Chapter 5
Mercury Emission Sources and Contributions of Atmospheric Deposition to the Everglades

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Abstract This chapter presents a summary of the literature on mercury (Hg) air emission sources in south Florida, the rest of the state, and the rest of the United States and the world, and their historical and current contributions to atmospheric deposition of Hg in the Everglades from monitoring and computer modeling. Hg deposited to the Everglades is a combination of gaseous oxidized Hg and particulate Hg emitted locally and gaseous elemental Hg emitted by distant sources and transported to south Florida where it is converted to the oxidized form that deposits more readily. While local emission sources of Hg likely contributed a large fraction (30% to over 45%) of deposition to the south Florida peninsula till the 1990s, stringent control measures have since been implemented on these sources and Hg reduced in commonly used materials. Currently, less than 1% of Hg deposition to the Everglades is likely due to Florida sources, with 85–95% due to the long-range transport of Hg from non-US sources. Summertime thunderstorms represent a key mode for the transfer of the Hg transported from distant sources to the Everglades as they reach into the upper troposphere where the oxidized form of Hg resides and mix it down to lower altitudes from where it is deposited to the Everglades under wet and dry conditions.

Keywords Source attribution · Modeling · Convective storms · Thunderstorms · Long-range transport · Atmospheric mercury transformations

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5.1 Introduction

As the deposition of inorganic mercury (Hg) emitted to the atmosphere represents most of the external contribution of Hg to the Everglades, it is important to understand the potential sources of these emissions. Hg emissions to the atmosphere result from the release during anthropogenic activities or natural phenomena of Hg present in minerals and soil, vegetation and water, and due to the release of Hg in products and industrial processes. The speciation of Hg emissions varies by source category and affects the eventual fate of these releases (see Chap. 3, this volume). Due to its long half-life in the atmosphere (approximately 6–12 months), gaseous elemental Hg [GEM or Hg(0)] emitted from sources far from the Everglades (such as in other US states and other countries) undergoes long-range transport, and can contribute to atmospheric deposition in the Everglades. Gaseous oxidized Hg [GOM or Hg(II)] emissions exhibit more local-scale deposition while the deposition of particulate bound Hg [PBM or Hg(p)] depends on the size of the particles to which Hg is bound, with coarse PBM deposition occurring primarily due to local sources. The elemental and divalent forms of Hg convert between each other, thus further affecting these deposition rates. Thus, when identifying the emission sources contributing to Hg contamination in the Everglades, it is important to examine not only local sources but also other sources in Florida, elsewhere in the US and the rest of the world. The speciation of Hg emissions also may change with the application of control technologies due to chemical transformation. In particular, coal-fired electric utilities could have very different Hg speciation depending on coal quality and controls implemented. For example, the use of a selective catalytic reduction (SCR) system increases the fraction of GOM due to oxidation from GEM while the use of a scrubber decreases the fraction of GOM and PBM. Because the effect of Hg emissions on ecosystems such as the Everglades may be seen for decades past their time of release (see Chap. 3, Volume III for a discussion of both legacy Hg deposits and the importance of newly deposited Hg in aquatic ecosystems), it is critical to consider the contributions of not only sources currently in operation, but also historical sources of Hg emissions in the Everglades region and further afield as discussed below.

5.2 Mercury Sources and Emissions in South Florida

An emissions inventory of Hg point and non-point sources for 16 counties in south Florida for 1996 (RMB 1999) identified soil degassing as the largest emission source category for both anthropogenic and natural sources (7158 kg/yr) while municipal waste combustors (MWC) were the largest anthropogenic contributor at (4445 kg/yr or 82% of all anthropogenic emissions) followed by power generation facilities, medical waste incinerators (MWI), and sugar refineries (435,213 and 136 kg/yr, respectively). Emissions from Broward, Miami-Dade and Palm Beach counties comprised nearly half (49%) of the total Hg emissions in the 16-county area. Emissions from these three counties were identified as important contributors to Hg deposition in the Everglades through a multivariate receptor modeling analysis of wet deposition measurements (Dvonch et al. 1999). Therefore, a subsequent emissions inventory study in south Florida (RMB 2002) focused on Hg emissions in these three counties and across a 20-year historical period from 1980 to 2000. This historical inventory included the three largest emission source groups (MWC, MWI and sugar refineries) in the 3-county area as well as power generation facilities that were negligible in this area but were of interest for many observers.

Hg emissions in the three counties with large historical contributions to Hg deposition in the Everglades increased three-fold from 1982 to 1983 as MWCs and MWIs became operational (Fig. 5.1). Emissions from these two source categories continued to increase through 1991 when total Hg emissions in the 3-county area were estimated to have reached their peak of 3077 kg/yr. After the USEPA promulgated new source performance standards (NSPS) in mid-1992 and subsequently required installation of Hg control technology, many MWIs ceased operations and medical waste was either autoclaved for steam sterilization or sent offsite for processing (RMB 2002). Actual test data were used to estimate emissions for MWCs and the reductions in emissions in the mid- to late-1990s are believed to be due to the implementation of controls such as carbon injection (RMB 2002). Total emissions from MWIs and MWCs declined by 93% from 1991 to 2000. Over the entire 20-year period examined, MWIs and MWCs together represented over half of the total emissions and as much as 96% of the total in 1990–1992. During the relatively high (> 1000 kg/yr) emission years of 1983–1994, sugar processing and power generation typically constituted less than 10% and 1%, respectively, of the total.

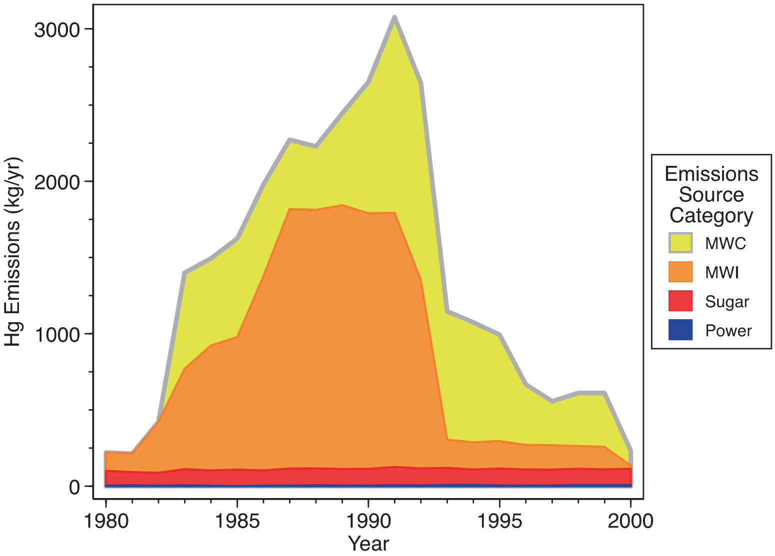

Fig. 5.1 Estimated annual Hg emissions during 1980–2000 from power generation, sugar processing facilities, medical waste incinerators and municipal waste combustors in Broward, Miami-Dade and Palm Beach counties. (Data source: RMB 2002)

While the inventory described above did not include Hg speciation, Dvonch et al. (1999) estimated the GEM/GOM/PBM split to be approximately 4/95/1% for MWIs and 24/75/1% for MWCs. Because emissions from MWIs and MWCs have such a large fraction of GOM (the form of Hg that tends to deposit near sources), these two source groups in the 3-county area likely had a large historical contribution to the Everglades during their period of operation.

