White Paper on Gulf of Mexico Mercury Fate and Transport:

Applying Scientific Research to Reduce the Risk from Mercury in Gulf of Mexico Seafood

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NOAA Technical Memorandum NOS NCCOS 192

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Cover photo: A large king mackerel coming aboard a sportfishing boat in the Gulf of Mexico. Credit NOAA Photo Library

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White Paper on Gulf of Mexico Mercury Fate and Transport:

Applying Scientific Research to Reduce the Risk from Mercury in Gulf of Mexico Seafood

David Evans
NOAA, National Ocean Service
National Centers for Coastal Ocean Science
Center for Coastal Fisheries and Habitat Research
101 Pivers Island Road
Beaufort, North Carolina 28516

Mark Cohen,
NOAA, Air Resources Laboratory
NCWCP, R/ARL, Rm. 4204
5830 University Research Court
College Park, Maryland 20740

Chad Hammerschmidt
Department of Earth & Environmental Sciences
Wright State University
3640 Colonel Glenn Highway
Dayton, OH 45435

William Landing
Department of Oceanography
Florida State University
Tallahassee, Florida 32306-4320

Darren Rumbold
Coastal Watershed Institute
Florida Gulf Coast University
Ft. Myers, Florida 33965

James Simons
Center for Coastal Studies
Natural Resources Center, Suite 3200
Texas A&M University-Corpus Christi
6300 Ocean Dr.
Corpus Christi, TX 78412

Steven Wolfe
Florida Institute of Oceanography
2902 Abbotsford Way
Tallahassee, FL 32312

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Penny Pritzker
Secretary

National Oceanic and Atmospheric Administration
Kathryn Sullivan
Administrator

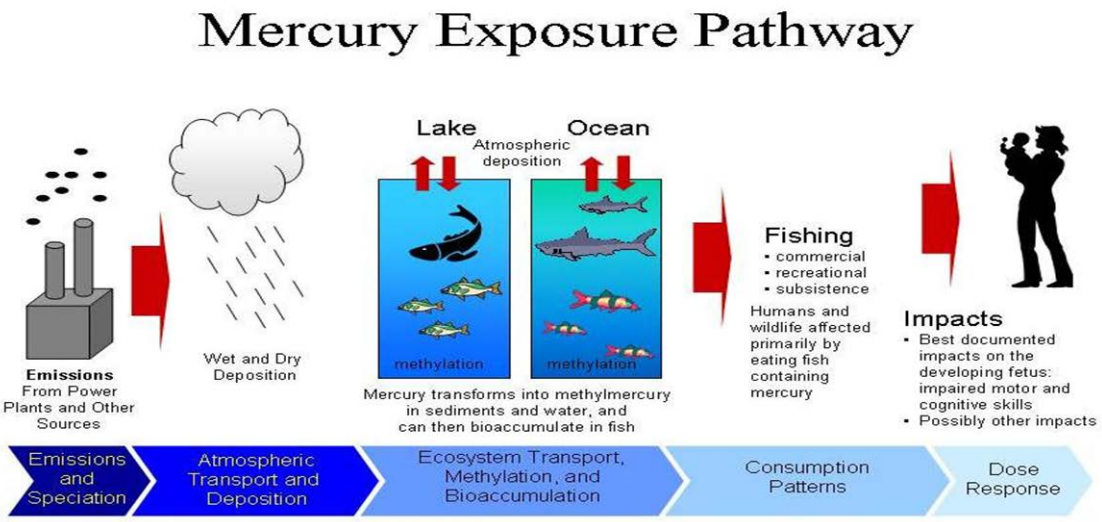
National Ocean Service
Holly Bamford
Assistant Administrator

Executive Summary

Consumption of marine fish is the greatest source of mercury exposure to United States residents. Consumers along the Gulf of Mexico coast are at enhanced risk because of their high levels of seafood consumption and the likelihood that many species of Gulf of Mexico (Gulf) fish have higher levels of mercury than the same species harvested on other coasts. It has been estimated that perhaps 30% of the Gulf coastal population exceeds EPA's reference dose (RfD) for methylmercury, which is used as a criterion to protect human health.

As an outgrowth of this concern, the Gulf of Mexico Alliance (GOMA) established a Mercury Workgroup within its Water Quality Team to develop an Action Plan. GOMA provides a unique partnership of federal, state, and industrial scientists and managers bringing together expertise seeking to reduce the risk of methylmercury to consumers in the Gulf of Mexico region through the application of rigorous and quantitative scientific methods.

GOMA's Mercury Action Plan is organized around the flow path of mercury from its anthropogenic and natural sources, through the environment, its biogeochemical transformation to methylmercury, and methylmercury's bioaccumulation in consumable seafood as shown in the diagram below, modified from U.S. Environmental Protection Agency (EPA).



Our approach begins with the premise that detailed scientific knowledge about the environmental behavior of mercury can be employed to design more effective management and mitigation strategies. A critical element is the need to develop models to assimilate our knowledge of mercury's behavior in the Gulf of Mexico and develop a predictive capability that can be applied to targeted and cost-effective efforts at mitigation.

To this end, we have developed a whitepaper that broadly outlines our current knowledge of mercury in the Gulf of Mexico. In addition, we make recommendations of research needs and

approaches that, if undertaken, would provide coastal managers with the ability to better ameliorate the toxicological risks of mercury to residents of the Gulf of Mexico, and help fulfill mandated requirements to improve impaired water bodies which EPA and the states typically engage through the Total Maximum Daily Load (TMDL) process.

Recommendations for scientific research to achieve these goals include:

- **Identification of at Risk Groups**
  - Updated Gulf-wide surveys of the fish consumption patterns of Gulf residents are needed. Demographic information beyond sex, age, and ethnic/racial group should be included to help identify the highest risk groups. This is the opportunity to include recreational anglers, subsistent fishers, and sub-groups such as different national origins or local communities, groups which can then be identified for special outreach.
- **Identification of Fish Species and Locales with High Mercury Concentrations**
  - The existence of geographic patterns in mercury concentrations within individual species, in the species preferences among consumers, and in the generally local nature of recreational fishing efforts means that more geographically focused monitoring of mercury in seafood species is needed if we are to protect the highest risk and most potentially exposed consumers. Neither the states nor the federal government currently conduct such extensive monitoring programs. Monitoring should be supplemented by research to predict locales with elevated mercury concentrations in seafood species.
- **Identification and Quantification of Locales where Methylmercury Enters the Food Web and Processes Leading to Mercury Biomagnifications by Seafood Species**
  - Field measurements of methylmercury concentrations in water, phytoplankton, and lower trophic level animals in the Gulf of Mexico are largely absent and critically needed. Quantitative predictive models of methylmercury bioaccumulation in Gulf of Mexico food webs depends on this information.
  - Region-specific methylmercury bioaccumulation models need to be developed and the models tested against their ability to accurately predict methylmercury concentrations in seafood species.
- **Identification of Locales where Methylmercury is Produced from Inorganic Mercury in the Gulf of Mexico**
  - Inorganic mercury and methylmercury measurements are needed in the waters of much of the coastal and offshore habitats of the Gulf. Without these measurements, we cannot bring closure to mass balance efforts tying mercury sources with mercury exposure to food web organisms.
  - Measurements of methylmercury and total mercury are needed from sediments throughout the Gulf in order to use these data as surrogates for mercury methylation rates. Measurements within the Gulf's major estuaries (especially in relation to seafood production) and coastal shelf areas are of particular concern as these are areas where methylmercury produced in sediments can most easily reach biota in surface waters.

    - Research should seek to understand the underlying conditions that favor methylmercury production in the Gulf in order to develop a predictive capability based on sediment and water column attributes that are already known.
- **Quantification of Inorganic Mercury and Methylmercury Inputs to the Gulf, its Estuaries, and Open Waters via Atmospheric Deposition, Watershed Deliveries, and Oceanic Deliveries from the Atlantic Ocean**
    - There are relatively few measurements of total and methylmercury concentrations in rivers entering the Gulf. New data is needed to remedy this deficiency.
    - An additional critical need is to quantify the changes in total mercury and methylmercury concentrations within estuaries because of addition and loss processes.
    - Both wet and dry atmospheric deposition of mercury needs to be measured over coastal lands and offshore Gulf waters.
    - Models of wet and dry deposition should be developed and tested against measured values.
- **Predicting and Measuring the Relationships between Mercury Inputs to the Gulf and Local, Regional, National, and Global Emission Sources**
    - Further application of models is needed to permit mercury source-attribution. Model intercomparisons of source-attribution results should be used to assess model predictions and should include sensitivity analyses to estimate the uncertainty in these estimates.
- **Mitigation**
    - Mitigation approaches will need to recognize the complexity of the mercury pathways from mercury sources to human exposure. They will also need to incorporate the spatial, temporal, and ecological variability of mercury concentrations among water, sediments, and biota within the Gulf of Mexico and the demographic variability among consumers.
    - Mitigation will need to be implemented at the appropriate spatial scale and implemented over a time scale adequate to achieve the desired results. Each of the four approaches (source reduction, consumption advisories, landscape modification, and fisheries management) can be appropriate for a specific situation in the Gulf. This will require an integrated strategy based on better scientific understanding.

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INTRODUCTION

There are significant health benefits from eating seafood, but consumption of marine fish is also the greatest source of mercury exposure to United States residents. Consumers along the Gulf of Mexico coast are at enhanced risk because of their higher than average consumption of marine fish and the likelihood that many species of Gulf of Mexico fish have higher levels than the same species harvested on other coasts (Mahaffey *et al.*, 2009; Lincoln *et al*., 2011). The toxicological risks of mercury exposure are now well known (for instance, neurological and cardiovascular disease) and widely documented (National Research Council, 2000; Mergler *et al.*, 2007).

In 2004, the Executive Office of the President, National Science and Technology Council (NSTC, 2004) issued *Methylmercury in the Gulf of Mexico: State of Knowledge and Research Needs.* A major goal was to "Identify data and information gaps that can be addressed by Federal agencies, working with state and industrial stakeholders and partners." The Gulf of Mexico region was chosen "as a prototype for other regional, national, and topical studies."

Reflecting this concern, the Gulf of Mexico Alliance (GOMA) established a Mercury Workgroup within its Water Quality Team to develop an Action Plan to implement and expand upon the recommendations of the NSTC. GOMA provides a unique partnership of federal, state, and industry scientists and managers bringing together expertise that seeks to reduce the risk of methylmercury to consumers in the Gulf of Mexico through rigorous and quantitative scientific methods.

GOMA's Action Plan is organized around the flow path of mercury from its anthropogenic and natural sources, through the environment, its biogeochemical transformation to methylmercury, and methylmercury's bioaccumulation in consumable seafood (Figure 1).

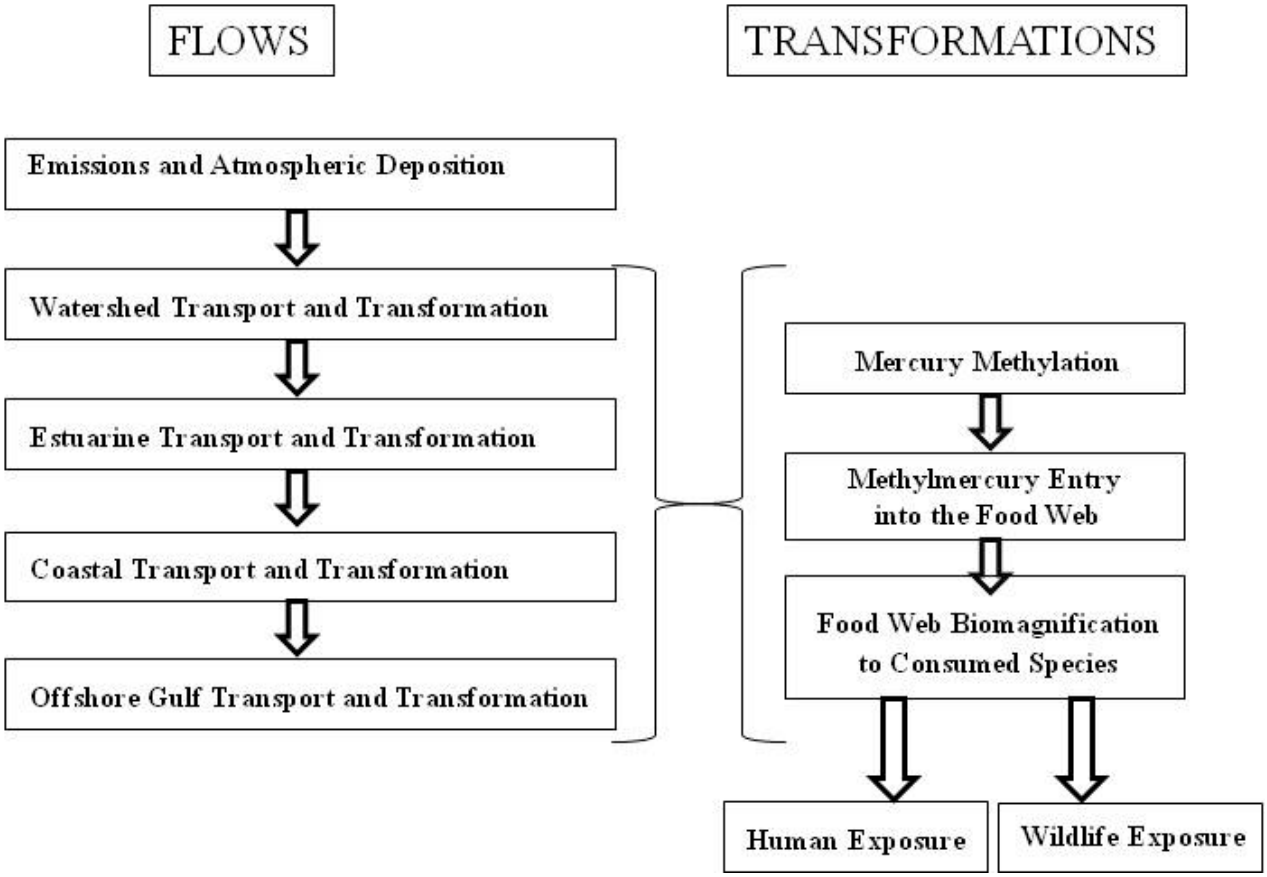
This white paper provides background and rationale for the priorities laid out in the Action Plan and summarizes our current understanding of mercury in the Gulf of Mexico ecosystem.

In addition, the Action Plan emphasizes the identification of special at-risk consumers who ingest larger than average amounts of Gulf of Mexico fish, especially fish with higher than average mercury concentrations. For this reason, we have structured this white paper starting with mercury exposure to humans and proceeding along the mercury flow path backwards toward mercury sources and emissions to the environment. By so doing, the major GOMA goal of mitigating the risks of mercury exposure is emphasized. This flow path approach also helps to clarify where the flow of mercury might be disrupted to reduce the risk to consumers. We need to identify the fisheries where mercury poses the greatest risk to humans (and wildlife), determine the sources of the mercury, and understand where it is being methylated.

The following sections will address the linkages in the mercury flow path. In each section, we will summarize what we currently know about mercury in the Gulf of Mexico, identify the critical gaps in our knowledge, identify the necessary quantifiable links to both the upstream and downstream elements of the overall flow path, and address our progress since the NSTC report of 2004 (see sidebar on page 3).

