) and watershed mercury inputs (20%). It is important to note that the simple model used in these calculations assumes steady-state conditions (i.e., the modeled system is in balance with mercury inputs and the model is not time-dependent). In addition, it is assumed that the 20% decrease in atmospheric mercury deposition also results in a 10% decrease in mercury inputs from the watershed. Simulations were run for the North Atlantic Ocean, an *open ocean system* in which mercury inputs largely occur by atmospheric deposition; Long Island Sound, a *watershed system* with large watershed mercury inputs; and Chesapeake Bay, a *multiple input system* with a mix of watershed and atmospheric inputs of mercury (Figures 6, 11). The results from these simulations of hypothetical decreases in mercury inputs were compared to simulations for the same systems under current conditions (i.e., no decrease in mercury inputs).

Model simulations suggest that fish mercury concentrations in the North Atlantic are relatively responsive to decreases in atmospheric emissions and deposition of mercury, but not as responsive to decreases in watershed mercury inputs (Figure 11). These results are consistent with the characterization that atmospheric deposition is the dominant pathway of mercury inputs to the open ocean (Figure 6b). Long Island Sound is more responsive to controls on watershed mercury inputs, as would be expected based on its characterization as a *watershed system* with large watershed mercury inputs. Long Island Sound also responds to decreases in atmospheric emissions, in part because atmospheric deposition to the watershed supplies mercury to the downstream marine basin by river flows. Finally, decreases in atmospheric emissions and deposition are moderately more effective at achieving decreases in fish mercury concentrations than watershed controls in Chesapeake Bay, a *multiple input system* with a mix of watershed and atmospheric inputs of mercury. Again, this is because decreased mercury emissions will not only decrease direct deposition to the Bay and mercury inputs from ocean currents, but also will decrease mercury inputs from the watershed associated with inputs of atmospheric deposition.

Figure 11. **Fish Mercury Response to Scenarios of Decreased Mercury Inputs**

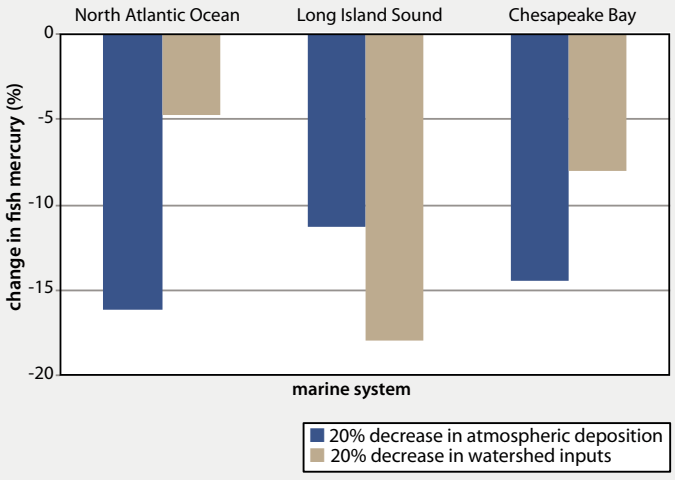


Figure 11: Simulations using a steady-state (no time dependency) model of changes in fish mercury concentrations in selected marine basins in response to a hypothetical 20% decrease in atmospheric mercury deposition and 20% decrease in watershed inputs of total mercury to the basin. Note that simulations of decreases in atmospheric mercury deposition reflect 20% decreases in direct deposition to the basin and 20% decreases in the supply of mercury due to mixing of ocean water with the basin, and a 10% decrease in mercury deposited to the watershed from atmospheric mercury deposition. The changes in fish mercury concentrations shown reflect simulated changes relative to fish mercury concentrations in the absence of any controls on mercury inputs to the basin.

Based on past trends and current mechanistic understanding of mercury cycling in oceans, the fisheries response to controls on atmospheric deposition to the open ocean is likely to occur within several years to several decades (Sunderland and Mason 2007). The fisheries response to controls on watershed mercury sources are likely to occur in two phases. When direct discharges are first eliminated, it is anticipated that mercury concentrations in fish will decline within a few years, as has been evident for the Hudson River Estuary (Levinton and Pochron 2008) and Gulf of Maine (Sunderland et al. 2012, Figure 10). However, if legacy sources and sediment contamination persist, the second phase of changes in fish mercury may be very slow and consumers of local fish could continue to be at risk for decades to centuries, as is the case for San Francisco Bay.

V. What are the implications of C-MERC science for mercury policy?

Mercury pollution has attracted considerable policy attention since the 1970s, when several nations developed agreements limiting the discharge of mercury directly into international waters. By the 1990s, individual states and nations had passed emission control regulations, limited discharges, and initiated remediation of industrial waste and mining sites, with measurable success. Nevertheless, mercury pollution persists and global progress has been hindered by the patchwork of federal and international policies that fail to adequately address the transboundary (e.g., atmospheric transport), multi-media (e.g., air to water to fish), and cross-cutting (e.g., pollutant interaction) challenges associated with mercury pollution in marine systems (Lambert et al. 2012).