A different approach to constructing a Hg emissions inventory was adopted by Husar and Husar (2002) who estimated the historical annual amount of Hg mobilized in fuels and consumer and commercial products, and inferred emissions to the atmosphere using information on fuel consumption and Hg use/content in the production of Hg containing products. These estimates were made for the US and then applied to the regional level in Florida for 1930–2000. Similar to the RMB (2002) study, Hg chemical speciation was not estimated. In addition, Husar and Husar (2002) did not consider the agricultural sector. Water-based paint applications dominated Hg-containing products from the 1960s to 1990 for Broward County and Miami-Dade County (and elsewhere in Florida; Fig. 5.2) and then abruptly declined when USEPA banned Hg use in paint in 1990. Husar and Husar (2002) assumed that 75% of the Hg in paint used was released to the atmosphere in the first year of use. They also assumed that Hg in products categories (electrical, controls, laboratory) were disposed as municipal solid waste (MSW). The fraction of waste burned was based on Florida Department of Environmental Protection (FDEP) reports for 1990–1998 and assumed to range from 10% to 30% for earlier years. The second largest Hg contributions to Broward and Miami-Dade were associated with the disposal of electrical (battery) devices.

The estimates for coal contribution to Hg emissions for Broward, Miami-Dade and Palm Beach counties were assumed to be negligible and were not included. Using the assumption that electrical, control, and laboratory use category will end up in MSW, Husar and Husar (2002) estimated the sum of Hg emissions for Broward, Dade, and Palm Beach Counties and compared these with the MWI and MWC (i.e., MSW) emission estimates of RMB (2002) (Fig. 5.3). Here, MSW_R represents the municipal solid waste incineration from RMB (2002) while MSW_R + MWI_R is the sum including medical waste incineration. The RMB Hg emission estimates are approximately 10–50% of the mercury amount mobilized in electrical use, laboratory use, and controls as estimated by Husar and Husar (2002)

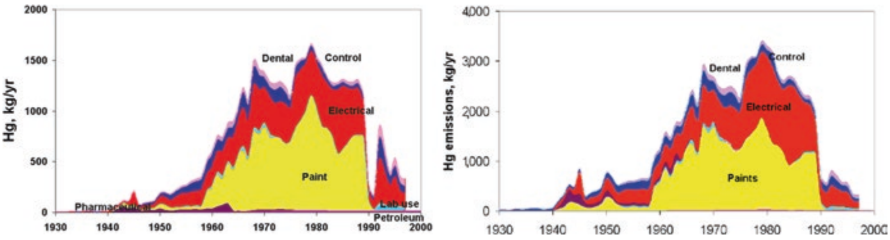

Fig. 5.2 Annual Hg releases during 1930–2000 estimated from mercury mobilized in fuels and goods in Broward (left) and Miami-Dade (right) counties. (From Husar and Husar 2002)

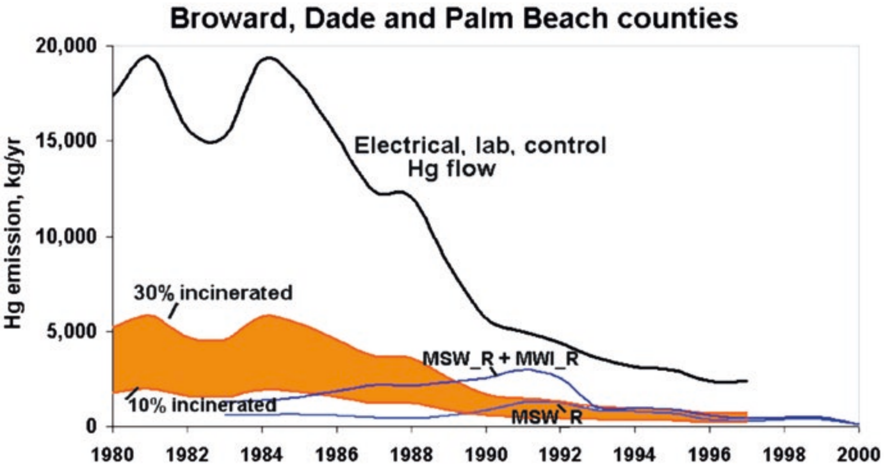

Fig. 5.3 Sum of municipal solid waste and medical waste incineration emissions for Broward, Miami-Dade, and Palm Beach counties. (From Husar and Husar 2002)

because only a fraction of the Hg in goods is incinerated and emitted. The shaded area in Fig. 5.3 represents a crude emission estimate by Husar and Husar (2002) assuming that 15–30% of Hg in goods is incinerated; the emissions estimates at the lower end of that range are generally comparable with those of RMB (2002). Husar and Husar (2002) judged the estimation of county Hg emissions from the Hg mass mobilization data to be the most uncertain part of their entire analysis.

5.2.1 Emissions Trends for South Florida from the USEPA National Emissions Inventory

The USEPA National Emissions Inventory (NEI) provides a detailed estimate of air emissions of criteria pollutants and precursors, and hazardous air pollutants including mercury.[1] The NEI is released every 3 years based primarily upon data provided by state, local, and tribal air agencies for sources in their jurisdictions and supplemented by data developed by the USEPA. Figure 5.4 and Table 5.1 present a summary of facilities with mercury emissions exceeding 10 lb/yr from the 2002, 2005, 2008, 2011 and 2014 NEI data for southern Florida counties with Everglades habitat. A reduction in the number of such facilities is evident over this 13-year period largely reflecting increasing controls on Hg emissions and closure of units with large sources mainly in Miami-Dade, Broward and Hendry counties. When considering emissions from all facilities (i.e., those at all emissions levels) in this region, emissions declined by 53% from 828 lb/yr in 2002 to 391 lb/yr in 2011 and have then subsequently increased by 6% in 2014 (Table 5.2). While emissions from

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[1] https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei

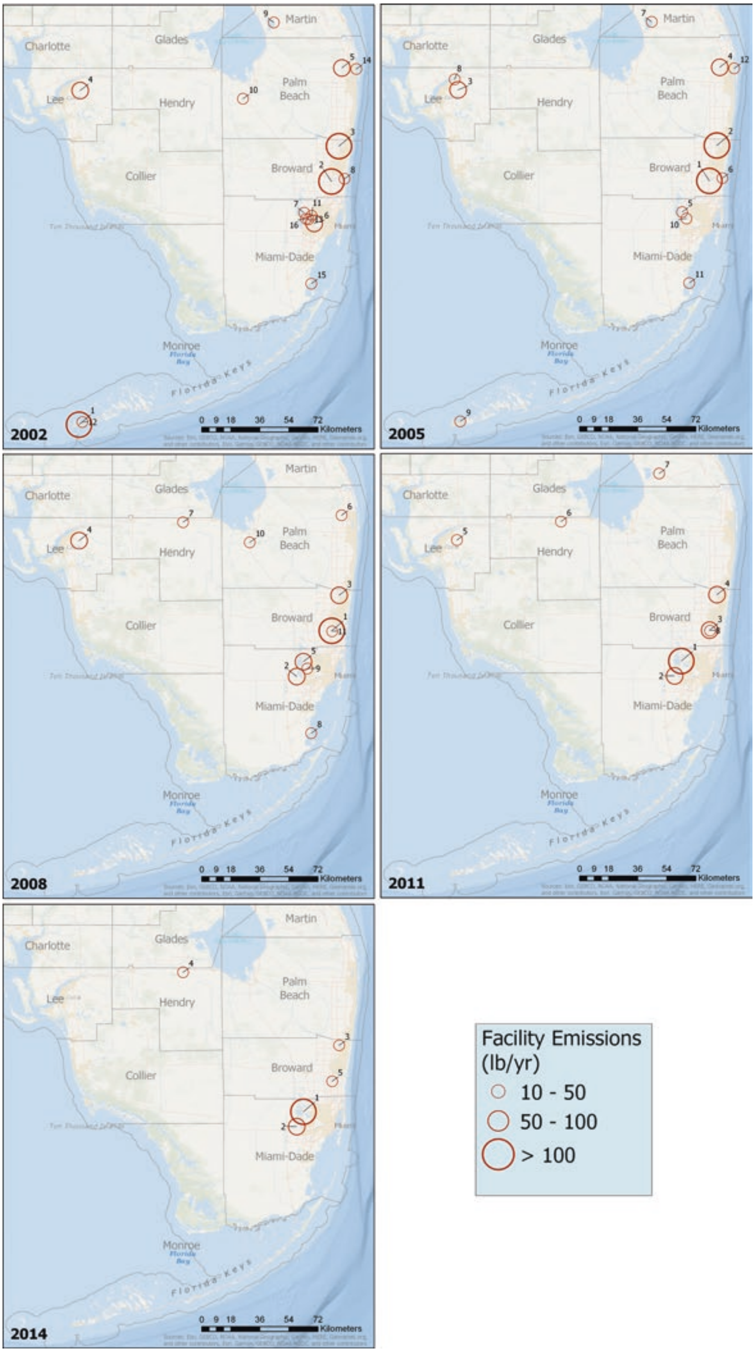

Fig. 5.4 Facilities with mercury emissions exceeding 10 lb/yr from National Emissions Inventory data for 2002, 2005, 2008, 2011, and 2014 for southern Florida counties (see Table 5.1 for emission values)

Table 5.1 Facilities with Hg emissions exceeding 10 lb/yr from NEI data for southern Florida counties (see Fig. 5.4 for locations) (note: ranks in different years correspond to different facilities)[a]

	2002	2002	2005	2005	2008	2008	2011	2011	2014	2014
Rank	Facility type	Hg (lb/yr)	Facility type	Hg (lb/yr)	Facility type	Hg (lb/yr)	Facility type	Hg (lb/yr)	Facility type	Hg (lb/yr)
1	MWC	151.4	MWC	116.9	MWC	141.8	Cement	112.0	Cement	222.0
2	MWC	116.8	MWC	100.6	Cement	84.0	Cement	61.0	Cement	74.0
3	MWC	100.6	MWC	96.2	MWC	81.7	MWC	55.7	MWC	35.1
4	MWC	96.2	MWC	73.8	MWC	73.0	MWC	54.2	Sugar mill	22.0
5	MWC	73.8	Cement	43.1	Cement	69.8	MWC	26.9	FFEPG	10.4
6	MWC	60.6	FFEPG	28.5	MWC	42.0	Sugar mill	19.0		
7	Cement	44.5	FFEPG	22.4	Sugar mill	20.6	FFEPG	18.0		
8	FFEPG	29.5	FFEPG	18.3	Nuclear	16.0	FFEPG	10.9		
9	FFEPG	24.8	MWC	15.6	MWC	15.5				
10	Biomass	22.9	MWC	15.2	Biomass	13.5				
11	Foundry	19.7	Nuclear	13.1	FFEPG	10.8				
12	MWC	15.6	FFEPG	11.8						
13	MWC	15.2								
14	FFEPG	13.9								
15	Nuclear	13.4								
16	MWC	12.6								

[a] *FFEPG* Fossil fuel electric power generation, *MWC* Municipal waste combustion

Table 5.2 Mercury emissions from facilities in southern Florida (lb/yr) derived from NEI estimates

Year	2002	2005	2008	2011	2014
Hg emissions (lb/yr)	828.3	562.5	592.5	391.0	415.1

MWCs and electric power generation have steadily decreased, cement manufacturing emissions have increased over the past decade and now represent the largest point source contributor to Hg in southern Florida (296 lb/yr in 2014).

5.3 Mercury Sources and Emissions in the Rest of Florida

As in south Florida, Hg sources and emissions in the rest of Florida have changed markedly over the last four decades due to regulations and the implementation of controls. Hg releases from the disposal of electrical devices dominated Hg releases in the state in the late twentieth century (Husar and Husar 2002) (Fig. 5.5) with Hg due to batteries in electrical devices peaking at over 50 Mg/yr. in the late 1980s (Fig. 5.6). The next largest category of Hg use in products was in water soluble

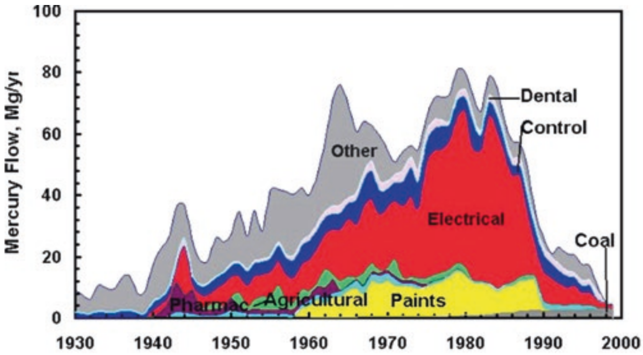

Fig. 5.5 Annual Hg releases inferred from material flow by major use category in Florida during 1930–2000. (From Husar and Husar 2002)

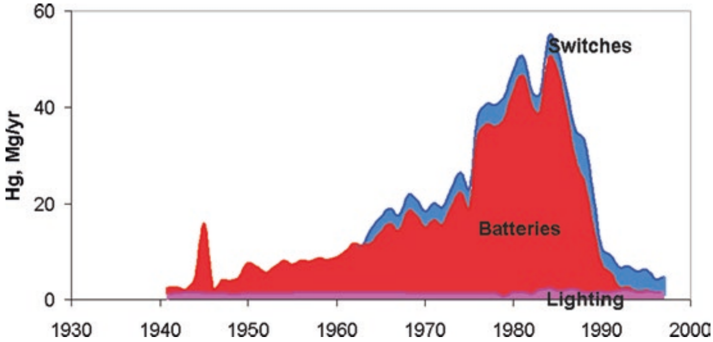

Fig. 5.6 Annual Hg releases from electrical device use in Florida during 1930–2000. (From Husar and Husar 2002)

paints due to the addition of organomercury compounds, and phenylmercuric acetate in particular, as fungicides to prolong the paint's shelf life, resulting in potentially over 10 Mg/yr emissions (Husar and Husar 2002) also in the late 1980s. Since that time, Hg has almost been eliminated from batteries and latex paints (FDEP 2013). Other historical sources of Hg emissions in Florida from electrical device use include the disposal of fluorescent and high intensity discharge (HID) lights and wiring devices and electric light switches. Non-Hg switches were introduced in the 1990s. However, switches have a life expectancy of up to 50 years (Husar and Husar 2002). Therefore, Hg releases due to the disposal of legacy Hg-containing electric light switches in Florida will continue through the 2030s.