This white paper is structured to follow the above NSTC organization, but expands the topic **Cycling, Fate, and Chemical Forms of Mercury** into three parts: 1) Identification of locales where methylmercury enters the food web and processes leading to mercury biomagnification by seafood species,

Figure 1. Flow path of mercury through the environment from sources to seafood, including some of the important transformations.



2) Identification of locales where methylmercury is produced from inorganic mercury in the Gulf of Mexico, and 3) Quantification of inorganic mercury and methylmercury inputs to the Gulf, its estuaries and pelagic waters via atmospheric deposition, watershed deliveries, and oceanic deliveries from the Atlantic Ocean. A critical part of these three sections is the need to develop models to synthesize our knowledge of mercury's behavior in the Gulf of Mexico in order to develop a predictive capability that can be applied to targeted and cost-effective efforts at mitigation.

Spatially-explicit process modeling is necessary in the Gulf of Mexico rather than just assuming that broad region wide proportional reduction will suffice to protect the populace, as has been done in other efforts (*e.g.*, total maximum daily load programs of Minnesota and the Northeast Region) This is due to the Gulf's complexity (*e.g.*, geographical variation in methylation potential, *etc*.) and multiple source pathways (*e.g.*, direct deposition, terrestrial runoff, inflow through the Yucatan Channel, *etc*.). This will allow us to identify variations in the attribution of mercury sources, and enable regionally-specific load reductions to reduce mercury bioaccumulation in fish and reduce human and wildlife exposure. Thus, it will be possible to design more effective and cost efficient mercury management and mitigation efforts.

SECTION 1. Identification of At-Risk Groups

More than 90% of the mercury exposure to the American population is from the consumption of marine fish and shellfish. Embryos and young children are particularly sensitive to methylmercury exposure because of their developing brains and nervous systems. As a result, pregnant women and young children have been identified as special at-risk groups.

Beyond sensitivity, risk depends on exposure, which is the product of ingestion rate and the concentration of methylmercury in the consumed seafood. In the coastal Gulf of Mexico, it is possible to identify demographic groups with above average consumption of seafood that also consume seafood species with above average methylmercury concentrations. In concert with source-reduction efforts, targeting high exposure groups with outreach and mitigation efforts provides an important way to reduce mercury's health impacts.

Identifiable groups with higher than average consumption of seafood include specific ethnic, cultural, occupational, and socioeconomic populations, including subsistence fishers. The largest group consuming elevated quantities of Gulf derived seafood is probably recreational anglers. The National Marine Fisheries Service estimates that there are more than two million recreational anglers along the coasts of the four Gulf states exclusive of Texas. A similar number of Gulf recreational anglers are resident in non-coastal counties, non-Gulf states, and foreign countries.

Sunderland *et al*. (2012) recently completed a report for GOMA through Florida's Department of Environmental Protection titled *Pilot Analysis of Gulf of Mexico State Residents' Methylmercury Exposures from Commercial and Locally Caught Fish*. The report asked an overarching question: "What proportion of mercury exposure in various demographic (ethnic) groups of Gulf state residents will be affected by reductions in mercury levels in Gulf of Mexico fish and shellfish?"

National Science and Technology Council Methylmercury Strategy 2004

- **Risk Characterization of Methylmercury Exposure**
  Identify at-risk sub-populations including diet and biopsy samples to characterize mercury exposure
  Characterize at-risk sub-populations in terms of fish consumption, methylmercury intake, blood or hair mercury concentrations and potential health impacts
- **Concentration and Regional Distribution of Mercury in Fish**
  Systematic monitoring program to measure bioaccumulation in fish and shellfish
  Consistent mercury concentration data in biota across species, regions, and habitats of the Gulf of Mexico
- **Cycling, Fate, and Chemical Form of Mercury**
  Atmospheric pathway emission sources research
  Research on methylation mechanisms in sediments, water column, and wetlands
  Fate and transport models of mercury cycling
  Chemical speciation of mercury in environmental media in various Gulf of Mexico regions
  Research on how methylmercury enters the food web
- **Sources of Mercury**
  Systematic assessment of historic and current sources of mercury emissions in Gulf of Mexico region
  Systematic monitoring program of atmospheric deposition on land and water in the Gulf of Mexico region
  Systematic monitoring program measuring mercury concentrations and chemical forms entering Gulf of Mexico in rivers and streams
  Identify natural sources of mercury in the Gulf of Mexico region
  Atmospheric modeling to estimate amounts and source-receptor relationships for the Gulf of Mexico region
  Improved sampling and analytical techniques for mercury speciation in environmental media
- **Risk Management and Mitigation**
  Evaluate success of current mercury education programs and advisories
  Develop a more effective education and outreach strategy

Reviewing and assessing 13 seafood consumption surveys over the past two decades, the authors came to several important conclusions.

- More than 90% of Gulf residents consume seafood.
- Florida residents (and by extension, residents in the coastal counties of the other Gulf states) consume nearly three times the seafood per person as the national average.
- For most groups, much of the seafood consumed is purchased and includes such items as canned tuna, pollock, and salmon caught outside the region. However, between 17% and 59% of the mercury consumed in seafood across the age, gender, and ethnic demographic groups is from Gulf of Mexico sources.
- Recreational anglers have the greatest percentage of their methylmercury exposure derived from Gulf of Mexico fisheries (estimated to range from 36% to 59%).
- Among all demographic groups, perhaps 30% of the coastal population is estimated to exceed EPA's reference dose (RfD) for methylmercury, which is used as a criterion to protect human health.

Of special value in the Sunderland *et al*. (2012) study is the effort to move away from broad assessments of average methylmercury exposures. Model simulations estimated exposures among demographic groups, but also among different theoretical statistical quantiles of each demographic group. From such simulations the percentage of a population that exceeds EPA's reference dose (RfD) for methylmercury can be estimated. These higher exposure, higher risk groups can be targeted for outreach and intervention to reduce their methylmercury exposure via seafood consumption. This approach can be extended to assess the species of fish contributing most to the methylmercury exposure of these higher risk sub-groups.

For the large population of Gulf coast recreational anglers, the quantity of fish harvested is available by species from data of the National Marine Fisheries Service (NMFS, 2010). Combining these data with the median mercury concentrations aggregated from a number of surveys allows us to identify those species contributing most to the potential mercury input to recreational anglers and their families. Table 1 summarizes the harvest, mercury concentrations (almost entirely as methylmercury), and potential mercury available to Gulf recreational anglers from the 24 most harvested species. The EPA considers only about half of harvested fish weight is available for consumption as muscle tissue. In addition, some of the harvest may be given away.

Red drum, spotted seatrout, and king mackerel introduce the most methylmercury into the diets from Gulf fish to coastal recreational anglers. Recreational anglers, however, do not typically target all of the listed species; king mackerel anglers, for example, fish different habitats with different gear than those fishing for spotted seatrout. Angler specialization in some high mercury species such as amberjack, little tunny, other tunas or mackerel, sharks, or crevalle jacks could lead to high methylmercury exposure among these specialist anglers. Again, targeting these high mercury exposure groups for outreach and designing mercury mitigation strategies to lower their exposure could minimize the most dose-dependent risks of methylmercury exposure.

Research Needs and Approaches

The recent study of Sunderland *et al*. (2012) drew heavily from a Florida-specific study (Degner *et al*., 1994) that is now nearly two decades old. Fish consumption patterns have changed as the population and demographics of Florida have changed. There is a need to update this kind of

Table 1. Annual recreational harvest of seafood species in the Gulf of Mexico ranked by the mercury amounts potentially introduced into the human diet as the product of harvest in metric tons (MT) and median mercury concentration in edible muscle tissue (NMFS, 2010).

Species	Recreational Harvest MT	median Hg µg g^{-1}	kg Hg in Harvest
Red drum	5397	0.47	2.54
Spotted seatrout	6576	0.35	2.30
King mackerel	1507	1.00	1.51
Black drum	1302	0.42	0.55
Spanish mackerel	889	0.52	0.46
Groupers	1215	0.37	0.45
Red snapper	1638	0.26	0.43
Sheepshead	1990	0.19	0.38
Greater amberjack	673	0.56	0.38
Sand seatrout	1084	0.33	0.36
Little tunny/Atlantic bonito	262	0.96	0.25
Other tuna/mackerels	409	0.52	0.21
Sharks	205	0.77	0.16
Pinfish	1290	0.12	0.15
Gray snapper	728	0.20	0.15
Blue runner	509	0.20	0.10
Crevalle jacks	142	0.70	0.10
Bluefish	129	0.62	0.08
Dolphinfish	615	0.13	0.08
Southern flounder	387	0.14	0.05
Mullets	568	0.06	0.03
Herrings	205	0.15	0.03
Vermillion snapper	147	0.16	0.02
Atllantic croaker	231	0.07	0.02

work with new surveys that also include the other Gulf states. The methodological limitations of consumption surveys need to be addressed in designing new surveys to provide consistent and confirmable estimates of the consumption of Gulf of Mexico derived fish among an expanded demographic of consumers. Collection of biomarker information such as hair mercury concentrations is needed to help evaluate the survey responses. Personal interviews are preferred over internet surveys because the interviews have less bias (Lincoln *et al*., 2011).

Demographic information beyond sex, age, and ethnic/racial group should be included to help identify the highest risk groups. This is the opportunity to include subsistent fishers and sub-groups such as different national origins or local communities, which can then be identified for special outreach. For example, among Asian-Americans and Pacific Islanders surveyed from Washington state, Japanese and Vietnamese consumed seafood at more than three times the rate

of members of the Mien, Hmong, and Samoan communities (Sechena *et al*., 2003). There were also differences in the fish species consumed among groups. Such variations are probably widespread (Shilling *et al*., 2010) and speak to the need to include the many sources of variability in fish consumption and associated methylmercury exposure. Subsistence fishers are a demographic group that will require a special effort to contact, as they can have exceptionally high methylmercury ingestion rates (Holloman and Newman, 2012). There is an additional need to move away from mean or median mercury exposure estimates and identify the high quantile (90% or 95%) populations with exposures most likely to have serious detrimental health effects.

SECTION 2. What Fish Species Have High Mercury Concentrations and Where Are They Found?

Fish Harvests in the Gulf of Mexico

Fish from the Gulf of Mexico enter the human diet largely from recreational and commercial harvests. The recreational harvest has been summarized in the prior section. Table 2 shows estimates of the amount of mercury introduced to the commercial market from the 24 most important Gulf species. For most species this is almost entirely methylmercury, although invertebrates and lower trophic level fish such as mullet can contain substantial proportions of non-methylmercury. In Gulf oysters, for example, methylmercury averaged only about a third of the total mercury content (Apeti *et al*., 2012). The total amount of mercury potentially introduced into the food supply from commercial harvest is nearly twice that from the recreational harvest. The commercial harvest, however, is distributed nationally and internationally, and it is dispersed over a much larger population than the recreational harvest. Unlike the recreational harvest, shellfish dominate the mercury load from the commercial harvest, comprising the top three species. Shellfish species are individually generally low in mercury.

The column listing the kilograms (kg) of mercury is a broad, average measure of mercury's entry into the human diet. Concern needs to be directed more specifically to the mercury concentration in individual seafood species because individual recreational anglers and purchasers of commercial seafood and their families can preferentially consume species with high mercury concentrations. For example, anglers or consumers who eat large amounts of king mackerel would experience much higher mercury exposure than those consuming similar amounts of shrimp or mullet.

It is widely observed that most fish species increase their mercury concentrations with increasing age and size. Recreational anglers have some control over the size of the fish they eat. State and federal fishing regulations often restrict the size of fish that can be harvested for conservation purposes. Where there is an upper size limit imposed, the higher mercury concentrations of larger fish would be removed from the human diet. In Florida, for example, recreational catches of red drum are limited to fish from 18 to 27 inches in length; for spotted seatrout the range is 15 to 20 inches. Fish in markets, where sold as steaks or filets, are often of indeterminate size. Consumer selection to limit mercury exposure is therefore restricted.

Natural Variability in Mercury Levels among Fish

While there is always uncertainty surrounding the relative importance of the physical, chemical and biological factors that control the amount of mercury biomagnified within an ecosystem (e.g., loading rates, methylation potential, bioavailability, community structure, etc.), we can be certain there will always be natural variability in methylmercury levels among individuals within

populations. For example, methylmercury concentrations can differ among individual fish due to variations in age, size, diet, metabolic rate, ingestion rate, assimilation and elimination rates, growth rate, health status, or sometimes sex (Monteiro and Lopes, 1990; Simoneau *et al*., 2005; Trudel and Rasmussen, 2006; Adams, 2009). Age can be thought of as a "master variable" for

Table 2. Annual commercial harvest of seafood species in the Gulf of Mexico ranked by the mercury (Mercury) amounts potentially introduced into the human diet as the product of harvest in metric tons (MT) and mean mercury concentration in edible muscle tissue (Cunningham *et al.*, 2003).

Group	Harvest metric tons (MT)	Mercury ppm	kg Mercury
White shrimp	41812	0.085	3.55
Blue crabs	18620	0.181	3.37
Brown shrimp	33587	0.078	2.62
King mackerel	1091	1.111	1.21
Black drum	2080	0.415	0.86
Red grouper	1583	0.334	0.53
Oysters	7203	0.072	0.52
Sharks	555	0.743	0.41
Stone crab claws[1]	2319	0.143	0.33
Red snapper	1478	0.216	0.32
Ladyfish	662	0.479	0.32
Spanish mackerel	575	0.513	0.30
Vermillion snapper	957	0.285	0.27
Striped mullet	3900	0.062	0.24
Amberjack	430	0.549	0.24
Herrings	915	0.148	0.14
Golden tilefish	108	1.044	0.11
Sheepshead	612	0.184	0.11
Yellowtail snapper	681	0.155	0.11
Gag grouper	264	0.351	0.09
Crevalle jacks	134	0.629	0.08
Swordfish	174.1	0.460	0.08
Yellowedge grouper	244.6	0.262	0.06
Yellowfin tuna	302.0	0.196	0.06

[1]Concentration data for stone crab claws from Marc Engel and Doug Adams, personal communication.

these other variables, in particular, metabolic rate, growth rate, and size. Size, in turn, can control the species of prey, especially in gape-limited fish. This is often observed as ontogenetic shifts in diet. Size has also been found to be negatively correlated with methylmercury elimination rate (Trudel and Rasmussen, 1997). Some of these factors affect rates of methylmercury exposure while others affect accumulation rates or simply the net resulting concentrations. Fast growth rates during at least a portion of a fish's life span may lead to "growth dilution" resulting in lower methylmercury concentrations (Braune, 1987; Desta *et al*., 2007; Jenssen *et al*., 2010). Braune (1987), for example, found a 'growth dilution' effect on methylmercury concentrations in 1- and 2-year-old Atlantic herring (*Clupea harengus harengus*), but demonstrated positive

correlations with weight and length in 3 to 5 year-olds. Rapid growth may also play a role in the consistently low levels of mercury found in the short-lived dolphin fish, *Coryphaena hippurus* (Adams, 2009) and has recently been suggested in young tiger sharks, *Galeocerdo cuvier* (Rumbold *et al*., 2014). Alternatively, low metabolic rates and slower growth in deep-dwelling fish, or their prey, may be responsible, in part, for higher mercury concentrations in some long-lived fish species of the deep ocean (Tracey, 1993, *cf.* Monteiro *et al*., 1996).

