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Mercury in the Gulf of Mexico: Sources to receptors ☆

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ABSTRACT

Gulf of Mexico (Gulf) fisheries account for 41% of the U.S. marine recreational fish catch and 16% of the nation's marine commercial fish landings. Mercury (Hg) concentrations are elevated in some fish species in the Gulf, including king mackerel, sharks, and tilefish. All five Gulf states have fish consumption advisories based on Hg. Per-capita fish consumption in the Gulf region is elevated compared to the U.S. national average, and recreational fishers in the region have a potential for greater MeHg exposure due to higher levels of fish consumption. Atmospheric wet Hg deposition is estimated to be higher in the Gulf region compared to most other areas in the U.S., but the largest source of Hg to the Gulf as a whole is the Atlantic Ocean (> 90%) via large flows associated with the Loop Current. Redistribution of atmospheric, Atlantic and terrestrial Hg inputs to the Gulf occurs via large scale water circulation patterns, and further work is needed to refine estimates of the relative importance of these Hg sources in terms of contributing to fish Hg levels in different regions of the Gulf. Measurements are needed to better quantify external loads, in-situ concentrations, and fluxes of total Hg and methylmercury in the water column, sediments, and food web.

1. Introduction

Gulf of Mexico (Gulf) fisheries account for 41% of the U.S. marine recreational fish catch and 16% of the nation's marine commercial fish landings (NOAA, 2011). These fisheries are economically important in the region (NMFS, 2006). While fish consumption has well established health benefits (Mahaffey et al., 2011) there is a global concern regarding exposure to methylmercury (MeHg), a toxic form of mercury (Hg) present in fish. Excess exposure to MeHg can cause neurotoxicological and cardiovascular effects in humans (Mergler et al., 2007). The benefits and risks associated with eating fish have led to confusing messages for the public, compounded by widely varying fish MeHg concentrations (2–3 orders of magnitude) among species, locations and sizes. Should people eat fish, and if so, which fish (see Oken et al., 2012)?

While MeHg occurs naturally and has undoubtedly always been present in fish, industrialization has resulted in higher levels of Hg mobilization and emissions in the biosphere, very likely producing higher fish MeHg concentrations on a global scale (Munthe et al., 2007). As MeHg is a toxin of no known use to organisms, lower MeHg levels in the environment would be beneficial. Efforts are ongoing to reduce Hg releases to the

☆This research has not involved human subjects or experimental animals.

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environment nationally and internationally (e.g. US EPA, 2011a; UNEP, 2011).

Elevated Hg concentrations have been reported in some higher trophic level fish species in the Atlantic and Pacific Oceans (Sunderland, 2007), and in the Gulf, where king mackerel (*Scomberomorus cavalla*) Hg concentrations have been reported up to 4.0 μg g^{-1} wet muscle (Adams and McMichael, 2007; Lowery and Garrett, 2005), and blue marlin (*Makaira nigricans*) concentrations above 10 μg g^{-1} were reported by Cai et al. (2007). The US EPA national Water Quality Criterion for the Protection of Human Health is, by comparison, 0.3 μg Hg g^{-1} wet muscle (US EPA, 2001). All five Gulf States (Texas, Louisiana, Mississippi, Alabama and Florida) have "do not eat" advisories on king mackerel for women of childbearing age and children (US EPA, 2003). Florida lists over 60 Gulf species in its fish consumption advisory regarding Hg (FDOH, 2012). While fish caught in the Gulf are marketed nationally, the Hg issue is of particular relevance to residents in the Gulf region, who eat more fish than the U.S. average (Mahaffey et al., 2009; US EPA, 2002; Degner et al., 1994).

Despite the importance of Gulf fisheries, our understanding of the main sources of Hg to the Gulf and factors controlling MeHg levels in the Gulf food web is inadequate. An improved characterization and understanding of Hg in the Gulf is needed to provide decision makers with a sound scientific basis to assess current risks and the benefits of emissions control strategies. Here we report on the current state of knowledge of Hg sources, cycling, bioaccumulation and human exposure in the Gulf of Mexico. A companion paper (Harris et al., this issue) describes the application of a screening level Hg mass balance model to examine Hg loading, cycling and bioaccumulation in the Gulf.

2. Description of Gulf of Mexico

The Gulf of Mexico has an area of approximately 1.6 million km^2 and a maximum depth of ~4 km (Fig. 1, Table 1). A large continental shelf represents about 30% of the total area. The Gulf is bordered by three countries: the United States (5 states), Mexico, and Cuba. There are 47 major estuaries (UNEP, 2009) and salinity shows significant seasonal variability in the northern Gulf due to seasonal shifts in circulation over the continental shelves and a large seasonal signal in freshwater discharge, primarily from the Mississippi and Atchafalaya Rivers (Morey et al., 2005). The Mississippi River drainage basin (Fig. 1) has an area of 3.2 million km^2, representing nearly two-thirds of the total drainage basin for the Gulf, and includes 41% of the contiguous continental United States area (US EPA, 2010). The largest source of water to the Gulf, however, is the Atlantic Ocean, via the Loop Current, whose flow is approximately three orders of magnitude greater than the water load from the Mississippi drainage basin (Morey et al., 2005). The Loop Current enters the Gulf through the Yucatan Channel and exits through the Straits of Florida. This current and other large-scale water circulation patterns in the Gulf redistribute Hg loads from the atmosphere, Atlantic Ocean, and terrestrial inputs.

Mean annual temperatures at the sea surface are in the range of 26–27 °C (UNEP, 2009). Dissolved organic carbon is on the order of 1 mg L^{-1} in open waters in the Gulf (Baskaran et al., 1996; Guo et al., 1995; Del Castillo et al., 2000) and the pH is ~8 (Solomon et al., 2007). The Gulf is a moderately high productivity system (150–300 g C m^{-2} yr^{-1}) although conditions range from eutrophic in some coastal waters to oligotrophic in deep water areas (UNEP, 2009). A large hypoxic area (~15,000–20,000 km^2) forms in summer in bottom waters over a portion of the northern shelf.