Estimates of Hg emissions from coal combustion in Florida by Husar and Husar (2002), based on annual coal production, coal Hg content, the fraction of Hg released and various control efficiencies, show a steady, albeit small, increase from 1980 to 2000; these estimates compare well with measurements of Hg emissions made during the 1999 USEPA Information Collection Request (ICR) at power plants. Depending on the estimated Hg content of coal, the Hg emissions from coal combustion in Florida in 2000 ranged between 1.4 and 1.9 Mg/yr.

Table 5.3 Mercury emissions from major source categories in Florida in 2005, 2009 and 2014

Source category	2005 (from FDEP, 2013)		2009 (from FDEP, 2013)		2014 (from USEPA NEI)	
	lb/yr	% of total	lb/yr	% of total	lb/yr	% of total
Coal-fired electric generation	2094	53%	1469	46%	1185	36%
Cement Industry	710	18%	326	10%	559	17%
Waste to energy plants	692	17%	663	21%	252	8%
Other electric generation	314	8%	314	10%	310	10%
Waste water treatment plants	102	3%	102	3%	25	1%
All others	60	2%	295	9%	916	28%
Total	3972		3169		3247	

Adapted from FDEP, 2013 and the USEPA NEI

More detailed estimates of Hg emissions from coal combustion since 2000 are available from the NEI. Table 5.3 shows the major source categories of anthropogenic Hg emissions in Florida from the 2005 and 2014 NEI databases and from a 2009 estimate from the state-wide Florida Total Maximum Daily Load (TMDL) determination. As noted by FDEP (2013) and seen in Table 5.3, Hg emissions from coal combustion at Florida utilities have declined over the past 10 years or so because lower natural gas prices have spurred many utilities that historically relied on coal to use natural gas (and which has a Hg content that is negligible compared to coal) by either retrofitting existing units to use natural gas, constructing new gas-fired units or operating existing natural gas electricity generating units (EGUs) at higher capacity. In addition to lower natural gas prices, Hg emissions from Florida coal-fired power plants have generally steadily declined due to the installation of controls prior or in anticipation of implementation of the Mercury and Air Toxics Standards (MATS), with reductions between 57% and 90% at several plants (FDEP 2013).

Emissions from waste-to-energy plants have decreased due to the implementation of Hg controls as well. However, the reduction in Hg emissions from cement production from 2005 to 2009 was due to a slowing of the housing market rather than controls (FDEP 2013). Cement kiln Hg emissions have subsequently increased through 2014 and represent 17% of the total state-wide Hg emissions. Coal-fired electric generation continues to be the largest Hg source category at 36% in Florida with most of these sources present in the northern and north-western parts of the state.

5.4 Mercury Sources and Emissions in the Rest of the United States and the World

The atmospheric modeling performed during the Florida state-wide Hg TMDL (FDEP 2013) showed that the overwhelming sources of Hg deposition in Florida are non-US in origin, transported by global weather patterns, and that only a very small percentage of Hg deposition occurring within Florida originated from within the state. As a result, we present here a review of Hg emissions from sources outside Florida including those in the rest of the world. In addition to emissions from anthropogenic sources, Hg emissions to the global atmosphere come from primary natural geogenic sources and the re-emission of historically-deposited Hg that was originally emitted from both anthropogenic and natural sources. The possible impacts of re-emissions of historically deposited or legacy Hg on future atmospheric deposition fluxes of Hg are discussed in Chap. 3 of Volume III.

5.4.1 Natural Sources and Re-emission of Mercury

Global primary natural (i.e., geogenic) sources of Hg include Hg released during geothermal activity in volcanoes and geothermal events and through the continuous natural weathering of Hg-containing rocks. Estimates of annual releases to air from natural sources worldwide vary widely, from 80 to 600 Mg/yr (Corbitt et al. 2011; Mason et al. 2012; Lei et al. 2014). Hg released from natural sources is primarily in the form of GEM which is either dry deposited or oxidized to GOM in the atmosphere before wet and dry deposition to the Everglades.

Re-emissions of historically deposited Hg occur from the Earth's surfaces (soil, rocks, snow and ice, oceans and other surface waters, and vegetation) that have previously received Hg either from atmospheric deposition or through another transport pathway (AMAP/UNEP 2013; Pacyna et al. 2016). Such Hg reflects the legacy contributions of both anthropogenic and natural sources and is re-emitted from the Earth's surfaces in the form of GEM. Oceans are the most important sources of these emissions and are estimated to be 52% of the total natural and re-emitted emissions (Pirrone et al. 2010) and 36% of the total global inventory when including direct anthropogenic sources (Pacyna et al. 2016). Because Hg evasion from ocean waters is significantly higher near the tropics than in temperate latitudes (Strode et al. 2007), evasion from the Atlantic Ocean of previously-deposited Hg potentially constitutes an important source of Hg to the Everglades. Total annual Hg re-emissions to the atmosphere of historically deposited Hg worldwide have been estimated to be in the range of 4000–6300 Mg/yr (Mason et al. 2012). The second largest source of Hg re-emissions is agricultural burning and wildfires where Hg stored in biomass is released as GEM and PBM. Estimates in the literature for annual global Hg emissions from biomass burning in the 2000s compare favorably with values reported at 675 ± 240 Mg/yr (Friedli et al. 2009), 600–678 Mg/yr (De Simone et al. 2015), and 612 Mg/yr (Kumar et al. 2018). Hg emissions from biomass burning in the United States have been estimated to be 43 Mg/yr on average during 2002–2006 (Wiedinmyer and Friedli 2007).

Global estimates of total primary natural emissions and re-emissions of Hg continue to remain uncertain with the combined estimates varying from 5207 Mg/yr (Pirrone et al. 2010; Pacyna et al. 2016) to 6500 Mg/yr (Lei et al. 2014). In any case, direct natural emissions and re-emissions worldwide constitute over two-thirds of the global Hg emission budget (Pacyna et al. 2016) and thus together represent an important source of worldwide Hg emissions.

5.4.2 Anthropogenic Mercury Emissions in the Rest of the United States and the World

Numerous inventories of global anthropogenic Hg emissions have been presented in the literature over the years with improvements made over time by considering more sources or applying more robust methods (e.g., Pacyna et al. 2003, 2006, 2010, 2016; Seigneur et al. 2004; Streets et al. 2009; Pirrone et al. 2010; AMAP/UNEP 2013). The increased application of Hg emission controls over the past two to three decades has slowed down or, in some cases, decoupled emissions from increases in energy demand, resulting in decreases in Hg emissions in Europe and North America. Conversely, a general increase in emissions in Asia, particularly in China and India, has been observed, resulting from increased industrial production and energy demands (Sundseth et al. 2017). The global geospatial distribution of anthropogenic Hg emissions reflects areas of industrialization (including known locations of point sources of Hg) and human population densities or remote extraction operations of metals (Fig. 5.7, from Pacyna et al. 2016).