In 2009, the Governing Council of the United Nations Environment Programme agreed to negotiate a legally binding mercury treaty among 140 nations. The stated goal of the treaty is to "protect human health and the global environment from the release of mercury and its compounds by minimizing and, where feasible, ultimately eliminating global, anthropogenic mercury releases to air, water, and land" (UNEP 2009). The international treaty process is slated for completion in 2013 with ratification and implementation continuing thereafter. These efforts and ongoing national policy initiatives represent important opportunities to comprehensively address mercury contamination in marine systems.

The four policy implications that emerge from this C-MERC synthesis are important for addressing mercury pollution in marine systems and human exposure from seafood consumption.

1. Given that most seafood consumers are general consumers, controlling sources of atmospheric mercury emissions will have substantial benefits for the largest fisheries that supply seafood to the most people.

The C-MERC synthesis suggests that previous national efforts to control sources of atmospheric mercury emissions have had a positive effect. Additional cuts in global mercury emissions will impact the open oceans (e.g., North Pacific, North Atlantic) that supply the majority of the world's seafood (*Figure 8b*), as well as coastal waters that have large indirect atmospheric inputs of mercury delivered by ocean currents or rivers (e.g., Gulf of Maine, Gulf of Mexico, the Arctic, Chesapeake Bay; *Figure 6b*). As such, controls on atmospheric emissions of mercury have the potential to benefit a large number of fish consumers locally and globally.

2. Controlling direct discharges and managing legacy sources of mercury can have a substantial impact on coastal fisheries that supply seafood to local populations of recreational and subsistence fish consumers.

Some coastal systems receive direct discharges of mercury into waterways or legacy mercury from contaminated sites. Controlling these sources should provide considerable benefits to local consumers. While this group represents a smaller population than the general consumer, some local consumers eat large amounts of fish from highly polluted waters (e.g., San Francisco Bay anglers). Beneficial control strategies for these coastal waters include curbing direct discharges (e.g., releases from wastewater treatments plants), mitigating legacy mercury from heavily contaminated sites (e.g., decommissioned mining operations or chlorine production facilities), and interrupting the methylation process to limit the bioaccumulation of existing mercury in fish (Davis et al. 2012, Matthews et al. in review). Simulations of control scenarios suggest that coastal waters with large river inputs will respond more to cuts from these local watershed sources than from atmospheric emissions and deposition (*Figure 11*).

3. Expanded marine monitoring of mercury in air, sediments, water, wildlife, and fish is needed to evaluate the effectiveness of national and international policies. Monitoring could also provide insight on the effects of global environmental change on mercury pollution in marine systems.

Controls on mercury sources are occurring concurrently with changes in nutrients, climate, fishing, and other environmental pressures. Excess nutrients, especially nitrogen, are a major problem facing coastal waters in developed areas around the world. A recent conceptual model suggests that decreases in nutrient levels may have unintended consequences for mercury bioaccumulation (Driscoll et al. 2012). Climate change also has the potential to alter the mercury cycle. For example, increasing air and water temperatures or changes in precipitation and river runoff patterns may alter ocean circulation and nutrient cycling, which could change marine food webs and their bioaccumulation of methylmercury. In the tropical coastal regions, climate change may alter annual wet and dry seasons which could impact mercury transport and transformation (Costa et al. 2012). Given these confounding factors and the lag times expected in the response of some marine systems to mercury controls, it is important to expand mercury monitoring in marine systems.