3. Inorganic Hg in the Gulf of Mexico

3.1. Hg sources to the Gulf

Fish Hg concentrations are affected by the rate of Hg loading to a waterbody (Munthe et al., 2007 review). This is clearly evident in studies of point source contamination in freshwater systems (Parks and Hamilton, 1987), coastal and estuarine systems (Herut

Fig. 1. Map of Gulf of Mexico and drainage basin. Drainage from Cuba not available.
Source: Adapted from map created by J.C. Allen, U.S. EPA Gulf of Mexico Program Office.

Table 1
General characteristics of the Gulf of Mexico.

Characteristic	Value	Data source
Surface area (km^2)	$\sim 1.6 \times 10^6$	NCOM model (S. Morey, Florida State University), UNEP (2009)
Drainage basin area (km^2)	$\sim 4.8 \times 10^6$	US EPA (2011b)
Volume (km^3)	$\sim 2.4 \times 10^6$	NCOM model (S. Morey, Florida State University)
Maximum depth (m)	~4000	NCOM outputs (S. Morey, Florida State University)
Mean depth (m)	~1500	Derived from NCOM outputs
pH[a]	8.1	Solomon et al. (2007)
Dissolved organic carbon ($mg\ L^{-1}$)[a]	~1	Baskaran et al. (1996), Guo et al. (1995), Del Castillo et al. (2000)
Suspended solids ($mg\ L^{-1}$)[a]	~2 to > 100	Goni et al. (2006), Carranza-Edwards et al. (1993), Baskaran et al. (1996)
Mean annual surface temperature (C)[a]	26–27	UNEP (2009)
Deep water temperature (C)[a]	5	Loubere et al. (1993)
Biological productivity ($gC\ m^{-2}\ yr^{-1}$)	Moderate (150–300)	UNEP (2009)
Sediment organic content[a]	Coastal: ~1–2%, Pelagic: ~1–2%	Kennicut et al. (1995), Yeager et al. (2004)
Total Hg concentration in water column ($ng\ L^{-1}$)	No data	N/A
Methyl Hg concentration in water column ($ng\ L^{-1}$)	No data	N/A
Total Hg concentration in sediments ($ng\ g^{-1}$)	5–80	Liu et al. (2009), Kannan et al. (1998), Delaune et al. (2008)
Methyl Hg concentrations in sediments ($ng\ g^{-1}$)	~0.02 to 0.3 (< =1% of total Hg)	Liu et al. (2009)

NA=Not applicable.

[a] Coastal shelf and pelagic areas.

et al., 1996; Francesconi et al., 1997), and has been demonstrated for smaller changes in ecosystem Hg loading relevant to changes in atmospheric Hg deposition (Harris et al., 2007). It is therefore critical to quantify the current sources of Hg to the Gulf, which can be broadly grouped as atmospheric, terrestrial and Atlantic inputs. External Hg loads to the Gulf were estimated as part of a mass balance modeling study by Harris et al. (this issue) and are summarized here. Hg inputs to the Gulf were assumed to be small from hydrothermal vents (Mason et al., this issue; Lamborg et al., 2006) and oil and gas exploration rigs (Neff, 2002).

Atlantic Hg inputs are dominated by the Loop Current which enters through the Yucatan Channel with a flow of approximately 27 Sv (1 Sv=$10^6\ m^3\ s^{-1}$). Inflowing Atlantic Hg concentrations were estimated from the literature, as no direct measurements of Hg concentrations in the Loop Current were available. Sunderland and Mason (2007) reported total Hg (THg) concentrations in Atlantic waters (north, south, equatorial) that averaged 0.43 $ng\ L^{-1}$ (n=6). Based on that estimate, the Atlantic input of THg to the Gulf was approximately 240 $\mu g\ m^{-2}\ yr^{-1}$ (Harris et al., this issue).

The riverine Hg load to Gulf coastal waters estimated by Harris et al. (this issue) was ~7 $\mu g\ m^{-2}\ yr^{-1}$, of which more than 90% was associated with the Mississippi and Atchafalaya Rivers. Rice et al. (2008) estimated loads of 6.25 and 3.25 $t\ Hg\ yr^{-1}$ respectively for these rivers, primarily associated with particulates. Dissolved Hg loads from these two rivers were estimated by Rice et al. (2008) using the US EPA's SERAFM model, rather than direct observations (Knightes, 2008). Site data for riverine Hg concentrations and loads to most other regions of the Gulf were not available. Using median stream concentrations of 1.90 $ng\ L^{-1}$ (THg) and 0.11 $ng\ L^{-1}$ (MeHg) from a USGS national survey by Scudder et al. (2009) (basins without mining activities), and allowing for estuarine Hg trapping of a portion of riverine Hg prior to entering coastal waters in the Gulf (~50%, see Harris et al., in this issue)), Hg concentrations assigned for other riverine inputs to the Gulf ranged from 1–3 $ng\ L^{-1}$ for inorganic Hg and 0.03 to 0.10 $ng\ L^{-1}$ for MeHg. River Hg concentrations can vary widely, e.g. Scudder et al. (2009) reported a range of 0.27 to 75 $ng\ L^{-1}$ in streams without mining activities. This highlights the fact that riverine Hg loads to the Gulf are under-constrained due to a lack of data on riverine Hg concentrations and uncertainty regarding the effects of estuaries on Hg delivery to marine waters.

Atmospheric Hg inputs to the Gulf were estimated with a combination of field observations and modeling. Wet deposition observations are available at several land sites in the Gulf region (Table 2), but atmospheric models are needed to estimate wet Hg deposition over Gulf waters, as well as dry deposition throughout the Gulf domain (no systematic monitoring of dry Hg deposition exists). While the mass balance modeling carried out by Harris et al. (this issue) used results from the AMSTERDAM model (the Advanced Modeling System for Transport, Emissions, Reactions and Deposition of Atmospheric Matter) (Vijayaraghavan et al., 2007,2008), here we also include outputs from two additional models: GEOS-Chem (Holmes et al., 2010) and REMSAD (the Regional Modeling System for Aerosols and Deposition) (Atkinson et al., 2008). Details regarding the mechanisms and rates used in these models for inter-conversion between inorganic atmospheric Hg forms and subsequent deposition can be found in those references. Modeled estimates ranged from 7–16 $\mu g\ m^{-2}\ yr^{-1}$ among the models for wet deposition and 14–28 $\mu g\ m^{-2}\ yr^{-1}$ for combined wet and dry THg deposition (Table 2). Dry deposition of Hg ranged from ~25–50% of the total estimated deposition across models but presented less seasonal variability than wet deposition. Additional information on the development of atmospheric Hg deposition estimates is provided in Appendix A.