Global anthropogenic Hg emissions are dominated by emissions from east and southeast Asia which represent 40% of the estimated 2010 worldwide inventory of 1960 Mg/yr (Fig. 5.8, prepared from data in AMAP/UNEP 2013). These emissions are primarily due to coal combustion, gold mining, cement production and non-ferrous metal (aluminium, copper, lead and zinc) production. North America contributes only 3% of the global anthropogenic total while Central America and the Caribbean contribute 2%. When considering all geographic regions in the world, artisanal and small-scale gold mining represent the largest global mercury source sector,

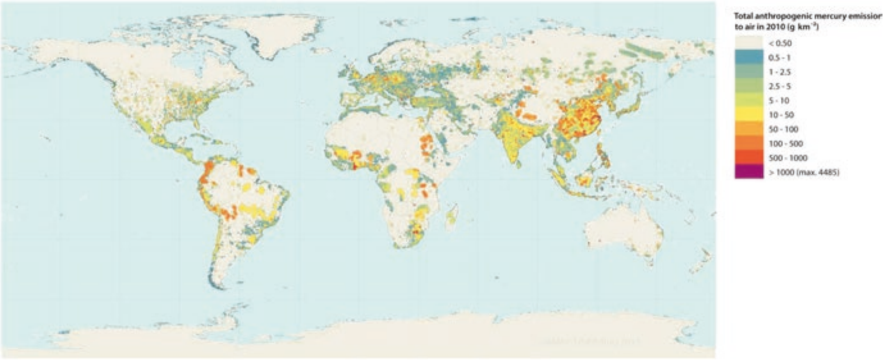

Fig. 5.7 Spatial distribution of global anthropogenic Hg air emissions in 2010. (From Pacyna et al. 2016)

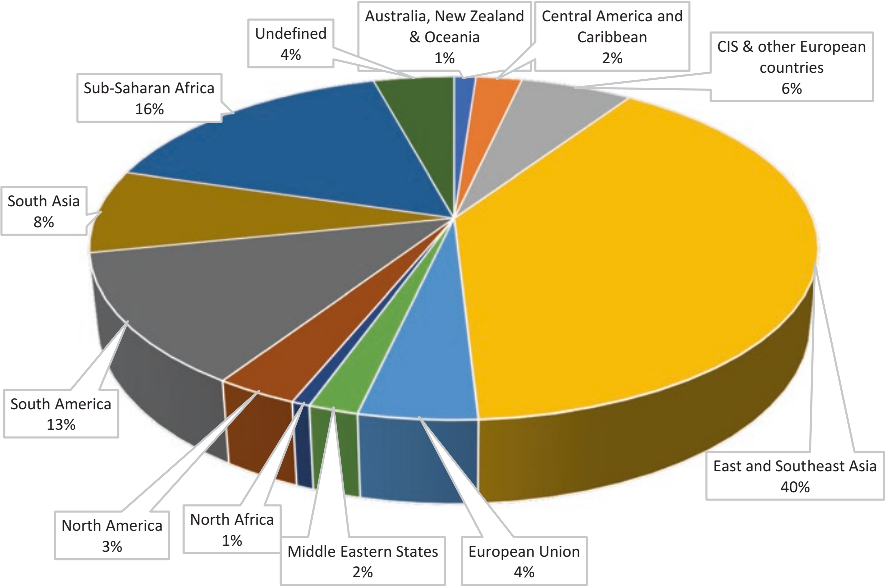

Fig. 5.8 Contribution of global anthropogenic Hg emissions in 2010 from different regions. (Figure created from data in AMAP/UNEP 2013)

constituting 37% of the total, followed by coal combustion at 24%. Other major emission sectors include production of nonferrous metals (10%), cement production (8.8%), and landfill waste and large-scale gold production at 5% each. There is significant uncertainty in the total estimate of global Hg emissions with an uncertainty range of 1010–4070 Mg/yr (AMAP/UNEP 2013; Pacyna et al. 2016) (Table 5.4).

The most recent U.S. national inventory of Hg emissions is from the 2014 USEPA National Emissions Inventory (NEI; USEPA 2014). Coal-fired power plants represent the largest anthropogenic source category of Hg emissions in the U.S., accounting for 44% of all domestic anthropogenic Hg emissions in 2014 (Table 5.5). U.S. Hg emissions have decreased significantly since the early 1990s due to the Hg co-benefits of the implementation of NOx, SO_2 and particulate matter (PM) source emissions controls for the implementation of USEPA NOx, SO_2 and PM emissions standards (FDEP, 2013). Emissions of Hg from coal-burning power plants have decreased considerably either due to Hg controls or from the retirement of coal-fired units in anticipation of implementation of the federal Mercury Air Toxics and Standards (MATS) rule finalized in 2012 (for implementation in 2016). For example, the USEPA's projected estimate of Hg emissions from US electric generating units in the 2016 MATS control scenario was 6.7 Mg/yr (14,800 lb/yr) compared to 47.8 Mg/yr (105,200 lb/yr) in 2005 (USEPA 2011). Furthermore, cheaper natural gas prices in the last decade have resulted in large transitions across the country from coal to natural gas usage for electric generation resulting in additional conversion or retirement of coal-fired power plants, thus further lowering Hg emissions due to the significantly lower content of Hg in pipeline natural gas versus coal.

Table 5.4 Global anthropogenic Hg air emissions by sector in 2010

Sector	Mg/yr	%
Artisanal and small-scale gold mining	727	37
Coal combustion	474	24
Non-ferrous metal production	194	10
Cement production	173	9
Large-scale gold production	97.3	5
Waste from consumer products (landfill)	89.4	5
Contaminated sites	82.5	4
Primary pig iron production	45.5	2
Chlor-alkali industry (Hg cell)	28.4	1.5
Oil refining	16.0	0.8
Mercury production	11.7	0.6
Oil and natural gas combustion	9.9	0.5
Waste from consumer products (incineration)	6.2	0.3
Cremation	3.6	0.2
Total	1959	100%

From AMAP/UNEP (2013)

Ferrous metals production, cement manufacturing and waste disposal comprise the next three largest source groups at 12%, 7%, and 7%, respectively, of the total U.S. anthropogenic inventory in 2014.

5.5 Source Contributions from Atmospheric Deposition to the Everglades

The Florida state-wide Hg TMDL (FDEP 2013) is the most comprehensive Hg modeling and monitoring study performed to date to understand atmospheric Hg deposition contributions to the Everglades and other parts of Florida. The methods and conclusions from this study are summarized below followed by other relevant studies.