Clearly, given this natural variability, caution is warranted when making comparisons between different fish populations to avoid spurious conclusions regarding spatial or temporal differences. In coastal and marine ecosystems, overharvesting and selective targeting of large pelagic predators such as tunas has reduced the average size of many species, thereby reducing the average mercury concentration in the remaining fish (Bundy *et al*., 2010). Consequently, before comparisons can be made, procedures are required to normalize the data. While standardization for size, age, or weight is a common practice, differences in growth or metabolic rate are often ignored (for review of how ignoring growth can lead to biased interpretations, see Stafford and Haines, 2001).To improve our ability to assess geographical and temporal trends requires improved coordination among state and federal agencies on what fish (*e.g.*, species, size age, gender, *etc*.) to use for biomonitoring.

Geographic and Habitat Variability in Mercury Concentrations in Seafood

Mercury concentrations in harvested seafood are likely to vary with the geographical location and ecological habitat as well as with size and age. This would result from variations in the delivery of mercury to these habitats due to source variability (atmospheric deposition, watershed delivery, and hydrology) and to heterogeneity in the processes that transport and transform mercury inputs (*e.g.*, partitioning to sediments and mercury methylation). For these reasons, we have a need to measure mercury concentrations in seafood species where they are locally high and to understand the underlying reasons for these patterns if we are to conduct effective mitigation.

The mapped distribution of mercury in spotted seatrout (no size or age adjustment) presented by Ache *et al*. (2000) clearly shows variations within the Gulf of Mexico (Figure 2). In particular, mercury concentrations in this species are lowest in south Louisiana, in the Mississippi and Atchafalaya River deltas, and highest along Florida's west coast. Other estuarine resident species such as red drum, hardhead and gafftopsail catfish, gulf flounder, sheepshead, and oysters show a similar regional pattern of differing mercury concentrations.

Geographic variations in the mercury concentrations in seafood species are likely to be greatest at the between-estuary spatial scale. This is because estuaries are where the majority of the recreational fish harvest takes place and because variations in mercury cycling processes are likely to be greatest between estuaries. Lowery and Garrett (2005) provide clear evidence of this variability.

Some coastal pelagic species such as king and Spanish mackerel and bluefish migrate seasonally. Migration could average out part of their geographic variability in mercury concentrations. Demersal species, such as those of the snapper/grouper complex, migrate less and could reflect regional differences in mercury source and process variability. We do not have evidence, yet, for such geographic differences in mercury concentrations, largely because of the issues in data comparability as described above. We do have a number of databases with which to begin cautiously making such an assessment (*e.g*., Hall *et al*., 1978; Texas Department of Health, 1998; Ache *et al.*, 2000; Adams *et al*., 2003; Cunningham *et al*., 2003; Lowery and Garrett, 2005;

Warner and Savitz, 2006; Cai *et al*., 2007; Louisiana Department of Environmental Quality, 2012).

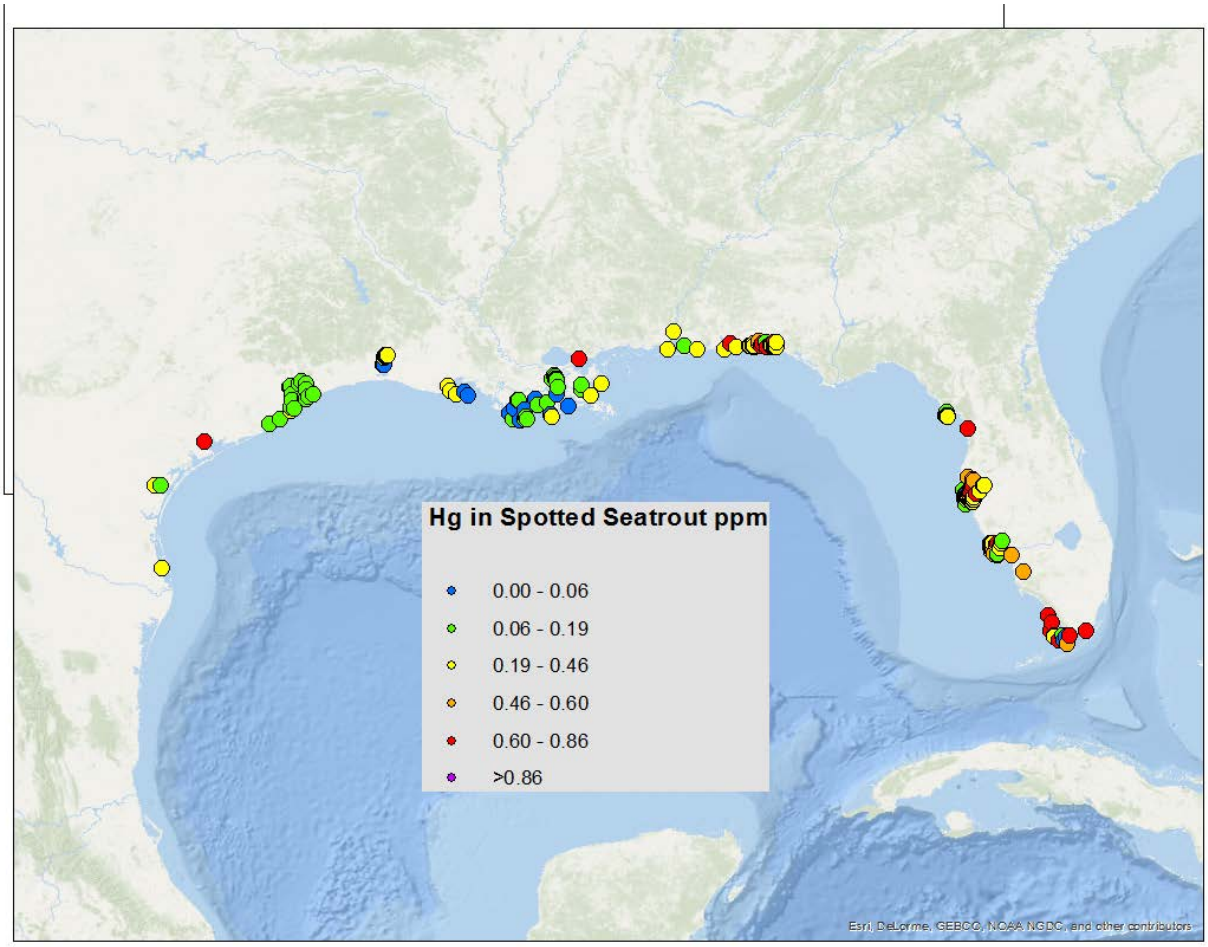


Figure 2. Geographic distribution of mercury concentrations in spotted seatrout (modified from Ache *et al.*, 2000).

Some offshore species such as yellowfin tuna, bluefin tuna, swordfish, blue marlin, and some sharks are highly migratory, some even migrating between the Gulf of Mexico and the Atlantic Ocean. As such, regional differences in mercury concentrations are likely to be smaller still.

Other Wildlife of Concern

Risk from mercury exposure is not just limited to humans as apex predators but also to sharks, marine mammals, and seabirds of the Gulf of Mexico. Regrettably, information on status, trends, and effects of mercury in sharks, marine mammals, and seabirds of the Gulf are scarce and dated.

The first advisory recommending limited consumption of sharks due to mercury was issued jointly by the Florida Department of Health and Rehabilitative Services (FDHRS) and the Florida Department of Agriculture and Consumer Services (FDACS) in Florida in 1991 after FDACS found high levels (ranging from 0.35 µg g^{-1} to 3.9 µg g^{-1}) in shark being sold in retail markets (FDHRS, 1991). A follow-up survey of mercury levels in sharks caught in coastal waters of Florida from 1988 to 1992 found levels as high as 2.87 µg g^{-1} (in the Caribbean reef shark, *Carcharhinus perezii*). Most sharks larger than 200 cm (total length) had levels exceeding 1 µg g^{-1}, the FDA action level at the time (Hueter *et al*., 1995). A survey of mercury reported in

the fishery resources of the Gulf published in 2000, found levels as high as 2 μg g^{-1} in blacktip sharks (*Carcharhinus limbatus*) sampled from 18 sites in the Gulf (Ache *et al*., 2000). Adams *et al*. (2003) reported 5.4 μg g^{-1} in a white shark sampled off Charlotte Harbor. An on-going study has found mercury levels as high as 4.5 μg g^{-1} in blacktip sharks off southwest Florida (Rumbold *et al*., 2014). The study of the effects of mercury on sharks is a nascent field (Nam *et al*., 2011). We do know, however, that observed concentrations exceed critical tissue thresholds suggested for bony fish (Adams *et al*., 2010; for review, see Sandheinrich and Wiener, 2011).

Brown pelicans (*Pelecanus occidentalis*) collected near Tarpon Key and Tampa Bay, Florida in 1969 were found to have "surprisingly high mercury" concentrations, 3 to 5 times higher than pelicans from California (Connors *et al*., 1972). The authors concluded that observed levels, as high as 17 μg g^{-1} (wet weight) in the liver of one bird, were potentially dangerous. Brown pelican eggs collected from Florida Bay in 1972 also had elevated mercury levels, ranging up to 0.65 μg g^{-1}, compared to eggs of ospreys, white ibis, and cormorants collected during the same study (Ogden *et al*., 1974). Pelican eggs collected from Texas from 1975-1981 contained mercury levels ranging from 0.04 to 0.60 μg g^{-1} (King *et al*., 1985). A more recent study found mercury levels in brown pelican eggs (ranging from 0.07-1.67 μg g^{-1} in albumin) elevated compared to eggs of laughing gulls in Mobile Bay, Alabama (Showalter, 2010). Concentrations in a portion of samples from each of these studies approached critical tissues concentrations suggested by Thompson (1996) for interpreting mercury levels in bird eggs. Based on a literature review, he concluded that adverse effects were unlikely to occur in birds at egg concentrations less than 0.5 μg g^{-1}, but that toxic effects were probable at concentrations greater than 2.0 μg g^{-1}. A recent survey of ospreys in Florida Bay found mercury levels in flight feathers to average 16.4 ±1.5 μg g^{-1} in adults and 13.7 ±5.76 μg g^{-1} in juveniles (Lounsbury-Billie *et al*., 2008). The authors report that these levels also approach critical tissue benchmarks for feathers and warrant further research to determine whether these mercury risks are reducing post-fledgling success.

Surveys of mercury levels have been done on Florida manatees (O'Shea *et al*., 1984) and dolphins in the Gulf (Bryan *et al*., 2007, Woshner *et al*., 2008). While the former tend to have relatively low levels (O'Shea, 2003), likely as result of their vegetarian diet, dolphins have been found with elevated mercury levels in a variety of tissues. Woshner *et al*. (2008) reported mercury levels that averaged 0.57 ± 0.43 μg ml^{-1} in blood and 2.15 ± 1.68 μg g^{-1} in the epidermis of dolphins from Sarasota Bay, FL. This study demonstrated associations between mercury concentrations in blood or epidermis and thyroid hormones, liver enzymes, and several hematologic parameters (Woshner *et al*., 2008). A similar study of dolphins along the Eastern coast of Florida and South Carolina, reported a negative correlation between mercury concentrations in blood or skin and total thyroxine, triiodothyronine, absolute numbers of lymphocytes, eosinophils, and platelets and a positive correlation with adrenocorticotropic hormone (ACTH), blood urea nitrogen, and gamma-glutamyl transferase (Schaefer *et al*., 2011). Based on these findings they concluded there was a potential for deleterious effects in highly exposed dolphins.

Human health guidelines do not directly protect the health of the marine ecosystem where mercury containing organisms are the dominant part of the diet of most wildlife. While we know too little about methylmercury's source, we know even less about its impact on marine species, in particular the impact biomagnified methylmercury may have on the other apex predators, such as sharks, dolphins and seabirds.

Research Needs and Approaches

We have a broad knowledge of the mercury concentrations in the most important seafood species in the Gulf of Mexico. This allows us to narrow our focus on what species provide the greatest risk to human and wildlife consumers. The existence of geographic patterns in mercury concentrations within individual species, the species preferences among consumers, and the generally local nature of recreational fishing effort means that we will need more geographically focused monitoring of mercury in seafood species if we are to protect the highest risk and most potentially exposed consumers. Neither the states nor the Federal Government currently conducts such extensive monitoring programs. To do so would be cost prohibitive. A better strategy would be to support the research that would allow us to predict locales with elevated mercury concentrations in seafood species. Part of such a strategy should be to take advantage of the existing historical databases of mercury concentrations in Gulf of Mexico species, with critical information on geographic location. Second, research should be emphasized which explores the habitat attributes and mercury source relationships that lead to locations having increased levels of methylmercury (hotspots) in seafood. For example, wetlands in estuarine watersheds are often associated with biota with high methylmercury concentrations. Sediment characteristics that support mercury methylation, coastal hydrology that limits dilution and flushing of produced methylmercury, and low primary productivity (oligotrophy) also seem to be associated with mercury hotspots. Monitoring of mercury in seafood in locales with a high potential for methylmercury bioaccumulation could confirm these predictions at a modest cost and help direct mitigation efforts which might be prohibitively expensive if applied to all habitats and locales in the Gulf of Mexico.

SECTION 3. Where Does Methylmercury Enter the Food Web and What Processes Lead to Biomagnification by Seafood Species?

Methylmercury is largely acquired by marine animals from their food. Food webs are descriptions linking predators with their prey. Food webs therefore define the pathways by which fish and shellfish acquire their methylmercury, ultimately leading back to the entry of methylmercury at the base of the food web by primary producers. Models exist that simulate this transfer process and can thus predict mercury concentrations in seafood from concentrations of methylmercury in water and sediments. Inorganic mercury can also be acquired through the food web, but unlike methylmercury, it is assimilated less well from food and excreted more rapidly. As a result, methylmercury is preferentially bioaccumulated and represents most of the mercury found in top predator fish. Endothermic birds and marine mammals can demethylate methylmercury, and as a result, methylmercury can be a smaller percentage of the mercury retained in these taxa.

Methylmercury is thought to be accumulated directly from water by phytoplankton and other primary producers. This initial point of entry into the food web becomes a critical target in understanding and predicting methylmercury bioaccumulation. Mason *et al*. (1996) developed empirical and theoretical models of methylmercury uptake in coastal phytoplankton. The high surface area to volume ratio of phytoplankton favors their bioaccumulation of methylmercury in comparison to other primary producers such as seagrasses, marsh grasses, and unrooted macrophytes. Benthic microalgae are also small in size, and their residence on or within surface sediments in shallow waters may allow their exposure to sediment porewater and its potentially high levels of methylmercury produced in these sediments. In the open Gulf of Mexico and most of its coastal waters, phytoplankton are the dominant primary producers supporting apex predators.

Stable Isotopes as Tracers of the Entry of Methylmercury into the Food Web

Carbon, nitrogen, and sulfur are essential components of all life forms, and they too are largely acquired from food. Their stable isotopes provide useful tracers of the origin of these elements (and by inference of methylmercury) in higher organisms of food webs. It is possible to employ these isotopes to infer the primary producers initially fixing C, N, and S (and methylmercury) that ultimately reaches an important seafood species such as king mackerel as has been done in lakes (Bowles et al., 2001; Vander Zanden and Vadeboncoeur, 2002) and coastal waters (Loseto *et al*., 2008).The recent ability to measure natural variability in mercury isotopes adds to the utility of such tracer approaches (Senn *et al*., 2010).