3.2. Relative importance of inorganic Hg sources

When the Gulf is viewed as a whole, the Loop Current dominates Hg loading (Fig. 2), accounting for 85–90% of the total supply. The whole-Gulf perspective is misleading however, because surface waters and Hg are not fully mixed around the Gulf of Mexico; some areas are not as influenced by the Loop Current as others. In a screening model analysis that divided the Gulf into 19 regions, the relative importance of external Hg sources varied widely among regions (Harris et al., this issue). Each of the three external Hg loads was the largest predicted source to at least one of the model regions. In the central Gulf the Loop Current was the largest predicted source of inorganic Hg. Inputs from the Mississippi and Atchafalaya River were the largest source of Hg in coastal waters in the vicinity of these rivers. Atmospheric deposition was predicted to be the largest source of Hg along the coast of Florida.

Although we examined the Hg contributions from direct deposition, the Atlantic Ocean and rivers, all of these Hg sources to the Gulf typically link back primarily to atmospheric Hg deposition (assuming hydrothermal vents are not an important source of Hg to the Atlantic). Exceptions include terrestrial areas

Table 2
Comparison of modeled mercury deposition to the Gulf of Mexico with measured wet deposition. All fluxes in μg m^{-2} for appropriate time period.

Month	REMSAD			GEOS-Chem				AMSTERDAM			PAMS (3 sites)	MDN AL24	MDN FL05
	Wet	Dry	Total	Wet	Dry (Hg(II))	Dry (Hg^o)	Total	Wet	Dry	Total	(2005–2010) Wet	(2005–2009) Wet	(2005–2010) Wet
January	0.74	0.45	1.19	0.57	0.52	0.27	1.35	0.83	0.75	1.57	1.40	1.10	0.95
February	0.74	0.45	1.19	0.51	0.37	0.20	1.08	0.85	0.99	1.83	0.83	0.93	0.52
March	1.00	0.57	1.57	0.59	0.52	0.25	1.36	0.81	0.96	1.77	0.68	1.77	1.15
April	1.00	0.57	1.57	0.51	0.46	0.18	1.15	0.52	1.04	1.56	1.17	1.81	0.68
May	1.00	0.57	1.57	0.66	0.29	0.15	1.10	0.66	1.52	2.18	1.33	1.74	1.12
June	2.01	0.33	2.34	1.08	0.31	0.10	1.48	1.94	0.93	2.87	1.43	1.30	2.91
July	2.01	0.33	2.34	0.83	0.31	0.09	1.22	1.71	0.82	2.54	1.75	3.50	3.96
August	2.01	0.33	2.34	0.60	0.24	0.07	0.90	2.30	1.01	3.31	3.31	1.76	3.43
September	1.34	0.52	1.86	0.60	0.30	0.12	1.01	2.63	0.83	3.46	1.18	1.65	1.69
October	1.34	0.52	1.86	0.53	0.47	0.15	1.14	1.76	0.77	2.53	0.93	0.88	0.77
November	1.34	0.52	1.86	0.30	0.51	0.16	0.96	0.90	1.03	1.93	0.66	0.69	0.71
December	0.74	0.45	1.19	0.31	0.42	0.23	0.95	1.04	1.00	2.05	0.81	1.44	0.71
Annual	15.26	5.61	20.87	7.08	4.70	1.93	13.71	15.96	11.65	27.62	15.48	18.56	18.60

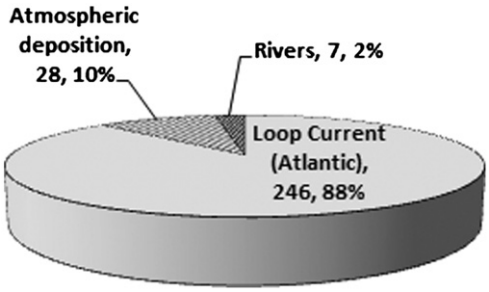

Fig. 2. Estimated Hg inputs to the Gulf of Mexico. From Harris et al. (this issue). First number for each input is in units of μg m^{-2} yr^{-1}. Second number is percent of overall Hg load.

with enriched Hg deposits (Fitzgerald et al., 1997) or where there is point source industrial Hg contamination. Reducing atmospheric Hg deposition is therefore central to reducing overall Hg loading to the Gulf of Mexico. Furthermore, GEOS-Chem and REMSAD simulations estimate that most of the Hg in atmospheric deposition to the Gulf, on the order of 75 percent or more, originates from sources outside the United States. This does not mean that actions are not needed within the United States to reduce Hg emissions; rather it points to a need for a coordinated international effort to reduce Hg deposition to the Gulf of Mexico.

4. MeHg sources in the Gulf of Mexico

Loop Current inputs of MeHg are likely the largest external source of MeHg to the Gulf (Harris et al., this issue), while rates of in-situ methylation, and the primary sites of in-situ MeHg production, are not known. In-situ methylation is a topic of ongoing research in the oceans generally (see Mason et al., this issue). MeHg production in sediments has been widely documented for freshwater and marine aquatic systems (e.g. Hollweg et al., 2009; Sunderland et al., 2006; Hammerschmidt and Fitzgerald, 2004; Gilmour et al., 1992). MeHg production has also been recently been identified in the water column of the Pacific Ocean and Mediterranean Sea where MeHg concentrations peaked in both cases at intermediate depths, in the range of 0.08 ng L^{-1}, while surface concentrations were lower, ~0.01 to 0.02 ng L^{-1} (Sunderland et al., 2009; Cossa et al., 2009, 2011; Heimbürger et al., 2010). The large hypoxic zone in the northern Gulf shelf is also potentially a zone of enhanced water column production of MeHg, as observed in other systems with low oxygen waters (Heimbürger et al., 2010; Sunderland et al., 2009; Cossa et al., 2009; Herrin et al., 1998). There are currently no water column Hg data available to resolve where MeHg is primarily produced in the Gulf.

It is essential to identify the primary sites of Hg methylation in the Gulf, especially those supplying MeHg to key fisheries with elevated MeHg levels, in order to evaluate the effectiveness of different Hg load reduction strategies. If water column methylation in intermediate depth waters is the primary source of in-situ MeHg production (Mason et al., this issue), inorganic Hg sources to these waters, e.g. Loop Current inputs, would be more important to control. Where sediment methylation in coastal zones is more important, terrestrial inputs would take on greater importance in terms of the source of Hg ultimately accumulating in fish.