5.5.1 Mercury Deposition Source Attribution from Florida State-Wide TMDL Study

The Florida state-wide Hg TMDL (FDEP 2013) included a rigorous atmospheric modeling effort supplemented with Hg deposition and air concentration monitoring by a team led by the University of Michigan Air Quality Laboratory (UMAQL) to characterize sources contributing to Hg deposition in Florida. An advanced multiscale modeling system was applied consisting of the global ECHMERIT

Table 5.5 Anthropogenic Hg emissions by sector in the United States in 2014. From the 2014 NEI (USEPA 2014)

Sector	Emissions (lb/yr)	%
Fuel combustion – Electric generation – Coal	45,912.9	44
Industrial processes – Ferrous metals	12,103.6	12
Industrial processes – Cement manufacturing	7384.5	7
Waste disposal	6774.4	7
Miscellaneous Non-industrial NEC[a]	6235.3	6
Industrial processes – NEC[a]	4515.4	4
Industrial processes – Non-ferrous metals	2681.7	3
Industrial processes – Chemical Manufacturing	2085.3	2
Fuel combustion – Electric generation – Natural gas	1977.4	2
Fuel combustion – Industrial boilers, engines – Coal	1665.0	2
Fuel combustion – Residential – Oil	1287.5	1.2
Fuel combustion – Industrial boilers, engines – Natural gas	1244.7	1.2
Fuel combustion – Other	3910.3	3.8
Industrial processes – Petroleum refineries	1232.3	1.2
Mobile – Locomotives	1212.1	1.2
Industrial processes – Storage and transfer	769.9	0.7
Mobile – On-road	726.0	0.7
Industrial processes – Mining	531.1	0.5
Industrial processes – Oil and gas production	434.4	0.4
Industrial processes – Pulp and paper	368.6	0.4
Mobile – Non-road	114.0	0.1
Solvent – Industrial surface coating and solvent use	288	0.3
Agriculture – Livestock waste	95.7	0.1
Other	75.9	0.1
Total	103,626	

[a] *NEC* Not easily classifiable

(Atmospheric General Circulation Model ECHAM5 with Mercury Chemistry) model and regional CMAQ (Community Multiscale Air Quality) model. ECHMERIT (Jung et al. 2009) and CMAQ (Bullock and Brehme 2002; Byun and Schere 2006; Bullock et al. 2008, 2009; Baker and Bash 2012) are both Eulerian tropospheric chemistry transport models that simulate Hg emissions, transport, atmospheric chemistry and deposition. ECHMERIT was applied at 2.81°× 2.81° horizontal resolution for calendar year 2009 using the global AMAP/UNEP Hg inventory for year 2000, the USEPA NEI Hg inventory for year 2005, primary natural and biomass burning emissions, and meteorology for year 2009. The ECHMERIT simulation provided boundary conditions (background concentrations) of GEM, GOM and PBM for CMAQ simulations conducted at 36 km resolution over the continental U.S., 12 km resolution over the southeastern U.S., and 4 km resolution over Florida. The CMAQ modeling was performed for year 2009 using meteorology driven by the Weather Research and Forecast (WRF) for 2009 and emissions updated to year 2009 for Florida using data from the 2005/2008 USEPA NEI and 2009 emissions data from Florida DEP. The air modeling was supplemented with monitoring in 2009 and 2010 for precipitation, Hg wet deposition, Hg dry deposition and air concentrations of GEM, GOM and PBM. In addition, multivariate receptor modeling was conducted using the positive matrix factorization (PMF) technique (Paatero and Tapper 1994).

The CMAQ modeling (FDEP 2013) provided speciated wet and dry loading estimates of Hg for TMDL eco-regions and basins across the state of Florida for year 2009. It also provided a measure of source attribution, i.e., the relative importance of in-state versus out of state emissions for TMDL eco-regions and basins in Florida, as well as a measure of potential reduction in atmospheric loadings resulting from future emission reduction scenarios. Multiple tags were incorporated in the CMAQ modeling to identify relative source contributions of: (1) Florida sources; (2) states in the southeastern U.S. other than Florida; and (3) other source regions. The PMF receptor modeling highlighted potential source groups contributing to deposition.

The monitoring in the TMDL study showed that Hg rain concentrations in summertime (June – August) in the Everglades were 2–4 times higher than in other seasons (Fig. 5.6). Similarly, summertime Hg wet deposition is also typically higher than in the other seasons. Annual Hg wet deposition fluxes simulated by CMAQ for 2009 were compared with observed wet deposition at the Everglades National Park and five other TMDL monitoring stations in Florida (see Fig. 5.5 for station locations). The ENP had the highest observed annual wet deposition across all six TMDL stations in the state with a measured wet deposition flux of 25.5 μg/m^2-yr. The CMAQ model performance at ENP was very good with a modeled annual wet deposition flux of 25.3 μg/m^2-yr. Modeled dry deposition fluxes of Hg from CMAQ for year 2009 were evaluated by comparison with dry deposition estimates determined using a separate inferential model based on surface measurements of ambient speciated Hg and meteorology measured at four TMDL project "supersites" located at Davie, Tampa, Jacksonville, and Pensacola. Annual Hg dry deposition modeled with CMAQ (31 μg/m^2-yr) at Davie, the supersite closest to the ENP, compared very well with dry deposition flux estimate from the inferential modeling (32 μg/m^2-yr).

Comparisons between CMAQ model results and measurements of GEM, GOM and PBM were also conducted for calendar year 2009 for the four TMDL supersite locations (Davie, Tampa, Jacksonville and Pensacola). The model shows very reasonable agreement with measured GEM with overall normalized mean bias of +15% and normalized mean error of 16%. However, the model severely overpredicts GOM and PBM concentrations by a factor of 8–15 for both species. FDEP (2013) attribute this over-estimation to the model's inability to capture the decrease in GOM and PBM concentrations at night due to potential errors in emissions speciation, Hg chemical conversion, or removal processes. Other atmospheric models have tended to overestimate GOM and PBM concentrations as well (FDEP 2013). Nonetheless, the reasonably good performance of the TMDL CMAQ modeling for wet and dry deposition provides confidence in the estimates of Hg loading to the Everglades.

The Hg source apportionment scheme applied in the TMDL CMAQ modeling analysis showed that over 99% of the 25 μg/m^2-yr wet deposition flux at ENP was due to non-Florida sources, with most of that from the global background (FDEP 2013). Similarly, the modeling showed that only 1% of the approximately 17 μg/m^2-yr dry deposition flux at ENP was due to Florida sources. The total (i.e., wet +

dry) Hg deposition at ENP in 2009 was 42 μg/m^2-yr. Of this amount, less than 1% was contributed by Florida sources and over 99% was due to sources outside the state, primarily from outside the U.S.

The PMF analysis of Hg wet deposition data at the ENP site identified four significant factors contributing to Hg wet deposition (FDEP 2013). The most important factor was identified as a combined oil combustion/industrial source contributor, with others representing a crustal source, a marine source, and waste incineration.

In summary, the Florida state-wide Hg TMDL study concluded that Hg wet deposition is especially important in Florida because of the high frequency of convective storms (thunderstorms) in the Everglades and other parts of Florida, especially in summertime, and due to the large size of these weather systems. Convective storms can climb more than 16 km which allows the removal of GOM that has been formed from GEM by oxidation in the free troposphere after long-range transport from outside Florida; thus, the wet deposition often represents the Hg in a very large volume of the atmosphere (FDEP 2013). Additionally, thunderstorms can produce high speed winds pulling in still more volumes of air vertically from which the rain, or hail, removes atmospheric pollutants. Also, as noted below, thunderstorms may increase Hg concentrations in non-convective precipitation by mixing GOM down to lower altitudes where it can be scavenged by all precipitation types. Across Florida, thunderstorms are more common in inland areas by about 20%, and across coasts to inland areas thunderstorms occur on average of 80–100 days per year (FDEP 2013). The scale of rain from summertime thunderstorms in the Everglades often exceeds 7.6 cm in an hour and enables Hg to be scavenged rapidly from the atmosphere.