In some estuarine and inshore waters, however, stable isotope signatures of C, N, and S have shown marsh macrophytes or seagrasses or even mangrove trees to be important originators of the nutrients accumulating in higher trophic level biomass (Chanton and Lewis, 2002). In Florida Bay, a gradient of sources of stable isotopes of carbon and nitrogen in fish can be observed, from phytoplankton in near-Gulf waters to the west, through seagrass domination in much of the western and central bay, to a mixture of primary producers in the eastern bay, and finally mangrove and freshwater grasses at the extreme northeastern margins (Evans and Crumley, 2005). In this latter study, mercury concentrations were not strongly associated with any single primary producer source as indicated by stable isotopes, although benthic microalgae were suggested as a potential source of mercury in fish. Table 3 summarizes similar studies around the Gulf.

Table 3. Some locales in the Gulf of Mexico where stable isotope studies have shown primary producers, other than phytoplankton, to be important in supporting the local food web.

Locale	Dominant Primary Producer	Reference
Lower Laguna Madre, Texas	seagrass	Riera *et al*. (2000)
Redfish Bay, Texas	seagrass	Fry and Parker (1979)
Mad Island Marsh, Texas	macrophytes, filamentous algae	Winemiller *et al*. (2007)
Louisiana estuaries	marsh plants	Fry (2008)
Mississippi Sound, Mississippi	seagrass, epiphytic algae	Moncreiff and Sullivan (2001)
Mobile Bay Delta, Alabama	seagrass, terrestrial plants	Goecker *et al*. (2009)
Apalachicola Bay, Florida	terrestrial plants,	Wilson *et al*. (2010)
Big Bend, Florida	seagrass	Nelson *et al*. (2012)
West Florida Shelf, Florida	benthic macrophytes	Burke *et al*. (2004)
Florida Bay, Florida	seagrass, epiphytes	Chasar *et al*. (2005)

The migration of fish between habitats can move carbon, nitrogen, sulfur, and mercury between the habitats. In northeast Florida, gag grouper (*Mycteroperca microlepis*) found in offshore reef areas reflect the nearshore C, N, and S stable isotope signatures of their pinfish (*Lagodon rhomboides*) prey which have migrated offshore (Nelson *et al*., 2012). These stable isotopes have also shown evidence of an inshore mangrove influence persisting in pink shrimp (*Farfantepenaeus duorarum*) that have migrated offshore (Fry *et al*., 1999).

Recently, stable isotope analysis has been used to estimate a food web magnification factor (FWMF, also known as a trophic magnification factor) integrating trophic transfer across all levels of the food webs in coastal waters and estuaries off southwest Florida (Thera and

Rumbold, 2014). Comparisons of FWMFs between different ecosystems may reveal differences in the efficiency with which mercury is transferred resulting from variation in food web dynamics. It can also reveal differences in mercury's basal concentration as it enters the food web as a result of differences in its availability (*i.e.*, due to loading or bioavailability). FWMFs have been reported for mercury in both freshwater and marine food webs (Campbell *et al*., 2005; Al-Reasi *et al*., 2007; Chasar *et al*., 2009; Chumchal and Hambright, 2009; Swanson and Kidd, 2010); however, the majority of these studies took place in temperate or polar systems. It is very likely that trophic transfer efficiency differs between climatically distinct regions due to differences in complexity or length of food webs.

Defining Food Webs from Feeding Habits

Stable isotopes can lead us to the likely source of methylmercury at the base of the food web, but more detailed knowledge of intermediate prey can both confirm conclusions reached this way and add necessary details about the trophic transfer process for methylmercury. This would permit quantitative prediction of methylmercury concentrations from initial concentrations in water and sediments. This detailed information is usually obtained by a quantitative understanding of feeding habits obtained from stomach content analyses of the various members of the food web. There is no single food web in the Gulf of Mexico, and the food web supporting any single species of top predator such as king mackerel or spotted seatrout can differ widely across the varied habitats and regions of the Gulf.

Work is underway to better define the food web structure of key fishes in the Gulf of Mexico. Development of a trophic database, focusing on fishes with high mercury concentrations which are consumed by humans along with food web structure based on these data, is in progress (Simons *et al*., 2013). The initial focal species for this effort are king mackerel and spotted seatrout. Since king mackerel are a coastal pelagic fish, this effort provides the opportunity to integrate both near shore and shelf margin methylmercury through its prey resources. Spotted seatrout is primarily an estuarine species and is very popular among recreational anglers. Because there are spatial differences in deposition of mercury, biological primary and secondary production rates, and species diversity and community structure in the Gulf, the food webs will be spatially distinct (Figure 3).

King mackerel are largely piscivorous, feeding mostly on small to medium sized pelagic fish (Saloman and Naughton, 1983). Frequently consumed prey across the Gulf of Mexico include ballyhoo (*Hemiramphus brasiliensis*), round scad (*Decapterus punctatus*), Spanish sardine (*Sardinella aurita*), Gulf menhaden (*Brevoortia patronus*), and Atlantic bumper (*Chloroscombrus chrysurus*). The species of prey fish seems to depend on what is locally available. Offshore of Louisiana, west of the Mississippi River delta (Web 4), drums of the family *Sciaenidae* such as sand seatrout (*Cynoscion arenarius*), and Atlantic croaker (*Micropogonias undulatus*) are among the dominant prey. Ribbonfish (*Trichurus lepturus*) are important prey for king mackerel along the Texas coast (Web 5). Squid and penaeid shrimp can be important prey in some sub-regions or seasons of year and for different life stages of king mackerel. The mercury concentrations of these many prey species are not well known, but concentration differences are likely to exist among prey species and locations. As a result, mercury bioaccumulation in king mackerel is likely to vary regionally with their local diet despite their extensive migrations.

Modeling, Simulating, and Predicting Methylmercury Bioaccumulation in Food Webs

To predict methylmercury accumulation at the level of primary producers, quantification of methylmercury concentrations in Gulf of Mexico waters is required. These concentrations will depend on concentrations of inorganic mercury, on rates of mercury methylation and demethylation, and on dilution and dispersion of produced methylmercury as described in the next section. In addition, consideration must be given to factors such as partitioning onto sediment particles and binding to dissolved organic carbon, both of which can reduce the bioavailability of methylmercury for uptake (Zhong and Wang, 2009). Thus one might expect that regions with high suspended sediment loads (*e.g.*, Mississippi River outflow) or high dissolved organic carbon concentrations (*e.g.*, estuaries of west Florida) would have lower methylmercury concentrations in phytoplankton that could translate into lower concentrations in higher trophic levels, including top predator fish.

An additional factor is the biomass of the primary producers themselves. As phytoplankton extract methylmercury from the surrounding water, aqueous methylmercury concentrations will decline. At high biomass levels the decline in water column concentrations will translate into a reduction in the steady state concentration of methylmercury in the phytoplankton (Pickhardt *et al.*, 2002). This process, termed biodilution, is well documented in freshwater ecosystems. Thus the high phytoplankton biomass stimulated by Mississippi River efflux off Louisiana or in other eutrophied locales may contain high masses of methylmercury but in lower concentrations that could translate into lower methylmercury concentrations in the food web supported by that phytoplankton.

To predict what these levels might be is a challenging task. Estimates of many rate constants and environmental measurements of mercury speciation are needed, and a model is required to systematically integrate the information to develop quantitative predictive capabilities. Much of this information is not known, or is poorly known. There is generally more data on the concentrations of methylmercury for fishes at higher trophic levels in the food web than there is for many of the invertebrates and forage fish which make up their prey.

There are a number of published food web models for the Gulf of Mexico, several that are unpublished, and several under development (Vidal and Pauley, 2004). The most commonly used software, Ecopath with Ecosim (Christensen and Walters, 2004), uses diet data from all levels of the food web in order to make predictions of the effects of various fishing management strategies on the dynamics of the fish biomass (Vidal and Pauly, 2004). Ecosim, as part of this software, has a module named Ecotracer that can be used to trace the pathways and movement of contaminants such as methylmercury through a food web.

One such Ecopath model examined the West Florida Shelf ecosystem (Okey *et al.*, 2004) which is large in size and supports most of Florida's Gulf of Mexico recreational and commercial fisheries harvests. The model has 51 groups of consumers, including dolphins, seabirds, turtles, and manatees, and includes seagrasses, benthic microalgae, and macroalgae along with phytoplankton as primary producers. It is an ecologically rich and complex ecosystem to model. D. W. Evans (pers. obs.) has explored mercury bioaccumulation in biota of the West Florida Shelf using this model coupled to the Ecotracer sub-module. The model simulations can generally track biomagnification of methylmercury between predators and their prey as well as the general enrichment of methylmercury with increasing trophic level. Of surprise was the importance of benthic microalgae and other benthic primary producers as sources of

methylmercury to consumers. This seems to be an important characteristic of the West Florida Shelf ecosystem. Current efforts are underway to disaggregate functional groups *(e.g.*, the group "large groupers" to individual species; the group "demersal coastal invertebrate feeders" to individual species such as spot (*Leiostomus xanthurus*) and red drum (*Sciaenops ocellatus*), Florida pompano (*Trachinotus carolinus*), red porgy (*Pagrus pagrus*), and hardhead catfish ( )). Some species will be split into age cohorts to better model the differences in mercury concentrations as fish age and grow.

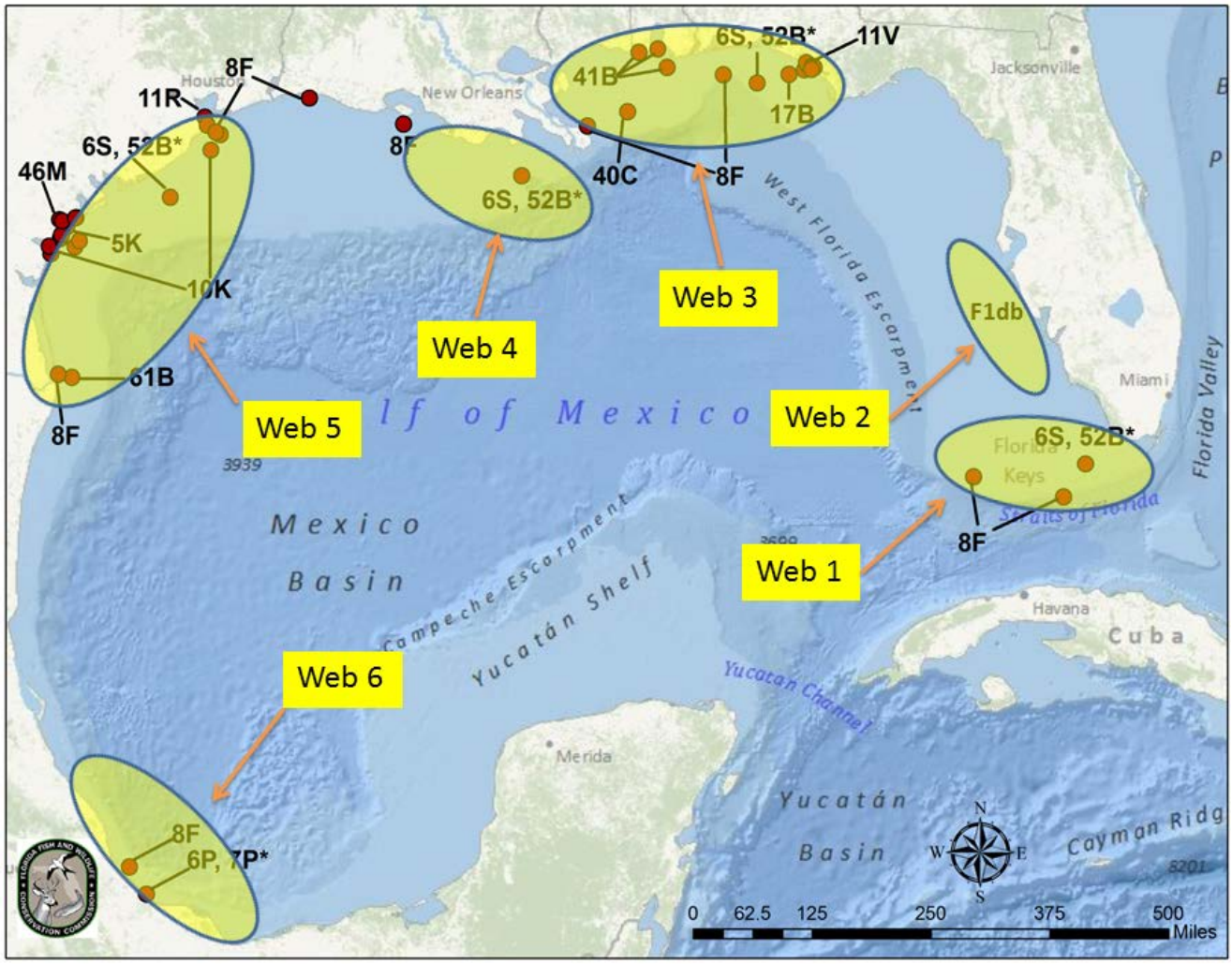


Figure 3. Location of six regionally-distinct food webs for king mackerel based on available data (individual studies indicated by numbered points).

Other food web modeling software includes Ecological Network Analysis (Ulanowicz, 2011) and Atlantis (Fulton *et al*., 2011). In addition, the U.S. Army Corps of Engineers has developed TrophicTrace (Bridges and von Stackelford, 2003), which is an Excel program that can be used to track the movement of contaminants through a food web. There are at present no published examples using any model that traces the movement of methylmercury through a food web in the Gulf of Mexico.

The recent Gulf of Mexico mercury screening model developed a trophic transfer sub-model to simulate methylmercury bioaccumulation in king mackerel as a target species of concern (Pollman *et al*., 2010). King mackerel are known to acquire high methylmercury concentrations.

The screening model postulated that methylmercury partitioned from Gulf of Mexico water into phytoplankton which were consumed by zooplankton and benthic consumers. These were consumed, in turn, by Atlantic thread herring and blue runner. Herring and blue runner became food for king mackerel, transferring much of the methylmercury moving through this series of food web linkages. The model simulations have been most useful in assessing the relative contributions of mercury sources (watershed, atmospheric deposition, and Yucatan Current introduction from the Atlantic Ocean) to the food webs of various regions of the Gulf of Mexico. The model, like all mercury bioaccumulation models in the Gulf, is limited by critical data. It is clear that existing datasets characterizing concentrations of total mercury and methylmercury in the Gulf of Mexico are inadequate and, in some cases, non-existent (Pollman *et al*., 2010).

The screening model and Ecotracer model are still under development, and their ability to predict concentrations of methylmercury in Gulf of Mexico seafood is largely untested. Both models are critically dependent on reliable measurements of methylmercury concentrations in Gulf of Mexico waters. Ongoing development of food habit databases in the Gulf will materially improve these modeling efforts. Ecopath models of some other sub-regions of the Gulf exist, down to the size of Weeks Bay, Alabama and St. Marks National Wildlife Refuge, Florida each of only a few square kilometers extent. These existing models can be used to model methylmercury bioaccumulation there and exploited in developing other sub-regional models.

Research Needs and Approaches

Field measurements of methylmercury concentrations in water, phytoplankton, and lower trophic level animals in the Gulf of Mexico are largely absent and critically needed. Development of quantitative predictive models of methylmercury bioaccumulation in Gulf of Mexico food webs depends on this information. Region-specific methylmercury bioaccumulation models should be developed with this data, and the models tested against their ability to accurately simulate methylmercury concentrations in seafood species using existing or newly acquired methylmercury data in seafood. A regional approach is necessary given the expected heterogeneity in methylmercury production and inputs, concentrations in water, biological productivity, and habitat diversity in the Gulf.