The timing of the response of fish Hg concentrations to changes in atmospheric Hg deposition is also likely affected by the primary site of methylation. Methylation in zones strongly influenced by terrestrial Hg inputs, such as some coastal sediments, should change at a rate ultimately controlled by the slow decline of terrestrial Hg inputs. Terrestrial Hg export in streams lags changes in deposition, likely on a scale of decades or centuries (Krabbenhoft et al., 2006; Harris et al., 2007; Munthe et al., 2007; Munthe and Hultberg, 2004). Water column methylation in large marine systems appears to peak in the upper waters below the mixed layer (200–700 m) (Cossa et al., 2009,2011; Sunderland et al., 2009). Inorganic Hg concentrations in waters at these depths were predicted by Sunderland and Mason (2007) to respond rapidly initially to reductions in direct atmospheric deposition, but require decades to reach steady state with lateral inputs from other ocean basins. Assuming that MeHg concentrations would respond relatively quickly to changes in MeHg production in these waters, fish Hg levels would similarly show an initial rapid response to changes in atmospheric deposition and a longer secondary phase response. Gulf regions with the potential for relatively rapid changes in MeHg production and concentrations (e.g. years to decadal scale) include locations where methylation in the water column (e.g. anoxic waters) or in a thin layer at the sediment water interface is important and supplied mostly by direct atmospheric Hg deposition. Harris et al. (this issue) predicted that this may be the case for example for some Florida coastal waters.

Table 3
Leading ten species in the Gulf of Mexico recreational and commercial harvests for human consumption (MT=Metric tonnes).

Species	Scientific name	Group	Harvest (MT)	Mean Hg (ppm)[a]	Sample size (*n*)[a]
Recreational harvest (NMFS, 2010)					
Spotted seatrout	*Cynoscion nebulosus*	Estuarine	6,576	0.320	546
Red drum	*Sciaenops ocellatus*	Estuarine/coastal demersal	5,397	0.497	594
Sheepshead	*Archosargus probatocephalus*	Estuarine/coastal demersal	1,990	0.180	226
Red snapper	*Lutjanus campechanus*	Reef fish	1,638	0.093	13
King mackerel	*Scomberomorus cavalla*	Coastal migratory pelagic	1,507	1.085	385
Black drum	*Pogonias cromis*	Estuarine/coastal demersal	1,302	0.443	233
Pinfish	*Lagodon rhomboides*	Estuarine/coastal demersal	1,290	0.131	6
Groupers	family *Serranidae*	Reef fish	1,215	–	–
Sand seatrout	*Cynoscion arenarius*	Estuarine	1,084	0.475	99
Spanish mackerel	*Scomberomorus maculatus*	Coastal migratory pelagic	889	0.527	204
Commercial harvest (NMFS, 2009)					
White Shrimp	*Litopenaeus setiferus*	Estuarine/coastal demersal	44,586	0.024	16
Brown Shrimp	*Farfantepenaeus aztecus*	Estuarine/coastal demersal	35,864	0.033	14
Blue Crabs	*Callinectes sapidus*	Estuarine	22,211	0.141	239
Eastern Oysters	*Crassostrea virginica*	Estuarine	9,363	0.080	1,634
Mullet	family *Mugil* spp.	Estuarine/coastal demersal	4,676	0.063	56
Pink Shrimp	*Farfantepenaeus duorarum*	Estuarine/coastal demersal	3,207	–	–
Stone Crabs	*Menippe mercenaria*	Demersal	2,769	1.360	3
Red Grouper	*Epinephelus morio*	Reef fish	2,531	0.324	44
Black Drum	*Pogonias cromis*	Estuarine/coastal demersal	1,836	0.443	233
Spiny Lobster	*Panulirus argus*	Estuarine/coastal demersal	1,350	–	–

[a] Hg data from US EPA (2003).

5. MeHg bioaccumulation

MeHg concentrations in fish and shellfish in the Gulf region vary depending on species, size, and conditions affecting MeHg supply and delivery through the food web. Table 3 shows the top ten species of fish or shellfish harvested recreationally and commercially for human consumption in the Gulf of Mexico, and associated Hg concentrations. The commercial harvest is much larger than the recreational harvest but is consumed both within and outside of the Gulf region. More than half of the commercial catch occurs within estuaries and in coastal waters within 5 km of shore (NMFS, 2009), dominated by invertebrates (shrimps, crabs, oysters) that are typically low in Hg concentrations. The recreational harvest includes a greater proportion of species with higher Hg concentrations than are typically harvested commercially (Table 3). Many datasets exist that characterize Hg in biota for the Gulf, and are further described in Appendix B.

5.1. Are biota mercury concentrations higher in the Gulf?

There is evidence that some fish species have higher Hg concentrations in the Gulf than in the adjacent Atlantic Ocean. Adams and McMichael (2007) reported that both king mackerel and Spanish mackerel from the eastern Gulf contain significantly higher concentrations of Hg in dorsal muscle than those from the southeastern U.S. Atlantic coast (king mackerel: 1.51 vs. 0.94 $\mu g\, g^{-1}$ means). Other studies consistent with this finding include Glover et al. (2010), Katner et al. (2010) and Cai et al. (2007). Bluefish (*Pomatotomus saltatrix*) and common snook (*Centropomus undecimalis*) were also reported to have higher Hg concentrations in Gulf waters than in the Atlantic (Adams et al., 2003). The golden tilefish (*Lopholatilus chamaeleonticeps*), a deepwater (200–300 m) demersal species has similarly been found to have Hg concentrations in Gulf waters that are two to three-fold higher (Fig. 3) than concentrations observed from the U.S. Atlantic coast (Hall et al., 1978). By contrast, the migratory pelagic dolphinfish (*Coryphaena hippurus*) seems to have Hg concentrations no higher than conspecifics from the U.S. Atlantic coast or elsewhere within its range (Adams, 2009; Evans et al., unpubl.). Whether Gulf fish Hg concentrations are higher than other regions needs further study.