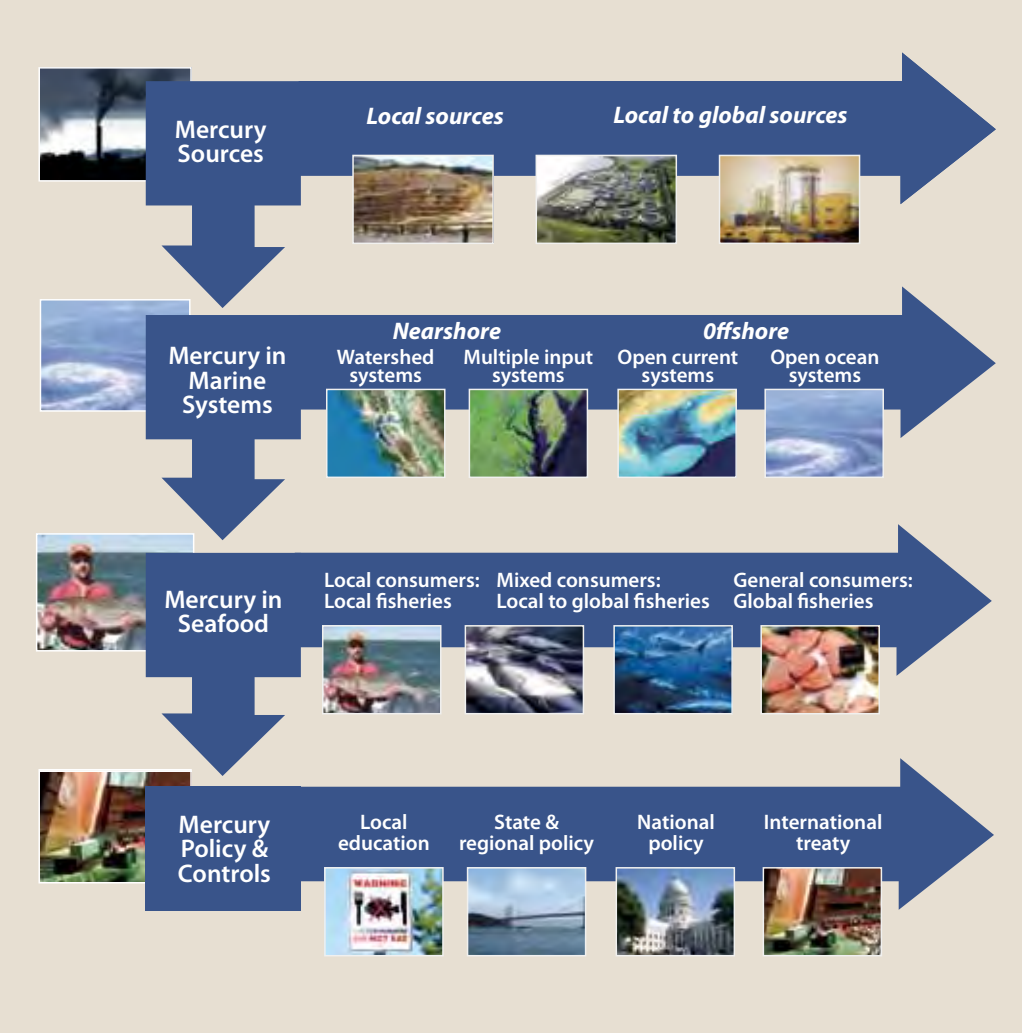
In order to evaluate the effectiveness of mercury control strategies, mercury and methylmercury should be broadly monitored in the atmosphere, water, and sediments of coastal waters and oceans; and these measurements should be linked to food web monitoring *in the same locations*. Food web monitoring should focus on wildlife, the fish species that people eat, and the geographic origin of food fishes, and link to information on mercury inputs and cycling to ensure that seafood measurements are interpreted accurately (Evers et al. 2008). While sampling for mercury has expanded in some of the oceans, most marine systems lack comprehensive mercury measurements.

4. Improved fish consumption advice could be beneficial, particularly for highly contaminated systems with large populations of local consumers. However, fish consumption advisories often have mixed results and are not a viable substitute for source controls.

Efforts to decrease human exposure to methylmercury traditionally rely on advisories that inform people about the need to limit their consumption of certain fish species. These limits may or may not be based on mercury data from local fisheries. While many advisories inform the consumer on the risks of consuming fish high in mercury, fewer also contain information on the health benefits of eating fish. Some studies have found that overall fish consumption decreases in response to advisories, with a parallel loss of nutritional benefits from eating fish (Oken et al. 2003, Shimshack et al. 2007, Shimshack and Ward 2010). Others suggest that local fishers are not aware of advisories or do not view them as credible (Gassel et al. 2011). Public health experts suggest that fish consumption advice should be improved to provide clear and simple messages on fish choice that integrate health, ecological, and economic impacts (Oken et al. 2012). Fish consumption advice, however, should be viewed as an interim strategy with variable effectiveness, not as a viable substitute for controlling mercury sources. ■

In Summary

The C-MERC initiative compiled and analyzed existing data from nine marine systems worldwide with varied mercury inputs, fish mercury levels, and seafood consumption profiles. By comparing data from across geographic regions, this C-MERC report links sources of mercury to seafood and consumers. In general, coastal waters can receive large riverine inputs of mercury from watershed sources that contaminate fisheries serving local consumers. In contrast, open oceans and large gulf systems largely receive mercury inputs from global atmospheric emissions and deposition that pollute fisheries serving worldwide populations of general consumers. The discernable local-to-global pattern of mercury pollution and exposure that emerged from the C-MERC synthesis can guide mercury policies and management strategies under consideration at local, national, and international levels.



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Note: **Bold** indicates C-MERC papers.

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