5.5.2 Mercury Deposition Source Attribution from Other Modeling and Monitoring Studies

Over the past three decades numerous other modeling and monitoring studies have attempted to explain the high wet deposition observed in the Everglades and the rest of southern Florida and to identify the relative contributions of various emission source groups and regions to both wet and dry deposition of Hg in the region.

As noted previously (Chap. 4, this volume), a hybrid air quality modeling approach was used in the Everglades Hg Pilot TMDL Study (FDEP 2003) to show that dry deposition was an important component of Hg deposition in the Everglades during June 1995 to June 1996, comprising 34–40% of the total Hg deposition. Dry deposition was dominated by the GOM fraction and showed a seasonal trend, with relatively greater deposition occurring during the climatological wet season. FDEP (2003) also concluded that the relatively modest decreases in measured volume-weighted mean Hg concentrations in rainfall from 1993 to 2000 at the Everglades National Park Beard Research Center agree reasonably well with the Hg emissions declines in Dade, Broward and Palm Beach counties during that time period. Two key issues relevant to Hg deposition were not addressed in the Pilot TMDL study and were recommended for follow-on work: (1) the inclusion of the global-scale contribution of Hg to deposition in the Everglades was considered outside the scope of the study due to paucity of data or models at the time; and (2) Hg species transformations in the atmosphere needed to be better characterized (FDEP 2003).

The FAMS study (see Chap. 4, this volume) allowed for the estimation of Hg deposition throughout Florida during 1992–1996 using precipitation and concentration monitoring (Guentzel et al. 2001). Most of the Hg wet deposition occurred during the summertime and Hg concentrations in precipitation were found to be significantly higher in southern Florida than at the north-central and north-western sites. Also, Hg concentrations in precipitation in southern Florida were similar at the urban and rural sites. In contrast, concentrations of related trace metals such as V, Ni, Cu, Zn, and Pb in precipitation were 20–80% higher at the urban sites. Guentzel et al. (2001) concluded that Hg was scavenged from the global pool in the free troposphere. They used box model calculations to hypothesize that long-range transport of GOM (referred to as reactive gaseous mercury or RGM by the authors), coupled with strong convective thunderstorm activity during the summertime, represents >50% of the Hg deposition in southern Florida. Their box model calculations also suggested that local anthropogenic GOM and particulate Hg emissions accounted for 30–46% of the summertime rainfall Hg deposition across southern Florida peninsula during 1992–1996 and the remainder from the global background.

The South Florida Atmospheric Mercury Monitoring Study (SoFAMMS) was a short-term study conducted from 6 August to 6 September 1995 to examine the potential impacts of local anthropogenic sources on Hg in precipitation in southern Florida using highly resolved sampling in both space and time (Dvonch et al. 1998). The study hypothesized that the large spatial and temporal variations in observed Hg concentrations in precipitation, which could not be explained by precipitation depth alone, indicated large impacts from local sources. As part of subsequent analyses, daily event precipitation sample data were used with a receptor modeling approach to investigate sources of Hg wet deposition in south Florida. A multivariate receptor modeling approach based on principal component factor analysis (PCA) estimated that municipal waste incineration accounted for 57% and oil combustion for 14% of the Hg wet deposited at five Florida Everglades sites located near or just within the eastern edge of the Everglades Protection Area (EvPA) (Dvonch et al. 1999). Dvonch et al. (2005) studied the influence of meteorological conditions on the wet deposition of Hg in south Florida using data collected in Davie, Florida during June 1995 to June 1996. A meteorological tracer analysis, utilizing the ratio of trace elements lanthanum to cerium as a tracer of oil-fired combustion emissions, determined that feed air to precipitation cells arriving at the site incorporated local urban emissions more frequently during the spring and summer seasons. The authors concluded that local anthropogenic sources played a dominant role in the wet deposition of Hg to southern Florida during the period investigated.

Lin et al. (2005) adapted the Biogenic Emission Inventory System (BEIS) for estimating Hg emissions from vegetation across the U.S. using land cover data, modeled meteorology and emission factors. The modeled Hg emissions were evaluated by comparison with evasion fluxes measured in Florida wetlands in June 1997

(Lindberg et al. 2002). The total vegetative Hg emissions in the continental US domain were estimated to be 44 ton/yr during 2001. They were highest in summer and were mainly contributed from the southeastern US. In Florida, they are highest in the southern (including the Everglades) and northwestern parts of the state.

Seigneur et al. (2004) used a nested multiscale modeling system consisting of a global chemical transport model (CTM-Hg) at 8° × 10° horizontal resolution and a continental chemical transport model (the Trace Element Analysis Model, TEAM) at 80–100 km horizontal resolution to simulate the chemistry, transport and deposition of anthropogenic, natural and re-emitted Hg over the continental US in 1998 (the horizontal resolution of TEAM varies with latitude and was, in this application, ~84 km over southern Florida). The models were evaluated against MDN wet deposition data and ambient Hg concentrations and subsequently used to conduct a source attribution analysis for 19 ecologically-sensitive receptor areas including three in Florida. The modeled contribution of North American anthropogenic emissions to Hg deposition at the Everglades National Park is 20%. The estimated contribution of natural sources at the ENP is 25% and the contribution of Hg emissions from anthropogenic sources in other continents exceeds 50%. Carlton et al. (2004) reported results from an application of the same model, TEAM, at a finer horizontal resolution (15–20 km). U.S. anthropogenic sources were modeled to contribute 8% to deposition in the Everglades in 2004. The USEPA (USEPA 2005) applied the CMAQ model at 36 km horizontal resolution with boundary conditions from GEOS-Chem for 2001 as part of Clean Air Mercury Rule (CAMR) modeling. They determined that global sources (i.e., those outside North America) contribute >85% to Hg deposition in southern Florida.

During the Florida Everglades Dry Deposition Study (FEDDS), Marsik et al. (2007) made measurements of Hg dry deposition during February – March 1999 and June 2000 using a surrogate water surface technique and compared these with modeled estimates of dry deposition using a single-layer inferential model. The average daily dry deposition fluxes measured during the 1999 and 2000 periods were 13.3 and 5.9 ng/m^2-day, respectively, while the modeled fluxes were lower, 3.4 and 1.8 ng/m^2-day, respectively. The authors hypothesize that the reductions in dry deposition from 1999 to 2000 could be due to reductions in waste incineration emissions in the interim period and/or to more wet removal in the summer period of 2000 compared to the winter of 1999. The authors note that the dry deposition of Hg is likely to be important across south Florida during the months of October–May, when precipitation is relatively infrequent. They attribute the discrepancy between measured and modeled estimates of Hg dry deposition to one or more of the following reasons: (1) errors in the measurement of ambient concentrations of GOM and PBM; (2) uncertainties in the surrogate water surface method employed to measure dry deposition; and (3) inaccuracies in the description of the resistances to uptake of GOM and PBM by the canopy.