Studies are also needed on the partitioning of inorganic mercury and methylmercury between solution and sediment particles and association with dissolved organic carbon. Bioavailability of methylmercury to phytoplankton (and other primary producers) as a function of this partitioning and association also needs further study.

Quantitative estimates of primary productivity in the Gulf of Mexico by type of primary producer (phytoplankton, benthic microalgae, seagrass, *etc*.) are needed. In particular the relation of primary production to methylmercury concentrations in the environment is needed to estimate aggregate and regional estimates of methylmercury that can enter the food web.

Finally, studies should assess the fluxes of mercury caused by the migration of fish and this biological transport must be included in models.

SECTION 4. Where Is Methylmercury Produced from Inorganic Mercury in the Gulf of Mexico?

Methylmercury can be imported into the Gulf of Mexico and its various habitats from adjacent watersheds, from the atmosphere, and from the neighboring Atlantic Ocean. It can also be produced within the Gulf through the methylation of inorganic mercury. Most mercury

methylation is believed to be mediated by anaerobic microorganisms, namely sulfate- and iron-reducing bacteria, although recent studies suggest that the ability to methylate mercury may be more widespread among microbes. A primary control on methylmercury production in estuarine and marine sediments appears to be the bioavailability of inorganic mercury, which is influenced by both loadings from external sources as well physicochemical factors that affect speciation of the mercury, including dissolved and solid-phase ligands (Hammerschmidt and Fitzgerald, 2004, 2006; Hammerschmidt *et al*., 2004, 2008; Fitzgerald *et al*., 2007; Hollweg *et al*., 2009).

Methylmercury will not be uniformly distributed among the Gulf's diverse habitats because of variations in methylmercury production, proximity to sources, and hydrologic transport processes. As a result, the locales of methylmercury origin or production can differ from locales where primary producers and other food web organisms reside. The transport connection between the two types of locales is critical in determining methylmercury's eventual bioaccumulation in seafood. For example, coastal wetlands can be sources of methylmercury, but phytoplankton in downstream estuarine receiving waters can be the critical recipients and bioaccumulators of this methylmercury rather than wetland biota.

Estuarine Sediments

Methylmercury produced within estuaries has a high potential for bioaccumulation because biological production, especially of harvestable seafood species, is high. In addition, shallow water depths mean that methylmercury produced in bottom sediments can be introduced into a limited volume of water, except where flushing rates are high. Pollman *et al*. (2010) used the concept of hydraulic load (estuarine area divided by freshwater flow into the estuary) to scale potential methylmercury contributions. Many estuaries along the west coast of Florida and estuaries of south Texas have low hydraulic loads which make them most susceptible to methylmercury inputs from sediments. In contrast, high flow estuaries such as the Mississippi River, and Atchafalaya/Vermillion Bays in Louisiana, the Brazos River and Sabine Lake in Texas, Mobile Bay in Alabama, and Apalachicola Bay and the Suwannee River in Florida have hydraulic loads 100 to 1000 times greater. Detecting methylmercury introductions from sediment sources would be difficult in these latter estuaries because of the great dilution from inflowing freshwater.

Potential mercury methylation rates have been estimated in a few Gulf of Mexico estuaries either by radioactive or stable mercury isotope incubations or by patterns of methylmercury concentrations observed in the water column in relation to salinity. Rumbold *et al*. (2011) found potential mercury methylation rates in eastern Florida Bay to range as high as 15% per day with a mean of about 2.5% per day. These potential rates are higher than in the Atlantic coast estuaries reported by Heyes *et al*. (2006).

If methylmercury is released from sediments in large enough quantities, it can be detected as an increase in methylmercury concentrations in overlying waters. Rumbold *et al*. (2011) found elevated methylmercury concentrations in the waters of Florida Bay at intermediate salinities, especially in the transitional mangrove ecotone. This suggested a methylmercury source within the ecotone. Bergamaschi *et al*. (2012) failed to observe mid-estuarine maxima in methylmercury in a similar mangrove dominated estuary (Shark River, Florida), although mangroves seemed to be the source of the observed methylmercury. Conversely, methylmercury was rapidly removed from the water column in Mobile Bay (D. W. Evans, unpublished) with no evidence of methylmercury additions. Han (2004) also reported losses of methylmercury from the waters of Galveston Bay. The most obvious difference between the first two and the latter two systems was the abundance of estuarine wetlands in the former. Although tidal excursions and flooding is

small in most Gulf estuaries, inundation of marginal wetlands by tides has the potential to flush methylmercury from the wetlands into the estuary proper and toward the Gulf of Mexico.

An additional, indirect measure of mercury methylation in estuaries can be observed in the ratio of methylmercury to total mercury in sediments (%MeHg). Large fractions are hypothesized to result from active methylation of sediment-bound inorganic mercury (Heyes *et al*., 2006; Drott *et al*., 2008). The median %MeHg in Mobile Bay sediments is 0.22% (D. W. Evans, unpublished) compared to 1.3% in Florida Bay. This mirrors the pattern in methylmercury in the water column and accumulated in biota. The presence of marsh plants seems to increase the fraction of methylmercury in some estuaries with a presumptive inference of enhanced mercury methylation (Canario *et al*., 2007). At a mercury contaminated site in Lavaca Bay, Texas, marsh and intertidal mudflats had the highest fraction of methylmercury, 0.5% to 1.4% (Bloom *et al*., 1999). Fluxes of methylmercury from these sediments to the water column were greatest in spring before anoxia in the sediments increased during the summer (Gill *et al*., 1999).

Coastal and Open Gulf Sediments

We have found no measurements of potential mercury methylation rates in the Gulf of Mexico proper. The fraction of methylmercury in the sediments is available as a possible surrogate at a number of Gulf locations. Trefry *et al*. (2007) found the fraction of methylmercury ranged from <0.01% to 1.45% with a mean of 0.6% ± 0.32% in shelf and slope sediments off the Louisiana coast. Delaune *et al*. (2008), Liu *et al*. (2009a), and White *et al*. (2009) found similar values. Keach (2006) found especially high fractions of methylmercury in sediments under the Mississippi River plume, in the hypoxic zone. Values ranged from 0.5% to an extraordinary 15.3%. Hypoxic bottom waters showed elevated methylmercury concentrations compared to oxic bottom waters and surface waters, suggesting a sediment methylmercury source. Such measurements have not been reported for the deep water sediments of the open water Gulf. Even if methylmercury is produced in these areas, however, the great depths and vast dilution volumes would minimize methylmercury contributions for bioaccumulation in near surface waters.

Coastal and Open Gulf Water Column

There is increasing evidence that inorganic mercury can be methylated in the water column as well as in sediments. Such evidence has been observed in the Pacific Ocean (Sunderland *et al*., 2009), the Mediterranean Sea (Cossa *et al*., 2009), the Southern Ocean (Cossa *et al*., 2011), and in Arctic waters (Lehnherr *et al*., 2011), although production in deeper water may be less important to its bioaccumulation than methylation in biologically productive surface waters (Hammerschmidt and Bowman, 2012).

With almost no measurements of mercury species in the water column, we are unable to assess the importance of water-column methylation in the Gulf. Keach (2006) found mean methylmercury concentrations in the Gulf of Mexico in the hypoxic zone of the Mississippi River plume of 0.0035 ng L^{-1} in surface waters, 0.0044 ng L^{-1} in oxic bottom waters, and 0.0065 ng L^{-1} in hypoxic bottom waters.

Availability of Inorganic Mercury for Methylation

Inorganic mercury that might be available for methylation can be bound to sediment particles or suspended matter, or in solution, often bound to dissolved organic carbon or other substrates. The current view is that for inorganic mercury to be available for methylation, it must be in solution or easily transferrable from particles to solution (Marvin-DiPasquale *et al*., 2009).

Moreover, to be available for methylation, inorganic mercury must be complexed to anions, such as sulfide or chloride, that form neutrally-charged complexes that can diffuse through the membranes of the microbes that are responsible for the methylation (Benoit *et al*., 2003). Complexation by dissolved organic carbon (DOC), while capable of transferring inorganic mercury from particles to solution, is thought to suppress its methylation (Hammerschmidt *et al*., 2008), although there is contradictory evidence (Graham *et al*., 2012). In addition, high concentrations of dissolved sulfide ion are thought to form charged complexes with inorganic mercury which also impedes methylation by limiting access to methylating microbes (Hollweg *et al.*, 2009).

Inorganic mercury methylation is thought to be most effective at intermediate sulfide ion concentrations in sediment porewaters (Benoit *et al*., 2003; Hollweg *et al*., 2010). Intense anoxia, under this hypothesis, would suppress methylation. Where sediment irrigation is increased by infaunal biota, methylmercury concentrations are observed to increase because higher oxygen fluxes into the sediments increases the area of the active methylation zone (Benoit *et al*., 2009). Bioirrigation also increases the advective flux of methylmercury out of the sediments into the water column.

An additional factor influencing the availability of inorganic mercury for methylation is the time interval since mercury was introduced into the terrestrial or aquatic environment. Isotopic tracer studies have shown that inorganic mercury introduced to soil or sediment containing systems decreases over time, in both the laboratory and the field, in its ability to be methylated due to increased binding to particulates (Hintelmann *et al*., 2000; Orihel *et al*., 2008). This suggests that inorganic mercury atmospherically deposited on the inland watersheds of Gulf estuaries will be less available for methylation than atmospheric mercury deposited on coastal plain estuaries of low slope and on small local watersheds. Atmospheric mercury deposited directly on the waters of the Gulf of Mexico and its estuaries would be most available for methylation because of reduced access to sediments which bind the inorganic mercury. Moreover, the mercury eroded from inland watersheds will include mercury of geological origin that is not easily transferrable to solution where it might be methylated. Marvin-DiPasquale *et al*. (2009) has observed that only a small fraction of sediment associated inorganic mercury is readily available for methylation, usually less than 1%, being greater under oxic conditions. Again, intensely anoxic conditions should decrease methylation of inorganic mercury.

Connecting Methylmercury Production with its Bioaccumulation.

Mercury methylation is but one of the sequence of critical processes by which inorganic mercury is ultimately accumulated in seafood as methylmercury. Other critical processes are the transport of methylmercury, after its production, to primary producers, their bioconcentration of methylmercury, and its subsequent trophic bioaccumulation. Methylmercury is produced in environments generally apart from these primary producers. The anoxic zones of sediments where methylmercury is commonly produced are inhospitable to most primary producers. Methylmercury must diffuse or advect from these sediment zones to the overlying water column in order to reach primary producers which inhabit oxic environments. Processes which impede this diffusion and advection will limit the physical availability of methylmercury to the food web.

In oceanic areas outside the Gulf of Mexico, maximum methylmercury concentrations are usually found at intermediate depths, below the photic zone where phytoplankton grow. The intermediate waters need to be upwelled for methylmercury to be available for bioconcentration by these primary producers. In the Gulf of Mexico, upwelling is episodic and it is associated

with eddies spun off from the Loop Current and with wind induced events in shelf waters. We know little of either the methylmercury distribution in Gulf waters or the possible impact of these episodic events in moving methylmercury into the photic zone.

Estuaries are one area where zones of mercury methylation in sediments reside near the photic zone because of the shallow depth of most Gulf estuaries. There is also evidence that mercury methylation in tidal freshwater and brackish water wetland habitats contributes to the high levels of mercury found in biota in these areas (Evans and Crumley, 2005; Farmer *et al.*, 2010; Fry and Chumchal, 2012).

Other areas where there is a relatively short physical pathway between sediment sources of methylmercury and the photic zone are the coastal shelves. Among these, the West Florida Shelf is a particularly likely area with a short pathway. The West Florida Shelf is the largest shelf area in the Gulf of Mexico, and its waters among the clearest. As a result, benthic primary production can occur over much of its area, and benthic microalgae, seagrasses, and macroalgae contribute significantly to overall primary production (Okey *et al*., 2004). Benthic primary producers are in close proximity to presumptive methylmercury sources in sediments with the likelihood of higher methylmercury exposure concentrations less influenced by dilution or other loss mechanisms. The sediments of the West Florida Shelf are very coarse textured with relatively low organic matter content (NOAA, 2014). Although largely unstudied, the prospect of mercury methylation in the sediments of the West Florida Shelf is plausible. They experience high rates of organic matter processing which is also characteristic of other shelf sediments (Jahnke *et al*., 2005). Equally important, their coarse texture permits high rates of advective flushing of the products of benthic metabolism. If mercury is effectively methylated in these sediments, then the high proportion of benthic primary production is positioned to intercept this methylmercury. The West Florida Shelf is a major region of recreational and commercial fish harvest in the Gulf. The mercury concentrations in several species of seafood are high in this region and can be linked to benthic primary production by stable carbon, nitrogen, and sulfur isotopic analyses. This lends indirect support for the hypothesized importance of mercury methylation in these sediments.

In contrast, the area receiving sediments from the Mississippi River is a net depositional environment for organic rich sediments. Much of the inorganic mercury deposited from the Mississippi River is quickly buried and becomes unavailable for methylation or for efflux to the overlying water column. Moreover, macrofauna that engage in bioirrigation are in low density. For these reasons, the Mississippi and Atchafalaya River deltas and adjacent Gulf shelf are likely to have limited potential for producing methylmercury that ultimately ends up in the food web. Comparing the mercury monitoring results by the states of Louisiana (Louisiana Department of Environmental Quality, 2012) and Florida, (Adams *et al*., 2003) mercury concentrations in many species of fish (especially estuarine and near coastal species) seem to be higher in Florida fish, largely sampled from the West Florida Shelf, than in Louisiana, largely sampled from the Mississippi River Delta and adjacent shelf (see Figure 2). Apeti *et al.* (2012) found a similar difference in total mercury and methylmercury concentrations in oysters sampled in Gulf estuaries.

Research Needs and Approaches

The paucity of measurements of methylmercury in the water and sediments of the Gulf of Mexico and its estuaries critically limits our ability to assess the sources of methylmercury that end up in Gulf of Mexico seafood. A current GOMA sponsored study of mercury in ten Gulf estuaries will help address this deficiency. Sampling transects along the salinity gradients in the estuaries with highest freshwater flows will provide total and methylmercury flux estimates from

their watersheds, deliveries to the Gulf, and estimates of methylmercury production or loss within the estuaries. Unsampled are lower-flow estuaries such as Lower Laguna Madre and other south Texas bays and Apalachee and Tampa Bays in Florida with small hydraulic loadings. They could experience elevated methylmercury concentrations in their waters if the sediments are net methylmercury sources. The consequence would be to expose the resident and transitory biota to concentrations of methylmercury that could significantly elevate mercury concentrations in the seafood harvested therein.

Inorganic mercury and methylmercury measurements are needed in the waters of much of the coastal and offshore habitats of the Gulf. Without these measurements we cannot bring closure to mass balance efforts tying mercury sources to exposure to food web organisms. We cannot employ the existing mercury screening model or its successors to assess where to expect elevated mercury concentrations in seafood. Particularly needed are measurements in the Yucatan Current as it enters the Gulf and in many of the shelf waters of the northern Gulf including the West Florida Shelf, the area adjacent to the Mississippi River debouchment, and the Texas shelf.