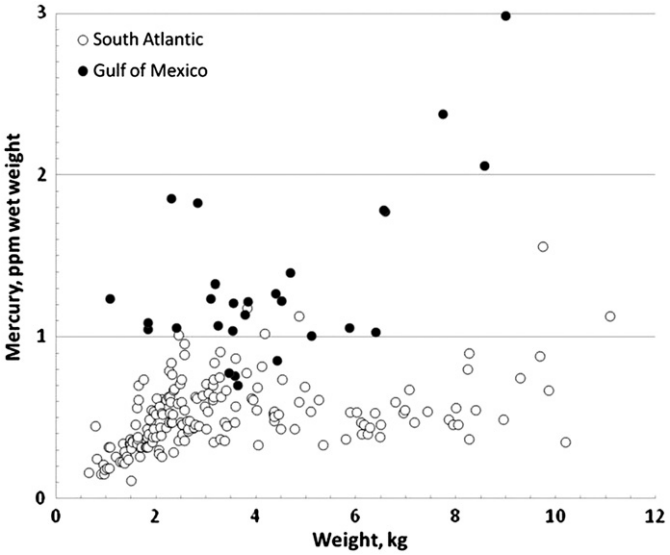

Fig. 3. Comparison of mercury concentrations in the golden tilefish as a function of weight in the Gulf of Mexico and U. S. Atlantic coasts.
Source: Hall et al. (1978).

5.2. Are there areas within the Gulf with elevated MeHg concentrations?

Inter-estuarine differences in fish Hg concentrations are to be expected given the differences in potential Hg sources, watersheds, and physical and biogeochemical characteristics of estuaries. Ache et al. (2000) identified two estuarine areas within the Gulf as biota Hg "hotspots". Lavaca Bay, Texas has local industrial contamination (Bloom et al., 1999; Gill et al., 1999), while eastern Florida Bay is within a national park with little industrial, urban, or agricultural Hg inputs. Marine and estuarine fishes in Florida Bay often accumulate higher concentrations of MeHg than in

other areas of Florida (Evans and Crumley, 2005; Adams et al., 2003; Adams and Onorato, 2005). Current fish consumption advisories are more restrictive for a number of species in Florida Bay as compared to advisories for these same species in other Florida coastal waters, including crevalle jack, great barracuda, spotted seatrout, red drum, common snook, tripletail, and gray snapper (*Lutjanus griseus*) (FDOH, 2012). Lowery and Garrett (2005) found differences in fish Hg concentrations among four Gulf estuaries, but these differences were not consistent across all species studied. Within estuaries, declining fish Hg levels have been observed along increasing salinity gradients. In Mobile Bay, Farmer et al. (2010) found that upstream areas of the estuary's delta had higher Hg concentrations in flounders and largemouth bass than downstream, though still brackish, areas. Lange et al. (2011) recently reported finding a steep gradient in Hg levels along the Shark River estuary in Everglades National Park, with concentrations declining seaward in gray snappers: 0.45 $\mu g\, g^{-1}$ at the upstream site, 0.15 $\mu g\, g^{-1}$ midstream, and 0.05 $\mu g\, g^{-1}$ at the mouth of the estuary.

In open Gulf waters, mixing should reduce MeHg concentration gradients observed in estuaries. Cai et al. (2007) did not find spatial differences in Hg levels in several offshore pelagic species sampled in Texas and Louisiana. Dolphinfish, a migratory pelagic species living offshore did not show an east/west Hg concentration difference. Biota from the northwestern and northeastern Gulf might be expected, however, to differ in Hg content because of the influence of the Mississippi River as a source of both Hg inputs (Rice et al., 2008; Harris et al., this issue) and nutrients that increase biological productivity (e.g. Dagg and Breed, 2003). Increased productivity can have competing effects on MeHg, including increased microbial activity that could enhance methylation rates, and increased fish growth rates, that tend towards lower fish Hg concentrations. Golden tilefish caught west of the Mississippi River plume, for example, had Hg concentrations approximately 30% higher on average than fish captured east of the plume, after adjustments for fish size (Lombardi and Evans, unpubl.). As in the case of spatial comparisons of the Gulf versus Atlantic, further effort should be made to assess within-Gulf geographic variations in fish tissue Hg levels, using existing databases with targeted monitoring to fill data gaps.

5.3. How important are fish movements and migrations in redistributing MeHg?

Fish movement within and among habitats affects fish MeHg exposure, and has the potential to represent a vector to transport MeHg among Gulf regions. Marine species have many life-history strategies by which they move across temporal and spatial scales, including daily, tidal or seasonal relocation of home range, ontogenetic shifts in habitat use, spawning migrations, and dispersal and directed movement of planktonic early life stages (Pittman and McAlpine, 2003; Gillanders et al., 2003). Migrations of estuary-dependent juveniles to adult habitats offshore (see Gillanders et al., 2003) and the diurnal vertical migration of zooplankton are among the most significant coordinated mass movements in oceans.

There is evidence of migrations of bluefin tunas, some billfishes, and king mackerel between the Gulf of Mexico and the Atlantic Ocean (Ortiz et al., 2003), and the Gulf has a high proportion of "estuarine dependent" species that migrate to adult habitats offshore. Examples include gag and other groupers as well as several species of snappers that move from estuarine or nearshore juvenile nursery grounds to offshore reefs as adults. Carbon, N, and S isotopes have been used in Florida Bay to track the migration of shrimp from estuarine nursery habitats to offshore feeding grounds (Fry et al., 1999). Although the effects of such movements on biota MeHg exposure and MeHg transport in the Gulf are largely unstudied, recent attention has been given to the potential importance of Hg biotransport in marine systems, in particular from estuaries out to coastal ocean (Fitzgerald et al., 2007; Chen et al., 2008,2009). Biotransport has also been documented for other regions and substances. Deegan (1993) estimated 5–10% of the total primary production of a Louisiana estuary was exported just in the form of menhaden, *Brevoortia patronus*. There are also numerous cases of biotransport of persistent organic pollutants as a result of large-scale synchronized movements of animals including fish, birds, and marine mammals (Merna, 1979; Ewald et al., 1998; Wania, 1998; Blais et al., 2005; Blais et al., 2007). Salmon were estimated to transport a substantial portion of a river's MeHg budget, over 1 $kg\, yr^{-1}$, to Bristol Bay, Alaska watersheds from the ocean (Zhang et al., 2001). Senn et al. (2010) recently concluded, however, that there was potentially a disconnect between the near coastal and oceanic food webs off Louisiana, based on the distributions of stable isotopes of C, N, and Hg in collected fish. Offshore, highly migratory species such as yellowfin tuna appear linked to a more phytoplankton-based food web and MeHg that has been subject to greater photo-degradation than nearshore species. These tuna are known to migrate between the Gulf of Mexico and the Atlantic Ocean.