Selin et al. (2007) applied GEOS-Chem at 4° × 5° horizontal resolution to simulate Hg deposition in North America and other continents and successfully validated the model against observed Hg air concentrations and MDN wet deposition data in 2003. They attribute the observed maximum MDN wet deposition flux in the southeastern U.S. to photochemical oxidation (by ozone or hydroxyl (OH) radicals) of the global GEM pool and frequent precipitation. They estimate that North American anthropogenic sources contribute, on average, less than 10% to wet plus dry deposition in southern and central Florida. Selin and Jacob (2008) applied an updated version of GEOS-Chem to simulate Hg deposition in 2004–2005. The model reproduced well measured Hg air concentrations and both the seasonal cycle and latitudinal gradient in the MDN wet deposition data (maximum in summer and increasing in amplitude from north to south). The authors interpret the seasonal cycle in the MDN data as largely driven by the global pool of Hg and attribute the high observed summertime Hg wet deposition in the Southeast (and, in particular, Florida) to the interaction of global-scale subtropical downwelling, which supplies elevated divalent mercury (GOM) in subsiding air masses, with frequent regional deep convection, which scavenges this free tropospheric GOM from high altitudes. They estimate that North American anthropogenic sources contribute, on average, 10–15% to wet + dry deposition in southern Florida. Vijayaraghavan et al. (2007) report that observed Hg wet deposition fluxes show a clear north-to-south gradient across the U.S. with greater deposition in Florida. This pattern is unlike observed sulfate wet deposition fluxes that show larger values in the northeast (near large emission sources) than the southeast, thus suggesting that sulfate wet deposition is more strongly influenced by local/regional emission sources than Hg.

There is an increased need for improved ambient measurements, including Hg speciation, as well as model sophistication towards reconciling model simulations and observed Hg chemistry and deposition. Sillman et al. (2007) applied CMAQ with modifications to include an integrated solution for gas phase and aqueous Hg photochemistry and expanded atmospheric chemistry over a model domain with 36 km horizontal resolution over the eastern half of the U.S. and parts of Canada, Mexico and the Caribbean. The model was applied for 15 days in June 2000 and reproduced the overall patterns in aircraft measurements in south Florida that show GOM varying between 10 and 230 pg/m^3 in the troposphere and increasing with altitude. However, the authors note that the model under-predicts the maximum observed GOM by a factor of two and, that if the reservoir of elevated GOM in the upper troposphere predicted by Selin et al. (2007) had been included here, this might have resulted in higher modeled GOM that would be closer to the observations. Gridded Hg models such as CMAQ dilute the mass of Hg emitted over the volume of each model grid cell. Vijayaraghavan et al. (2008) reported better model predictions with CMAQ for Hg wet deposition at MDN monitoring stations in the U.S. when applying a plume-in-grid treatment (advanced plume treatment or APT) that initially models the chemistry and transport of Hg in the puffs released from elevated point sources before transferring the mass to the grid farther away from the source. Baker and Bash (2012) modeled Hg deposition over the U.S. at 12 km horizontal resolution with CMAQ and the Comprehensive Air Quality Model with extensions (CAMx) (Ramboll 2018) and concluded that speciated ambient observations that provide more insight into air-surface exchange and an increased understanding of atmospheric Hg chemical processes will further constrain model parameterizations and improve model performance.

Using a box model for the marine boundary layer (MBL), Holmes et al. (2009) concluded that a major source of Hg deposition in the MBL is from the oxidation of GEM transported in the free troposphere over long distances to GOM by bromine atoms and subsequent deposition after uptake onto sea-salt aerosols. The incorporation of a halogen chemical mechanism that includes bromine-induced gas-phase oxidation of GEM in CMAQ by Ye et al. (2018) resulted in improved model predictions of observed GOM and PBM concentrations and wet and dry deposition in the eastern U.S. Through measurements of vertical profiles of bromine monoxide (BrO) radicals, GEM and GOM over the Gulf coast of Florida, Coburn et al. (2016) hypothesize that the oxidation by bromine radicals is the dominant pathway for GEM oxidation to form water-soluble GOM (with less than a 5% increase due to chlorine-induced oxidation) that is subsequently available for wet scavenging by thunderstorms or transport to the boundary layer. Thus, the literature indicates that the oxidation of GEM aloft is driven by a larger natural than anthropogenic component.

Holmes et al. (2016) used rainwater samples from over 800 individual precipitation events and radar and satellite observations at seven locations across the eastern U.S., including one in the Florida panhandle, to show that thunderstorms increase Hg concentrations by 50% relative to weak convective or stratiform events of equal precipitation depth due to the strong convection reaching the upper troposphere where GOM resides. The authors also note that thunderstorms may also raise baseline Hg concentrations in non-convective precipitation by mixing GOM down to lower altitudes where it can be scavenged by all precipitation types. Using aircraft measurements of GOM and total Hg air concentrations in 2013 and the GEOS-Chem model, Shah et al. (2016) showed that high summertime GOM concentrations in the free troposphere in the southeastern US were associated with clean subsiding air masses originating in the upper troposphere within the Pacific or Atlantic anticyclones. The anticyclones are characterized by large-scale sinking motion which transports higher GOM concentrations (> 300 pg/m^3) from the upper troposphere due to the fast oxidation of GEM resulting from higher Br concentrations. The sinking air in the anticyclones also suppresses cloud formation and precipitation, thereby preventing loss of GOM by reduction and wet deposition (Shah et al. 2016). The authors conclude therefore that the transport of GOM produced in the Pacific anticyclone could be an important source of GOM deposition over the southeastern U.S. if these high GOM air masses are exposed to deep convection.

5.6 Conclusions

In summary, the historical Hg deposition to the Everglades until the mid-1990s was likely due to a combination of emissions from local sources with a high oxidized Hg speciation fraction (e.g., >75%) and short stacks such as waste incinerators, as well as regional and more distant sources. Local anthropogenic GOM and particulate Hg emissions accounted for 30–46% of the summertime rainfall Hg deposition across southern Florida peninsula during 1992–1996 with the remainder from the global background. Despite reductions in local emission sources since that time, the continuing high deposition of Hg to the Everglades appears to be largely due to the oxidation of gaseous elemental Hg in the free troposphere (originating from regional and global emission sources) to water-soluble gaseous divalent Hg by bromine and other oxidants followed by: (1) scavenging during deep convective activity and subsequent wet deposition especially during summertime thunderstorms in the Everglades; or (2) transport to the boundary layer and subsequent dry deposition or wet deposition during non-convective precipitation. By the late 1990s and early 2000s, due to local source emission reductions, over 99% of the 25 μg/m^2-yr wet deposition flux at ENP was due to non-Florida sources, with most of that from the global background (FDEP 2013). Similarly, computer modeling from the Florida Hg TMDL study (FDEP 2013) showed that only 1% of the approximately 17 μg/m^2-yr dry deposition flux at ENP was due to Florida sources. The total (i.e., wet + dry) Hg deposition at ENP in 2009 is 42 μg/m^2-yr. Of this amount, less than 1% is contributed by Florida sources and over 99% is due to sources outside the state, primarily from outside the U.S.

More research is needed to identify missing Hg emission sources, if any, in current southern Florida emission inventories and to improve our understanding of Hg dry deposition velocities and atmospheric Hg chemistry and scavenging over the Everglades, all of which affect the analysis of Hg deposition and biogeochemical cycling in the region.

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