Measurements of methylmercury and total mercury are needed from sediments throughout the Gulf in order to use these data as surrogates for mercury methylation rates. Measurements within the Gulf's major estuaries (especially in relation to seafood production) and coastal shelf areas are of particular concern as these are areas where methylmercury produced in sediments can most easily reach biota in surface waters. Isotope measurements of mercury methylation potential should be performed at some of these sites to evaluate the sediment methylmercury and total mercury measurements as surrogates for methylation rates.

Research should attempt to understand the underlying conditions that favor methylmercury production in the Gulf of Mexico in order to develop a predictive capability based on sediment and water column attributes that are already known to a limited extent (*e.g.,* reactive mercury concentrations, redox status, sulfur speciation, grain size, organic matter content, bioirrigation, reoxygenation, sediment deposition rates, sediment mixing rates). Because it would be prohibitively expensive to attempt this everywhere, research sites could be selected based on existing information on mercury concentrations in biota (both high and low level sites) and on the site attributes mentioned above.

SECTION 5: How Much Inorganic Mercury and Methylmercury Enter the Gulf of Mexico, its Estuaries, and Open Waters?

Mercury enters the Gulf of Mexico at its boundaries through runoff from its watersheds, with ocean water from the Atlantic Ocean via the Yucatan Current, and from atmospheric deposition. None of these inputs of mercury is well characterized, and mercury flux estimates from the first two sources are almost non-existent. Pollman *et al.* (2010) made estimates of total mercury and methylmercury fluxes to the Gulf of Mexico on a regional basis as part of a mass balance modeling effort. Although water fluxes from the three main sources of mercury inputs are reasonably well known, the estimates of mercury concentrations in the input waters are largely extrapolated from other areas. Direct measurements of mercury concentrations are distinguished by their absence, and estimates range over one to two orders of magnitude or more. As a result, estimated fluxes of total mercury and methylmercury inputs have a similar uncertainty. Table 4 below summarizes mercury flux and concentration estimates calculated from Pollman *et al.* (2010). Note that more-recent data presented after the table indicates that Yucatan Channel values may be lower.

Yucatan Channel Inputs

The Yucatan Current delivers enormous volumes of water from the Atlantic Ocean via the Caribbean Sea to the Gulf of Mexico. Concentrations of total mercury and methylmercury in the Yucatan Current are unknown and have been extrapolated from the North Atlantic (Table 4). Direct measurements are critically needed. Data from the tropical western Atlantic (total mercury = 0.060 ng/L, methylmercury = 0.008 ng/L) suggest that the flux of total mercury may lower by a factor of 7 and of methylmercury by a factor of 3 (Hammerschmidt, pers. comm.).

Table 4. Estimated total fluxes and presumed concentrations of average total mercury and methylmercury from the three major sources to the Gulf of Mexico (Pollman *et al*., 2010).

Mercury Source	Total Mercury Flux, kg/yr	Methylmercury Flux, kg/yr	Total mercury ng/L	Methylmercury ng/L
Yucatan Channel	160,000	9,200	0.43	0.025
Watershed Inputs	1,100	33	1	0.030
Atmospheric Deposition	44,000	320	22	0.160

Watershed Inputs

Freshwater flowing off the land delivers both inorganic mercury and methylmercury to the Gulf of Mexico. Methylmercury is produced within watersheds and its concentration seems to be positively related to the percentage of woodlands and wetlands in the watershed (Scudder *et al*., 2009). Watershed methylmercury concentrations have been estimated from rivers entering the Gulf of Mexico (Table 5). In the Mobile River watershed in Alabama, Warner *et al*. (2005) found wetlands to favor higher methylmercury concentrations in water. Most dramatically, the mangrove wetlands along Florida's southwest Gulf coast have some extraordinarily high methylmercury concentrations, reaching 26.8 ng/L in one sample (Bergamaschi *et al*., 2012). The wetland dominated coastal plain watersheds of Georgia and Florida have especially high methylmercury concentrations in their drainage waters. Watersheds with most of their runoff coming from low lying coastal plain areas, rich in wetlands, are also important methylmercury sources along the southeastern U.S. Atlantic coast (Guentzel, 2009; Bradley *et al*., 2011).

Table 5. Observed methylmercury concentrations in some rivers draining into the Gulf of Mexico (Pollman *et al*., 2010).

study area	Mean ng/L	Maximum ng/L	Minimum ng/L
Acadian-Pontchartrain Drainages (south Louisiana)	0.165	0.458	0.031
Apalachicola-Chattahoochee-Flint River Basin	0.156	0.733	0.025
Georgia-Florida Coastal Plain	0.934	4.108	0.029
Mobile River Basin (Alabama)	0.066	0.208	0.024
Trinity River Basin (Texas)	0.207	0.300	0.158
Mobile River Basin (Alabama)-Warner	0.225	1.470	0.010
SW Florida-USGS	4.280	26.800	0.150
Shark River	0.160	0.180	0.130

Deliveries of methylmercury from watersheds vary seasonally with runoff flow. Concentrations of methylmercury are typically higher under flood conditions. As a result, most methylmercury should be delivered to the Gulf during high flow seasons. For example, methylmercury concentrations peak during the late winter:spring rainy season in the waters of the lower Mobile River drainage basin (D. W. Evans, unpublished). Maximum methylmercury concentrations in the watershed of eastern Florida Bay are observed during the summer high flow period (Rumbold *et al*., 2011). Most of the flood-derived methylmercury is associated with DOC in watersheds dominated by wetlands and forests; in agriculturally dominated watersheds, most methylmercury is associated with suspended particulate matter because of increased soil erodibility (Babiarz *et al*., 1998).

Watershed loading of mercury to the Gulf of Mexico can be significantly altered due to estuarine processing. High salinity samples from lower Mobile Bay, Alabama, have methylmercury concentrations of 0.02 ng L^{-1} or less and total mercury concentrations were typically less than 0.5 ng L^{-1} in solution, which are much smaller than observed in the Mobile Bay watershed (Warner *et al*., 2005; D. W. Evans, unpublished).

Atmospheric Inputs

Based on deposition measurements from coastal sites, atmospheric deposition is believed to be an important loading pathway for mercury to the Gulf of Mexico although we lack direct measurements of total mercury and methylmercury deposition over Gulf of Mexico waters. Deposition occurs to the open waters of the Gulf, to estuaries, and to Gulf of Mexico watersheds, via precipitation (wet deposition) and surface exchange (dry deposition). Different species and forms of mercury are deposited, including inorganic mercury (*e.g.*, elemental, reactive gaseous, and particulate) and organic mercury (*e.g.*, methylmercury). There can be large spatial and temporal gradients of mercury deposition, for each of the different forms and deposition pathways. For some species or forms and pathways (*e.g.*, surface exchange of elemental mercury), the net flux of mercury may be upwards (*i.e.*, evasion rather than deposition) at some locations and at some times. There are two primary approaches to assess surface fluxes – measurements and modeling. As discussed below, both methods need to be used in combination to provide the most complete understanding.

Atmospheric mercury measurements from the Gulf of Mexico area are summarized in Figure 4 and Table 6. Total mercury wet deposition has been measured at several sites in the region (Figure 4), including several Mercury Deposition Network (MDN) sites and three Pensacola Atmospheric Mercury Study (PAMS) sites (Caffrey *et al*., 2010: Landing *et al*., 2010). Nearly half of the MDN sites in the region have been discontinued (*e.g.*, two sites in Alabama, four sites in Louisiana, and two sites in Florida). Methylmercury wet deposition is currently measured at only one coastal Gulf of Mexico site (MS12). There are few, if any, measurements of wet deposition of mercury over the open waters of the Gulf of Mexico.

Only a small fraction of the mercury deposited in rainfall arrives as methylmercury. Rumbold *et al*. (2011) found only about 0.4% of the total mercury in wet deposition deposited in south Florida was methylmercury. These measurements were made over land. There is evidence that wet deposition over the coastal ocean may contain methylmercury produced within the rainwater (Hammerschmidt *et al*., 2007; Conaway *et al*., 2010). In eastern Florida Bay, wet deposition of methylmercury was much less than that delivered in runoff from the land (Rumbold *et al*., 2011). Dry deposition of mercury is difficult to measure directly. For reactive gaseous and particulate mercury, the flux is generally “down” and dry deposition can be estimated based on measurements of mercury concentration in the atmosphere. Elemental mercury is both deposited

and volatilized, and so in addition to measurements in the atmosphere, measurements of the concentration at the earth's surface must also be made *(e.g.*, in the surface layer of Gulf of Mexico water) in order for the net flux across the surface to be estimated. There are only a few sites in the coastal Gulf of Mexico region at which speciated atmospheric concentrations of mercury are being made – the Grand Bay NERR (MS12) and the Outlying Landing Field (FL96, OLF). Comparable measurements have been made at several sites in Florida, but these have been discontinued. Few speciated concentration measurements have been made over Gulf of Mexico waters, but Florida's Department of Environmental Protection has sponsored a few such measurements on recent research cruises. Dry deposition of reactive gaseous mercury (RGM) and particulate mercury [Mercury(p)] can be estimated from these over-land and over-water measurements. Measurement-based estimates of wet and dry deposition are critically important as they provide quantitative "answers" to the question of how much mercury is being deposited. Of course, these answers are only provided at the locations of the measurements. The extent to which these few measurement sites represent the deposition over the entire Gulf of Mexico region is not well understood. Figure 5 shows the monthly wet deposition of total mercury measured at three relatively nearby sites (MS12, MS22, and FL96). On a monthly basis, there are significant differences in mercury wet deposition. While not shown, the differences on a weekly basis for these three sites are even more dramatic. Figure 6 shows data from all MDN sites in the Gulf of Mexico region for 2008-2010 that have at least 85% data completeness for at least one year during this period. Data for the full year 2011 are not yet available at the time of this writing. There can be significant year-to-year variations at a given site, and significant site-to-site variations in a given year.

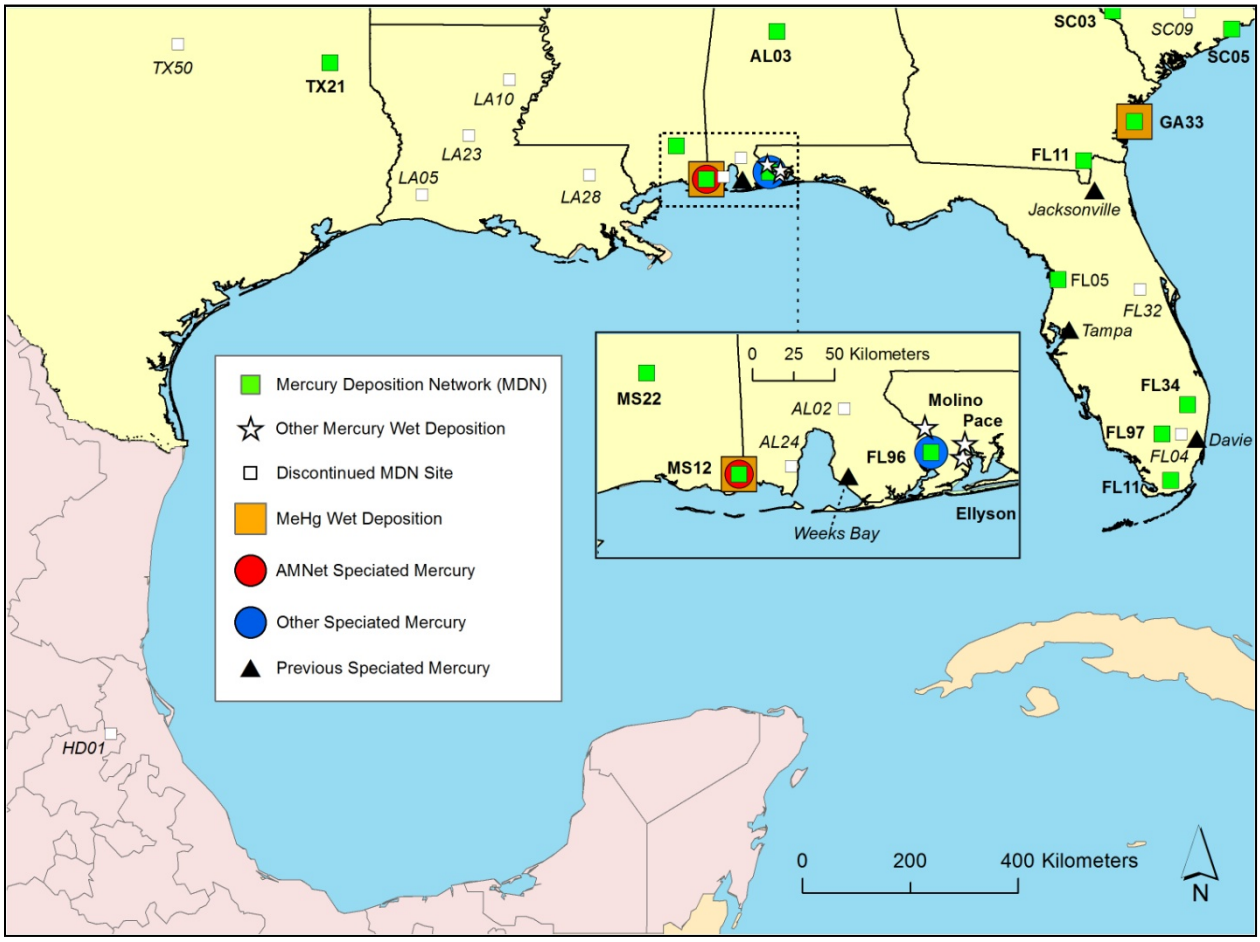


Figure 4. Atmospheric mercury monitoring sites in the Gulf of Mexico region.

Table 6. Summary of Gulf of Mexico-related mercury deposition measurements.