5.4. Are there different food webs in the Gulf supporting different Hg bioaccumulation?

Fisheries production in open Gulf waters is likely to be supported by phytoplankton-based food webs, while terrestrial, seagrass, salt marsh and mangrove productivity, and benthic pathways are more important in some estuarine and nearshore waters. As discussed in Section 5.2, the Mississippi River introduces large quantities of nutrients that support high primary and secondary production and the largest fish harvests in the Gulf. These food-web differences can ultimately affect fish MeHg concentrations, as the largest single biomagnification step for MeHg is at the base of the food web (e.g. Wiener et al., 2003). Measurements of stable isotopes of C, N, and S in Gulf biota confirm geographic and habitat differences in food webs (Senn et al., 2010). Within Florida Bay, gradients in the importance of different primary producers in supporting fish and invertebrates are evident over even small spatial scales (Evans and Crumley, 2005; Chasar et al., 2005). The effects of these differences on MeHg bioaccumulation have not yet been fully evaluated. Existing datasets described in Appendix A may be useful in this regard, particularly those with isotopic data on nutrients and Hg.

6. Human exposure

MeHg is a neurotoxin that affects the central nervous system in humans and can cause long-term delays in neurocognitive development of children (Mahaffey et al., 2011). Several studies have shown that other effects of MeHg exposure on adults may include cardiovascular impairment and endocrine disruption (Salonen et al., 2000,1995; Tan et al., 2009). The primary source of MeHg exposure for most North Americans is consumption of marine and estuarine fish (Mahaffey et al., 2011, 2009, 2004). The most recent Hg exposure study for U.S. women of childbearing age suggests that between 3% and 15% pregnant women have blood Hg concentrations that are high enough for fetal blood levels to exceed the US EPA's MeHg reference dose (safety standard), depending on whether concentrations of MeHg in umbilical blood are considered (Mahaffey et al., 2009). The same

study showed that blood Hg levels in coastal residents are significantly elevated compared to the rest of the country.

Per-capita fish consumption in the Gulf of Mexico region (46 g day^{-1} for fishers, 24.4 g day^{-1} when adjusted for non-fish consumers) is elevated compared to the U.S. national average of 16.9 g day^{-1} (Degner et al., 1994; US EPA, 2002). Mahaffey et al. (2009) showed that women of childbearing age in the Gulf of Mexico most commonly consume tuna, shrimp, salmon, and finfish like grouper and snapper at an average rate of 5.9 meals per month and >8 meals per month for Asian and Native Americans. Data from Imm et al. (2007) suggest that children consume fish at a similar frequency to their mothers (average 5.17 meals/month and 12% >8 meals per month). Reported rates of fish consumption in the Gulf of Mexico region from the NHANES survey are higher than all other areas of the U.S. except the Atlantic coastal region (Mahaffey et al., 2009). Similarly, Warner (2007) reported significantly higher levels of Hg in hair from coastal residents and anglers in Alabama compared to residents from northern parts of the state.

Since Hg concentrations across fish species can vary by more than 100-fold, data on both quantities and species consumed are needed to accurately estimate MeHg exposures. Many dietary surveys report the frequency of fish meals consumed but do not provide information on meal sizes or species, needed to estimate variability in Hg exposures (Karouna-Renier et al., 2008; Patch et al., 2005; Mahaffey et al., 2009; NRDC, 2010). Fish consumption rates reported from a survey by Degner et al. (1994), averaged across all demographic groups for the top 25 categories of fish consumed by Florida residents are shown in Fig. 4. Also shown are the respective fish Hg concentrations (measured as THg and assumed to be MeHg) and the ranked contributions to overall Hg intake by residents. The data in Fig. 4 reflect the consumption of seafood items caught both within and outside the Gulf. Tuna, grouper, and snapper account for almost 60% of the Hg intake because of the large quantities of these fish consumed (Fig. 4B). Shrimp are the second most frequently consumed category of seafood in the Gulf but account for only slightly more than 2% of Hg intake because of low tissue Hg concentrations (Fig. 4). The relative contributions of different fish species to Hg exposure vary considerably across different demographic groups. For example, recreational fishers Hg exposures are dominated by species like red drum and spotted seatrout (Fig. 5) (Katner et al., 2011; Lincoln et al., 2011).

Lincoln et al. (2011) showed that recreational fishers and their families in Louisiana consume large quantities of fish harvested from the Gulf (74% of total consumption) and that hair Hg concentrations in 40% of the population surveyed exceeded levels associated with the US EPA's reference dose for MeHg. These groups are therefore likely to be among the most vulnerable groups to effects associated with high levels of MeHg exposures (Imm et al., 2007; Lincoln et al., 2011; Sunderland et al., 2012). Improved data on the sources of fish consumed by Gulf state residents are needed to understand how human exposures to Hg will change with reductions in Hg inputs and fish MeHg levels. This information could also be used to better inform the public of existing risks and options to reduce risks.