	Wet deposition		Dry deposition	
	Watersheds & Estuaries	**Open Waters**	**Watersheds & Estuaries**	**Open Waters**
Elemental mercury	Probably not very significant	Probably not very significant	Atmospheric conc. measured at a few sites, but net fluxes difficult to estimate because surface concentrations generally not known	Atmospheric conc. measured during recent cruises, but net fluxes difficult to estimate because surface concentrations generally not known
Reactive gaseous mercury			A few sites where RGM is measured, from which dry deposition can be estimated	RGM has been reportedly measured during recent cruises, from which dry deposition can be estimated
Particulate mercury			A few sites where Mercury(P) is measured, from which dry deposition can be estimated	Mercury(p) has been reportedly measured during recent cruises, from which dry deposition can be estimated
Methylmercury	One coastal site where methylmercury is measured in rainfall, and a few additional sites in the extended Gulf of Mexico watershed			
Other organic mercury				
Total mercury	Several regional MDN sites have been discontinued recently, but there are still some MDN sites in the region			

Atmospheric modeling is another approach to estimating Gulf of Mexico-relevant mercury deposition. In this approach, mercury emissions are used as inputs to an atmospheric fate and transport model, and the model outputs the deposition of mercury over its entire domain. There are several models that have been or are being applied to estimate mercury deposition to Gulf of

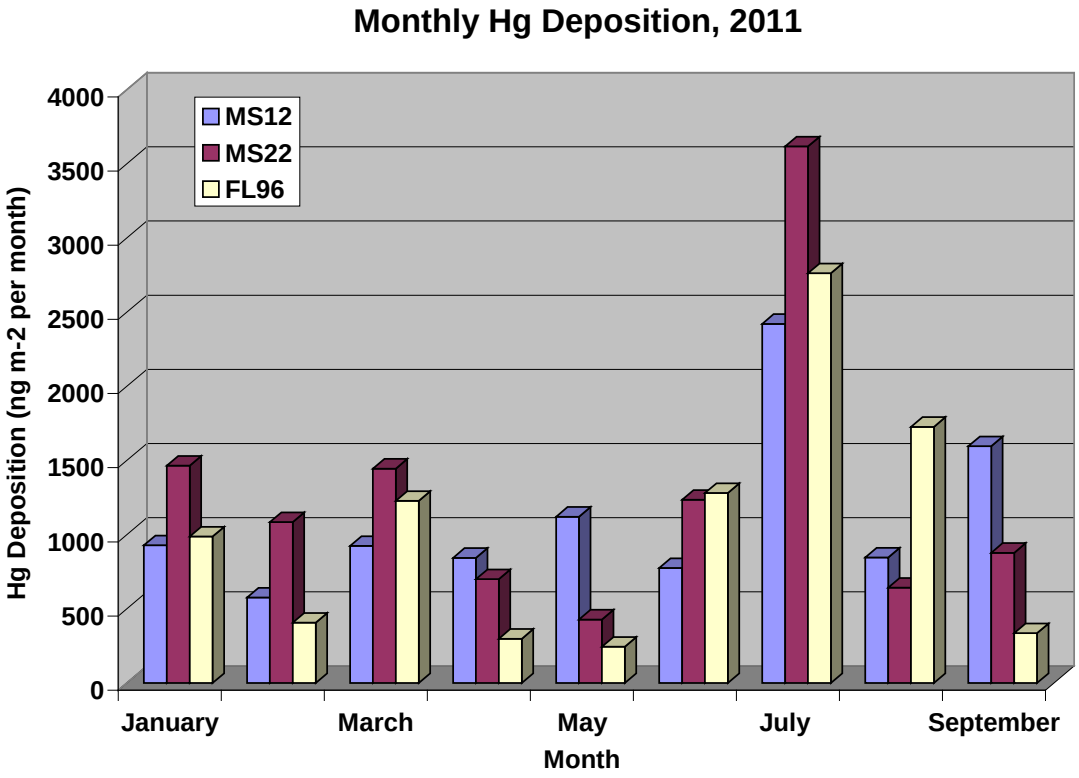


Figure 5. Monthly wet deposition of mercury at three sites near Mobile Bay (MS12, MS22, and FL96) during 2011

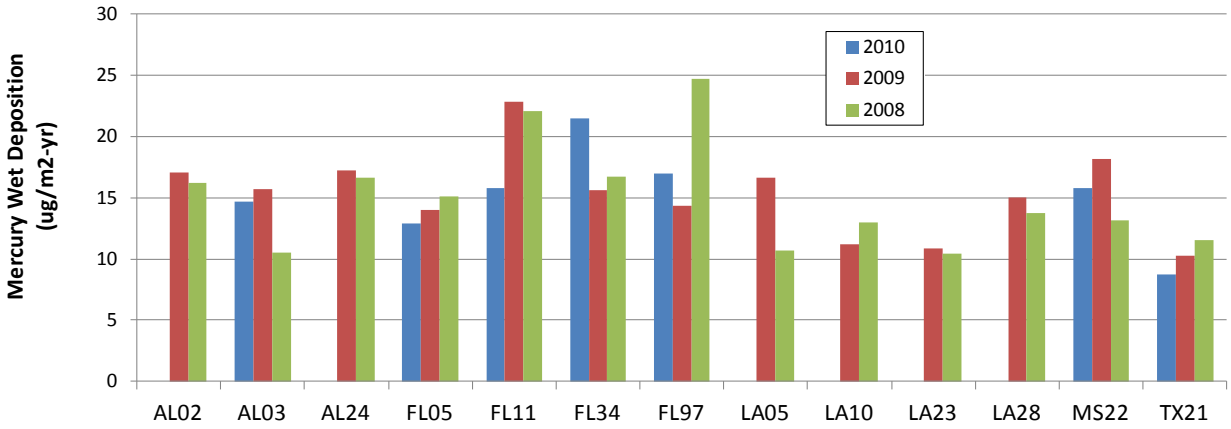


Figure 6. Annual mercury wet deposition for all MDN sites in the Gulf of Mexico region. Values are shown for sites with at least 85% data completeness for a given year.

Mexico waters and its watersheds (*e.g.*, Harris *et al.*, 2012a, 2012b). Advantages of modeling include (a) estimated deposition over the entire region, including spatial and temporal variations; (b) estimates for both dry and wet deposition, and for different mercury forms and species; (c) the possibility of developing detailed source-attribution information (discussed further in Section 6 below). Limitations include uncertainties in model inputs (*e.g.*, mercury emissions and

meteorological data) and atmospheric mercury fate processes. By comparing model results with ambient measurements, the accuracy of the simulations can be estimated.

Neither measurements nor modeling alone can provide accurate estimates of atmospheric mercury deposition to the Gulf of Mexico and its watersheds. Used together, such estimates can be made.

Geographical Distribution of Input Estimates

Although estimated mercury fluxes into the Gulf of Mexico through the Yucatan Channel are significant, this flux enters distant from most of the U.S. and Mexican coasts and can be a minor source of total mercury and methylmercury to coastal waters where biological production and fisheries harvests are greatest (Pollman *et al*., 2010). Moreover, along the coast, higher variability in mercury concentrations would be expected because of seasonal variability in the mercury concentrations in atmospheric and watershed sources and variability in the contributions of the three sources. This variability will require different spatial and temporal scales of monitoring and modeling to capture the source influences.

As might be expected, the mass balance model of Pollman *et al*. (2010) finds the influence of runoff as a source of methylmercury dominates in the coastal waters of Louisiana and Texas where the Mississippi River's influence is strongest. Rice *et al*. (2008) reached a similar conclusion. Atlantic water entering through the Yucatan Channel dominates methylmercury inputs in the central Gulf of Mexico. Atmospheric methylmercury inputs are predicted to be dominant in coastal areas where river flows are small, the West Florida and Yucatan shelves. Predictions of source domination are similar for total mercury, but atmospheric inputs dominate most coastal areas not under the strong influence of the Mississippi River.

There is also the possibility that the availability of inorganic mercury for methylation and the availability of methylmercury for incorporation into the food web will vary with their sources as discussed in Sections 2 and 3. For example, inorganic mercury carried into the Gulf from watersheds is more likely to be bound to sediment particles with reduced availability for methylation. The binding of methylmercury to particles or dissolved organic matter in watershed deliveries could reduce methylmercury bioavailability to primary producers in the food web. By contrast, freshly deposited inorganic mercury and methylmercury from the atmosphere is likely to be more reactive and more readily available to enter the food web.

An additional consideration for methylmercury inputs is *in situ* production, much of which is expected to take place in estuarine and near coastal sediments as discussed in Section 4.

Research Needs and Approaches

Yucatan Channel Inputs

The absence of either total mercury or methylmercury concentration measurements in the Yucatan Channel, at multiple depths, is a critical deficiency that fatally constrains any effort at mercury mass balance and modeling efforts for the Gulf of Mexico. Such measurements are of high priority.

Watershed Inputs

There are relatively few measurements of total and methylmercury concentrations in rivers entering the Gulf. Most serious is the near absence of data from the Mississippi River, source of about 55% of the riverine input of freshwater to the Gulf of Mexico (Solis and Powell, 1999). Pollman *et al.* (2010) summarized many of the published estimates. Inputs from other river

systems of lesser magnitude are also important because they will dominate inputs in individual estuaries with their own food webs which channel mercury to seafood.

An additional critical need is to quantify the changes in total mercury and methylmercury concentrations within estuaries because of addition and loss processes which can substantially alter deliveries to the Gulf of Mexico proper. Both total mercury and methylmercury are substantially lost from the water column of Mobile Bay as water flows toward the Gulf of Mexico (D. W. Evans, unpublished). Such losses have also been observed in Galveston Bay (Han, 2004). These losses reduce deliveries of mercury to the Gulf and thereby reduce the importance of watershed deliveries relative to the other two mercury input sources.

As mentioned earlier, there is a critical need to quantify the availability of inorganic mercury from each of the sources for methylation to methylmercury. In particular, watershed derived mercury may be of legacy origin with a lower reactivity than that from the two other sources.

Aerial Deposition

More extensive mercury wet deposition measurements are needed. Maintainence of funding for existing MDN sites is critical. Discontinued sites need to be restored. Additional MDN sites should be initiated in the Gulf of Mexico region, including over-water sites (*e.g.*, on an oil drilling platform). Additional types of measurements at MDN sites (methylmercury, measurement of different forms/species in precipitation, other heavy metals) would increase our abilities in source attribution and modeling. Event-based wet-deposition measurements as opposed to weekly measurements would also help predictive and interpretive efforts. Development and utilization of models to interpret measurement data would improve our confidence in predictions.

Funding needs to be maintained for existing speciated ambient concentration measurement sites to permit measurement based mercury dry deposition estimates. To make progress towards a routine network of dry deposition measurements, we need co-located alternative dry deposition measurement and estimation methods (*e.g.*, passive samplers vs. surrogate surface vs. micro-meteorological techniques. Elemental mercury concentration measurements in the terrestrial and aquatic ecosystems are needed to allow modeling of the two-way surface flux of this form of mercury. Additional speciated mercury measurement sites, including possible over-water sites are needed; Cruise-based, over-water measurements should be expanded. Model development is needed to help interpret measurement data.

Accurate and frequently updated regional, national, and global mercury emissions inventories should be assembled to provide inputs to mercury deposition models. There is a need to develop and evaluate enhanced meteorological data and model output for driving fate and transport models (*e.g.*, higher resolution; data-assimilation; more actual data over Gulf of Mexico waters). Enhanced research, development and application of atmospheric mercury fate and transport models and their evaluation against measurements is warranted. Multiple models should be applied for the “same” simulation to investigate differences among models and to create more robust, ensemble estimates of deposition. Collaboration should be encouraged with watershed researchers to learn more about the processes and rates of mercury delivery of mercury to the Gulf of Mexico via initial atmospheric deposition to its watersheds.

SECTION 6. Predicting and Measuring the Relationships Between Mercury Inputs to the Gulf of Mexico and Local, Regional, National, and Global Emission Sources

In addition to estimating how much mercury is deposited from the atmosphere, we need to know where it comes from. Such source-attribution information describes the relative importance of different source types and source regions contributing to the atmospheric deposition of mercury to the Gulf of Mexico and its watershed. This information is needed to prioritize actions and develop policies to reduce the atmospheric deposition loading. Moreover, it would allow us to estimate changes to the Gulf of Mexico and its watershed from alternative mercury emission and control scenarios at regional, national, and international scales. Both measurements and modeling approaches can be used to develop source-attribution information.

Time Trends

In this approach, time trends in measurements at a given site are compared with time trends in emissions. Butler *et al*. (2008) found that mercury wet deposition remained relatively constant in the Southeastern U.S. (SE), as a region, over the period 1998-2005. Emissions trends over the same period could not be accurately estimated, but mercury emissions in the region likely declined. In contrast, deposition (and likely, emissions) declined in the Northeast (NE) and Upper Midwest (MW) U.S. The author's speculated that the lack of mercury deposition trends in the SE may be evidence that long-range transport from global emissions regions plays a larger role in this region than in the NE or MW. As Butler *et al*. (2008) found, a major difficulty in using this methodology is that emissions inventories are typically updated only infrequently and with substantial delays. As an example of the limitation, until a version of the 2008 National Emissions Inventory (NEI) was released in April 2012, the EPA's 2005 NEI was the most recent comprehensive U.S. mercury emissions inventory. Global emissions inventories are typically updated even less frequently. Thus, although speciated mercury concentration measurements have been made at FL96 (Pensacola Outlying Landing Field) since 2005 and MS12 (Grand Bay NERR) since 2006, little comprehensive mercury emissions inventory data is available for comparative trend analysis over this period.

Back-Trajectories

In this approach, measurements at a given site are analyzed in conjunction with estimates of the source regions of air parcels arriving at the site, estimated via back-trajectories. An example of this type of analysis is the work of Weiss-Penzias *et al.* (2011), who analyzed measurement data at FL96 (OLF, Pensacola) and at a site in Yorkville, Georgia. Based on a back-trajectory analysis, the authors found evidence to suggest that a significant fraction of mercury dry deposition was derived from sources outside of the local area surrounding FL96.

Receptor-based Multivariate Statistical Modeling

In this approach, measurement data alone are used to develop estimates of source contributions. Examples include chemical mass balance (CMB), positive matrix factorization (PMF), and principal components analysis (PCA).

Based on 3-4 years of continuous rainfall monitoring for mercury and other trace elements in southern Florida, Guentzel *et al.* (2001) concluded that the pronounced seasonal pattern in rainfall mercury deposition, the relatively uniform summertime rainfall mercury concentrations,

and the low concentrations of particulate mercury, indicated that processes other than particulate mercury transport and scavenging govern rainfall mercury deposition in southern Florida. They hypothesized that long-range transport of reactive gaseous mercury (RGM) species coupled with strong convective thunderstorm activity during the summertime represented >50% of the mercury deposition in southern Florida. Model calculations indicated that local anthropogenic particulate mercury and RGM emissions accounted for 30-46% of the summertime rainfall mercury deposition across the southern Florida peninsula.

Landing *et al*. (2010) evaluated ratios of mercury to volatile elements released during coal combustion (*e.g.*, As, Se, excess sulfate) in rainfall samples from the Pensacola Florida region, and concluded that from 22–33% of the rainfall mercury deposition at these sites could be attributed to local and regional coal combustion. These estimates are consistent with REMSAD atmospheric modeling results (using the 2001 EPA mercury emissions inventory) which suggested that 22% of the total atmospheric (wet plus dry) mercury deposition in the Pensacola area was due to local and regional sources, with 78% coming from the global background (D. Atkinson, U.S. EPA, pers. comm.).

These estimates are somewhat lower than, but not inconsistent with, the factor analysis results of Landing *et al*. (2010) where they estimated that 43±10% of the rainfall mercury deposition was associated with the factor believed to reflect emissions from local and regional coal combustion. They concluded that their data supported a conceptual model in which long-range transport and slow oxidation of gaseous elemental mercury are responsible for the majority of the rainfall deposition of mercury in the Pensacola region, and that local and regional coal combustion contribute a smaller fraction. The conclusions from these long-term rainfall monitoring projects in the northern Gulf of Mexico and over the Florida Everglades are quite similar; that long-range transport of oxidized mercury species from the “global background” is the dominant contributor to mercury deposition in both regions. It is reasonable to assume that this same mechanism applies to the open waters of the Gulf of Mexico, especially during the summer months when air masses come in off the open waters of the Caribbean Sea and the Atlantic Ocean and mercury deposition is at its highest.

Isotopic Analysis

Emerging isotope-based mercury measurement techniques are beginning to provide information on source-attribution (*e.g.*, Gratz *et al*., 2010; Sherman *et al*., 2012). Rolison *et al*. (2013) have recently made speciated concentration measurements at two coastal Gulf of Mexico sites and have analyzed the isotopic composition of the different atmospheric mercury forms. It is hoped that the results will shed light on the source(s) of mercury measured at the sites.

Comprehensive Fate and Transport Modeling

In this approach, explicit modeling of the fate and transport of mercury emitted from specific sources is carried out, and the contribution from different sources is tracked numerically. Initial source attribution results for mercury deposition in the Gulf of Mexico region has been generated in this way through the application of several different modeling systems. Seigneur *et al*. (2004) used the CTM-TEAM model to estimate the relative contribution of major source regions (*i.e.*, North America, Asia, and Europe) and natural sources to atmospheric mercury deposition at several sites in the Gulf of Mexico region for the year 1998. North American anthropogenic emissions contributed from approximately 10% to more than 40% at the various sites, with an average of approximately 20%. Asian emissions and natural emissions were each estimated to contribute about 30% to the sites. The REMSAD model has been recently used to estimate

source attribution for deposition in the Gulf of Mexico region (U. S. Environmental Protection Agency, 2008). Results were developed for each state in the region, for several of the major watersheds in the region, and for the northern "half" of the Gulf of Mexico. The application of models to develop source-attribution information for deposition is ongoing. The HYSPLIT-Mercury model is currently being applied to develop such information for Gulf of Mexico, in an analysis comparable to one recently completed for the Great Lakes (Cohen *et al*., 2011).

Indirect Contributions of Atmospheric Deposition

While the quantity and source attribution for atmospheric deposition to watersheds draining into the Gulf of Mexico can be estimated by methods discussed above, the complexities of the subsequent fate and transport of the mercury through the watershed play a major role in determining the ultimate contribution of the deposition to the Gulf of Mexico. At least a portion of the mercury contributed to the Gulf of Mexico by the Yucatan Channel derived is from atmospheric deposition.

Research Needs and Approaches

The research needs and approaches regarding atmospheric processes and deposition described in Section 4 are relevant to the estimation of source-attribution for mercury deposition discussed in this section. The methods to estimate source-attribution rely on the same measurements and modeling used to estimate the quantity of deposition. In addition to these, the following research needs and approaches are noted:

- Further application of isotopic measurement approaches in the region
- Further application of models to estimate source-attribution, including model intercomparisons of source-attribution results – with attendant efforts to characterize and explain differences in model results – and sensitivity analyses to estimate the uncertainty in these estimates.

SECTION 7. Mitigation

Mitigation seeks to reduce the methylmercury exposure to humans. This can be attempted by interrupting the flow of mercury from its sources to its bioaccumulation in seafood, and to the consumption of that seafood. Historically two approaches have been targeted: 1) source reduction and 2) seafood consumption advisories. At least two other points of vulnerability exist in the flow of mercury: 3) altering landscapes and habitats to reduce the methylation of mercury to the form bioavailable to biota, and 4) managing fisheries to limit fish with high mercury concentrations from entering the market or human food web. Each of these approaches can be made more effective and potentially more economical to implement if the pathways of mercury flow are better understood and if patterns of seafood consumption in terms of geographic and species specific mercury concentrations are predictable.

Each strategy would work best if applied at the spatial scale commensurate with the scale of the source spatial variability. For example, if atmospheric deposition of mercury is the major source and it is regionally uniform, and the mercury emission sources are located largely within the region, then restriction of regional emissions can be an effective mitigation goal. If, however, emission sources are global, regional emissions restrictions are likely to be less effective. At smaller spatial scales, such as an estuary, mitigation might seek local removal of existing mercury deposits in sediments, alteration of the potential for mercury methylation by treating or

removing salt marshes as sites of methylation, or targeting consumption of locally contaminated fish through fish consumption advisories tailored to the local fish consuming population. A mercury research program that acknowledges the scale of source, process, and bioaccumulation heterogeneity can help in choosing among possible approaches.

Source Reduction

Emission Controls

A major strategy to limit mercury exposure is EPA's Total Maximum Daily Load (TMDL) assessment. In a TMDL, sources of mercury to the environment of concern are quantified, and proportional or other formulaic reductions mandated to reduce methylmercury concentrations in specific species of fish to a level that is presumed to protect public health. The degree to which the complexity of mercury's behavior in the environment and its geographic variability is employed in TMDL assessments can vary greatly. Many statewide or regional TMDLS largely ignore this complexity and variability. The Minnesota Statewide and the Northeast Regional TMDLs assume that the main source of mercury to the state or region is from atmospheric deposition. The Minnesota TMDL states, *"Ideally, the link between emissions and mercury bioaccumulation in fish would be known quantitatively and the effect of a given reduction in emissions accurately modeled. Such models are under development. In the absence of a validated model that accurately incorporates the complexities of atmospheric chemistry, watershed transport, methylation, and bioaccumulation in fish; we rely on the following rationale (Jackson et al., 2000):*

> *a. A reduction in emissions from sources in a given source area (local, regional or global) results in a proportional reduction in the rate of deposition in Minnesota attributable to those sources. b. A reduction in deposition results in a proportional reduction in mercury loading to water bodies. c. Within a given water body, a proportional reduction in mercury loading in the water results in a proportional reduction in mercury concentrations in fish."*

The Northeast Regional TMDL echoes this: "*The Northeast region's ability to achieve the calculated TMDL allocations is dependent on the adoption and effective implementation of national and international programs to achieve necessary reductions in mercury emissions. Given the magnitude of the reductions required to implement the TMDL, the Northeast cannot reduce in-region sources further to compensate for insufficient reductions from out-of-region sources.*" Moreover: "*If there are differences in sources, loadings, or fish mercury levels across the state, states are encouraged to separate waterbodies into groups according to those differences, i.e., groupings may include waterbodies that are similar in fish mercury levels, source distribution, and other factors such as (but not limited to) land use/land cover, presence of wetlands, drying and re-wetting cycles, water chemistry, and soil type that may affect methylation rates and bioavailability of mercury. Areas with significantly higher mercury levels or local sources may be treated as a separate region with a separate TMDL calculation or excluded from the regional TMDL and a separate TMDL developed. Alternatively, states may include certain waterbodies with higher mercury levels than other areas within the regional TMDL if there is a reasonable site-specific rationale for including such waterbodies.*"
This is certainly the case for such a complex waterbody as the Gulf of Mexico, where varying source distributions, fish mercury levels, land use/cover, wetlands, hydrology, water chemistry, and soil (sediment) types all exist.

Recent comparisons by EPA among four coastal watersheds found that spatial heterogeneity in mercury distributions within each watershed reduced the ability of proportional TMDL calculations to achieve mercury exposure reduction goals (Rothenberg *et al*., 2008). The authors suggested the need for additional site characterizations to identify other predictors of mercury exposure as well as the need to include all sources (loadings) of mercury to the system (*e.g.*, atmospheric deposition)

Clean Air Act Mercury Controls

The U.S. Clean Air Act (CAA) identifies and regulates 188 air toxics, also known as "hazardous air pollutants." Mercury is one of these air toxics. Section 112 of the CAA directs EPA to establish technology-based standards for both new and existing sources within certain source categories that emit these air toxics. Those sources also are required to obtain CAA operating permits and to comply with all applicable emission standards. Two prominent industrial categories, related to mercury emissions, are coal-fired Electric Generating Units (EGU) and cement production. The EPA recently issued regulations and standards to address both industrial categories. The Mercury Air Toxics Standards (MATS) was issued in December 2011 to reduce emissions from EGUs. Legal challenges to MATS are currently on-going and EPA is in the process of formally reconsidering its issued standards for new sources. On August 9, 2010, EPA issued a final rule to limit emissions of mercury and other toxics from Portland cement plants.

United Nations Environmental Program (UNEP) Mercury Program

United Nations Environmental Program (UNEP) has been working to address mercury issues since 2003. Currently, the UNEP mercury program is acting under two main efforts: (1) UNEP Global Mercury Partnership [UNEP Mercury Program Current Partners] and (2) ongoing negotiations. The UNEP Global Mercury Partnership is a consortium of entities that have agreed to undertake cooperative research into mercury's impacts in the environment while at the same time limiting, and where possible eliminating, anthropogenic mercury sources to the environment for the protection of human health and the global environment. UNEP has released a report of global mercury sources inventory for 2010, along with an associated refereed report (United Nations Environmental Program, 2013). Ongoing efforts are to increase the level of participation in the UNEP Global Mercury Partnership.

Consumption Advisories

Fish consumption advisories are employed in all of the Gulf states to steer consumers away from eating fish with higher mercury concentrations, and towards eating healthful fish. Consumption of marine fish is the dominant exposure route for Americans to highly toxic methylmercury. Excessive exposure to methylmercury from fish consumption by pregnant and breastfeeding women may impair neurodevelopment in the fetus and young children. As well, inadequate consumption by pregnant and breastfeeding women of long-chain omega-3 fatty acids, which in our food supply are derived predominantly from eating fish, may also hamper neurodevelopment in the fetus and young children. Fish consumption advisories thus represent a communication challenge. To the extent that consumers of large quantities of fish, especially those fish with higher mercury concentrations, can be identified and targeted for fish consumption guidance, the better the fish consumption advisory approach can work to limit dangerous exposures to individuals.

The Gulf states vary in their approaches to implementing fish consumption advisories for mercury. Women of child-bearing age and young children are universally identified as sensitive risk groups. Specific areas and resident fish species are usually identified along with recommended maximum meal frequencies. The most common means of communicating advisories are through internet sites, news releases, and brochures. Florida has in recent years distributed fish consumption advisory wallet cards, designed by Purdue University with input from Florida agencies, and has plans for another distribution program (100,000 wallet cards). In a study in Louisiana, however, recreational anglers identified television, newspapers and magazines, and mailings as their preferred means of being informed about consumption advisories (Ogunyinka and Lavergne, 2009). These means are rarely used by the states, presumably for reasons of cost. Anglers also suggested including posting advisories at licensing locations and with fishing regulation brochures, as well as at marinas. Some states have done this to a limited extent. It is surprising that information is not made widely available at sites where those who seek to catch fish are most likely to see the advisories. Marine waters are especially underserved in the posting of advisory information.

Better outreach is needed to inform not just the most sensitive groups, but also those groups that could be expected to consume large quantities of marine seafood. Some programs include outreach to commercial fishers, presumed to consume seafood at high levels. In California, it was recognized that certain ethnic, immigrant, and subsistence fishing groups were likely to be large consumers of seafood (Shilling *et al*., 2010), and that multiple languages were needed to communicate risk and consumption advisories adequately. Because of cultural behaviors, it was also recognized that outreach strategies sensitive to these behaviors and preferences are needed (White, 2009).

As with everything involved in the mercury problem in the Gulf of Mexico, there is diversity in the groups targeted by fish consumption advisories. Effective mitigation through consumption advisories will need to be targeted at specific at risk groups and communicated in ways that these groups can understand and respond to. These are likely to involve more respectful communication that acknowledges the recipient's interest and understanding and invites their participation in decision making processes (Shilling, 2009; Shilling *et al*., 2010). It is likely to be costly to actively reach people at an individual level. Some progress is being made at this effort. Florida, for example, employs as many as nine languages in trying to communicate consumption advisories. The state uses many state agencies to communicate mercury information including the Women, Infants, and Children (WIC) program of the Department of Health, the Department of Environmental Protection, and the Fish and Wildlife Conservation Commission. Communication tools include children's books and recipe books as a means to broaden and better communicate advisory recommendations.

Landscape Modification

The terrestrial and aquatic habitats through which mercury flows, from sources to seafood, offer a number of opportunities to intercept mercury and lessen its chance for methylation and bioaccumulation. These opportunities will operate on the scale of individual watersheds, estuaries, or marshes rather than on the scale of the whole Gulf, as international controls on mercury emissions might. At small scales, this may offer advantages in terms of cost and practicality for states and local governments to play a part in mercury mitigation. In California, for example, dams and other detention structures have been employed or proposed to prevent mercury-laden sediments from legacy deposits in the watershed from contaminating downstream habitats under high flow, erosion events. Another example is the removal or treatment of

freshwater marshes or salt marshes to reduce rates of mercury methylation (Windham-Myers *et al*., 2009). In Lavaca Bay, Texas, the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) remediation plan involved digging up contaminated bay sediments, removing contaminated salt marsh areas, and burying existing contaminated sediments with cleaner sediments to remove the former from the zone of methylation.

Recent research has suggested the possibility of treatment of marshes with iron compounds to suppress mercury methylation (Mehrotra and Sedlack, 2005; Liu *et al*., 2009b; Ulrich and Sedlack, 2010). In the 1970s and 1980s, both Sweden and Canada investigated the treatment of mercury contaminated lakes with various chemical agents to reduce mercury bioaccumulation (Rudd *et al*., 1983; Lindquist *et al*., 1991). These treatments included lime, selenium compounds, and phosphate.

A variety of landscape modification activities have been shown to enhance mercury methylation and methylmercury bioaccumulation. These include watershed damming and reservoir formation (Mailman *et al*., 2006), altered drying and wetting of wetlands, and enhanced water and sediment delivery to coastal marshes through river diversion (Fry and Chumchal, 2012). By studying these actions, it might be possible to mitigate their impacts and to reduce methylation in habitats of concern.

Landscape modification in its various forms has been applied with some success at grossly-contaminated sites. However, its use at larger geographic scales or at sites with lower levels of contamination is unlikely to be effective or economically practical.

Fisheries Management

Fisheries managers have a limited ability to reduce the mercury exposure to consumers through seafood consumption. One means is to remove fish with high mercury concentrations from harvest or market. The U.S. Food and Drug Administration (FDA) has the authorization to remove fish from the market, but with the exception of imported swordfish, seldom does so. The current FDA action level of 1 $\mu g\ g^{-1}$ methylmercury in seafood is only used to advise state regulators, and FDA does not directly intervene in the domestic market.

Managers inadvertently control mercury exposures, in some cases, by placing size restrictions on harvested seafood for conservation reasons. For example, all Gulf states ban the commercial harvest of red drum and restrict recreational anglers to fish less than 26 to 28 inches in length, with minimum size restrictions as well. This effectively removes large red drum with the highest mercury concentrations from the human food chain. Commercial red drum harvest from federal waters is forbidden gulf-wide, which further protects larger red drum from harvest. A few other species such as spotted seatrout, black drum, and sheepshead also have maximum size limits. It is unlikely that setting maximum size limits to control mercury exposure will be implemented because of public and commercial opposition and the needs for fishery conservation.

Targeted harvesting of large fish has been tried as a means to reduce mercury concentrations in lakes. Göthberg (1983) reports on the utility of this approach as a means to change both secondary production and trophic biomagnifications of mercury. Such approaches worked in some lakes but not in others. This is an approach applicable only to smaller, generally enclosed, aquatic ecosystems.

Research Needs and Approaches

Mitigation approaches will need to recognize the complexity of the mercury pathways from mercury sources to human exposure. They will also need to incorporate the spatial, temporal, and ecological variability of mercury concentrations among water, sediments, and biota within the Gulf of Mexico and the demographic variability among consumers. A prerequisite to effective and cost conscious mitigation depends on better understanding of mercury's environmental complexity as detailed in earlier sections and on fulfilling many of the recommended research needs and approaches identified in these sections. Mitigation will need to be implemented at the appropriate spatial scale and implemented over a time scale adequate to achieve the desired results. Each of the four approaches to mitigation listed above can be appropriate for a specific situation in the Gulf of Mexico. In deciding which approach to apply, and its likelihood of success, will require an integrated strategy based on better scientific understanding. We are beginning to see more comprehensive and integrated strategies of mitigation. The Delta Tributaries Mercury Council (2002) prepared a strategy based on a TMDL approach, but included a recognition of the need to incorporate environmental variability. It included multiple specific and diverse mitigation recommendations. It could be a model for mitigation efforts in the Gulf of Mexico.

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