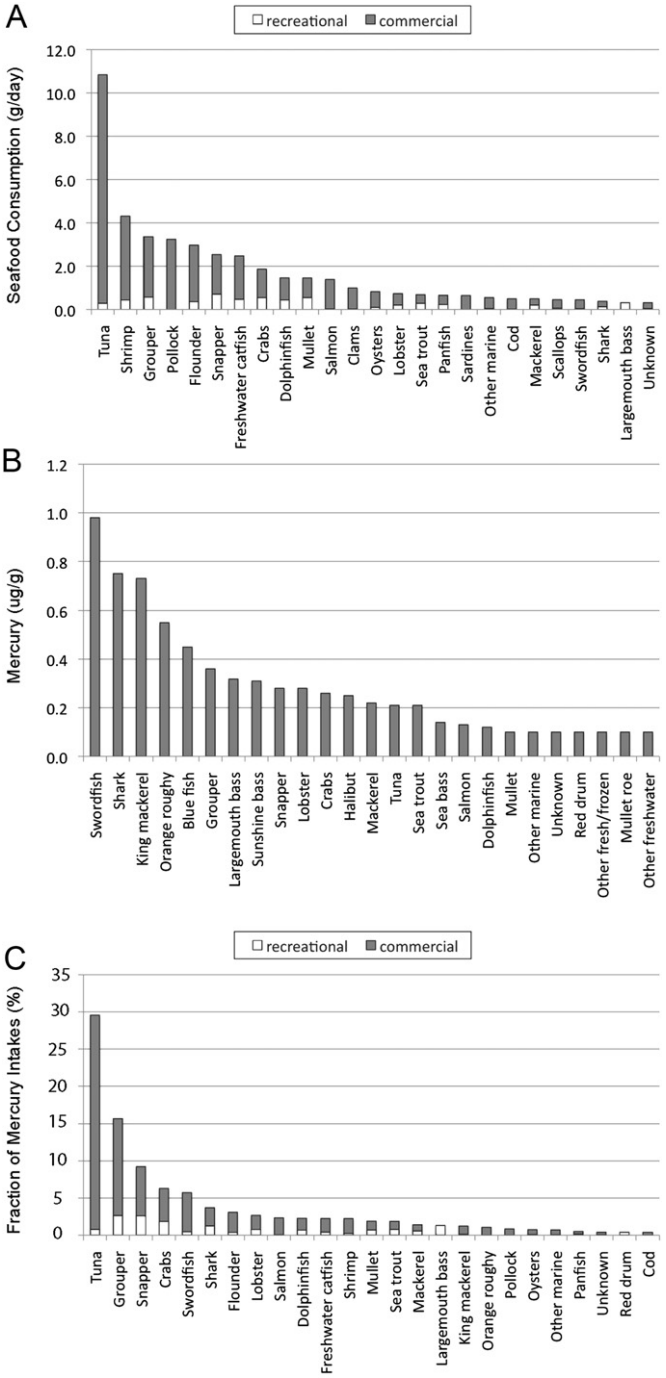

Fig. 4. Top 25 seafood categories for Florida residents per-capita: (A) seafood consumption, (B) mercury concentrations, and (C) mercury intakes. Seafood consumption data based on Degner et al. (1994) and fish mercury data are from Sunderland (2007) and Lowery and Garrett (2005). Mercury concentrations are not exclusive to the Gulf and include results from other regions.

7. Conclusions

The Gulf of Mexico has major commercial and recreational fisheries of national importance in the United States. Fish Hg concentrations are elevated in some species in the Gulf, including king mackerel, sharks, and tilefish. Fish consumption advisories for Hg are in place for all five Gulf states. Per-capita fish consumption in the Gulf is elevated compared to the U.S. national average. Swordfish have the highest Hg concentrations consumed by Floridians, but tuna, grouper, and snapper account for more than half of the Hg intake because of the larger quantities consumed. Because Gulf recreational fishers and family members consume greater quantities of Gulf fish than the national average, and because Hg levels can be elevated in these fish compared to other areas, they have a greater potential for increased MeHg exposure. This is supported by surveys of recreational fishers in the Gulf region showing high percentages of respondents exceeding the US EPA's reference dose for MeHg (Lincoln et al., 2011).

There is evidence that fish Hg concentrations in the Gulf are higher than in Atlantic waters for some but not all fish species.

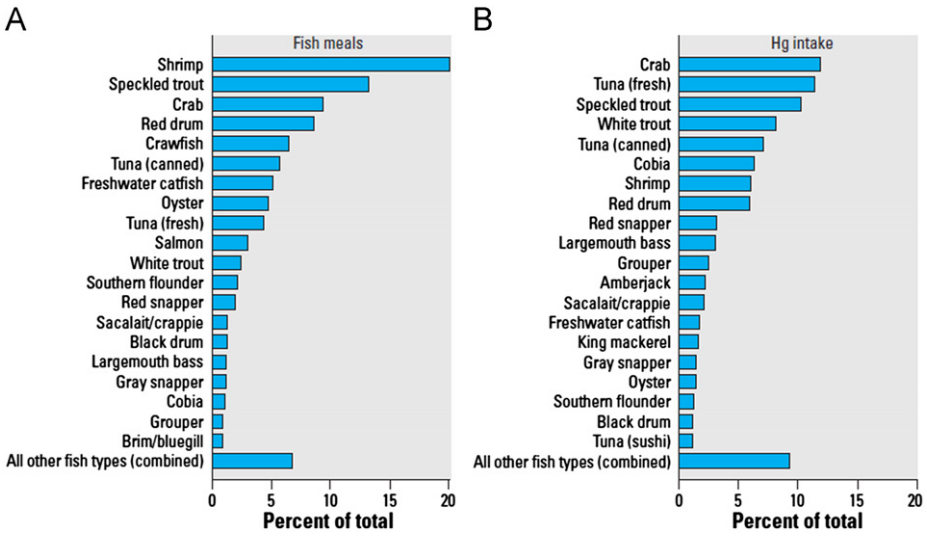

Fig. 5. Percent of total fish meals (A) and Hg intake (B) contributed by individual fish types to total intake across all Louisiana recreational anglers. From Lincoln et al. (2011).

Factors governing fish Hg levels in the Gulf of Mexico, and in oceans generally, are not adequately resolved. Atmospheric wet Hg deposition is elevated to the Gulf region in comparison to most other areas in the U.S., but the dominant source of Hg to the Gulf as a whole is estimated to be Atlantic inputs via the Loop Current. Redistribution of atmospheric, Atlantic and terrestrial Hg inputs to the Gulf occurs via water circulation but is under-constrained. Fundamental questions remain regarding where in-situ methylation occurs and what trophic factors lead to high bioaccumulation factors in marine waters (high MeHg in some fish, generally low MeHg concentrations in surface waters).

8. Future needs

Our analysis reinforces the disparity between the high importance of the Gulf fishery and a limited understanding of factors controlling Gulf fish Hg levels and associated risks to humans. There has historically been an emphasis on studies of Hg in freshwater systems, partly because they are often more manageable in terms of size and complexity. Increased attention is now needed for Hg in marine systems generally, including the Gulf of Mexico. This need has been recognized for the Gulf of Mexico at a high level federally in the United States for several years (NSTC, 2004). Field data are needed to better describe THg and MeHg sources, sinks, and concentrations in estuaries, coastal and pelagic regions, including measurements in the water column, sediments, and lower food web. Hg data are essentially non-existent for Hg in open waters of the Gulf. Mexico and Cuba also border the Gulf of Mexico and a multinational effort is encouraged. Finally, to understand the link between Gulf fish Hg levels and human exposures, updated dietary survey data are needed for Gulf state coastal residents. The only statistically representative survey of Gulf of Mexico state residents was conducted in Florida in the 1990s (Degner et al., 1994) and few data are available for other Gulf states.

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Appendix A. Development of atmospheric Hg deposition model scenarios

The version of the GEOS-Chem global biogeochemical model applied here (Holmes et al., 2010) used the Streets et al. (2009) global anthropogenic emission inventory for 2006, and the spatial pattern from the GEIA 2000 inventory (Pacyna et al., 2006). The simulation with AMSTERDAM (also known as CMAQ-MADRID-APT) was conducted for calendar year 2002 and used speciated Hg emissions estimated for (1) U.S. coal-fired electric generating units from 2002 Continuous Emissions Monitoring System (CEMS) heat input data and known coal quality and controls information and (2) for other US sources from US EPA's, 2001 Clean Air Hg Rule (CAMR) modeling which reflected a late 1990s/early 2000s time frame (US EPA, 2005). Boundary conditions for speciated Hg were obtained from GEOS-Chem modeling for 2002. The REMSAD simulation used the USA emissions data utilized by the US EPA in the CAMR modeling, with updates to key emitters to better match the 2001 modeling year. Further information on REMSAD emissions can be found in "Emissions Inventory and Emissions Processing for the Clean Air Hg Rule (CAMR)" (US EPA, 2005; Atkinson et al., 2008). REMSAD boundary conditions consisted of boundary layer concentrations of inorganic Hg(II) and Hg(0) from three global models: CTM (Seigneur et al., 2001), GRAHM, (Dastoor and Larocque, 2004) and GEOS-Chem.

The AMSTERDAM and REMSAD models used meteorological inputs that predicted excessive rainfall over the USA; consequently, these models also over-predicted wet Hg deposition when compared to MDN sites in the USA (27% and 33%,

Table B1
Major Gulf-wide compilations of mercury concentrations in biota.

Reference	Samples	Species	Dates	Comments
Hall et al. (1978)	1,699	53	1975	Includes invertebrates
Ache et al. (2000)	26,322	181	1990–2000	Includes invertebrates
US EPA (2003)	7,224	108	1990–2001	Includes invertebrates
Lowery and Garrett (2005)	1,660	24	2003–2004	Estuarine, reef, and pelagics
Kimbrough et al. (2008)	10,051	1	1986–2009	Estuarine invertebrate

respectively). Modeled wet deposition fluxes over the Gulf of Mexico were subsequently adjusted by those percentages to match the observed wet deposition from MDN sites along the Gulf coast more closely (Table 2). GEOS-Chem simulations did not exhibit the same tendency to overestimate wet Hg deposition and were not adjusted.

Differences among the model estimates of THg deposition may be partly due to the different emission inventories in each model. All three models predicted significant seasonal variability in the wet deposition of Hg, mostly driven by summertime increases in rainfall. Approximately 60–70% of the annual wet deposition is predicted to occur between May–October. These predictions are consistent with data from the NADP Hg Deposition Network (MDN) that show maximum wet deposition rates (~60–75%) during the May through October "wet season" at two active MDN sites (AL24 and F05) (Table 2). Data from the three Pensacola Atmospheric Hg Study sites (PAMS) showed an average annual wet deposition from 2005 through 2010 of 15.5 $\mu g\, m^{-2}\, yr^{-1}$, with 64% depositing from May through October (Table 2).

Appendix B. Mercury in biota databases

Many datasets exist characterizing Hg concentrations in biota of the Gulf of Mexico. The majority of these data are for edible seafood species from monitoring programs in U.S. waters, designed to assess the need for consumption advisories. Little published information on Gulf biota Hg concentrations was identified for the two nations that border the southern Gulf, Mexico and Cuba. Existing data collectively permit a preliminary assessment of what species have the highest Hg concentrations and therefore present greater risk to consumers. To a lesser extent, the data suggest geographic differences in Hg concentrations that can help to identify hot spots of concern. Extensive maps document the distribution of harvested species within the Gulf and general patterns of movement and migration (NOAA, 1985). Finally, there is information available for many food webs in the Gulf that can be useful to help infer the transfer of MeHg from water and sediments to primary producers and subsequent biomagnification to higher trophic levels. Hg data for these lower trophic level organisms, especially invertebrates, are rare, however for the Gulf and marine systems generally.

Table B1 lists the major published Gulf-wide data sets on Hg in Gulf biota. Several are compilations of original data published elsewhere. The US EPA also maintains a fish consumption advisory database documenting Hg and other contaminant concentrations in biota used by the states, tribes, and the Federal Government to justify consumption advisories. Smaller, area and species-specific published studies include pelagic fish off of Texas and Louisiana (Cai et al., 2007), gamefish and forage fish from Florida Bay (Evans and Crumley, 2005), recreational gamefish off of Alabama (Warner and Savitz, 2006), snappers off Louisiana (Bank et al., 2007; Wells et al., 2008), and groupers and seabasses off Florida (Tremain and Adams, in press). Adams et al. (2003) summarized a large body of Hg concentrations in marine species along Florida's Gulf coast. Other important unpublished datasets include those of state monitoring programs (e.g. Texas Department of Health, 1998).

A broad array of isotope studies for nutrients also exists in the Gulf (Evans and Crumley, 2005; Bank et al., 2007; Cai et al., 2007; Akin and Winemiller, 2008; Senn et al., 2010), some also measuring Hg, that would help to distinguish features among food webs and test the validity of food web models such as EcoPath and NETWRK (Vidal and Pauly, 2004).

Finally, fish Hg databases typically report on THg concentrations rather than MeHg under the assumption that most Hg in fish muscle is methylated. However, with invertebrates, and some lower trophic level fish, the percentage of THg that is MeHg has been found to be highly variable (Thera, 2011). Careful consideration should be given to this issue when using datasets characterizing Hg levels in aquatic biota.

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