Chapter 4
Atmospheric Deposition Flux of Mercury to the Everglades

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Abstract This chapter presents a summary of data on atmospheric mercury (Hg) deposition under precipitating and non-precipitating conditions within and near the Everglades from monitoring data and other estimates and a comparison with data from elsewhere in the United States. Hg deposition to the Everglades is among the highest in the country and has continued to be so over the past two decades.

Keywords Mercury · Wet deposition · Dry deposition · Spatial variability · Oxidation states · Temporal trend

4.1 Introduction

The deposition of inorganic mercury (Hg) from the atmosphere to the Earth's surface represents a significant fraction (over 95%) of the external loading of Hg to the Florida Everglades (USEPA 1996; Guentzel et al. 1998; FDEP 2003, 2013). Atmospheric deposition may occur under both wet and dry conditions and is influenced by precipitation patterns and other meteorological conditions, the presence of emission sources, local terrain and land cover, atmospheric concentrations and transformations of Hg, and long-range atmospheric transport of Hg. As discussed in Chap. 3 (this volume), Hg in both atmospheric emissions and the ambient atmosphere can be present in a variety of oxidation states and phases, which in turn exhibit varying deposition characteristics. Due to its limited solubility in water, Hg(0) (gaseous elemental Hg or GEM) has negligible wet deposition. The wet deposition of Hg primarily consists of gaseous divalent Hg(II) (gaseous oxidized

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Hg or GOM) which is highly soluble in water, with a smaller fraction of particulate-bound divalent Hg [PBM, also referred to as Hg(p)]. GEM undergoes dry deposition under non-precipitating conditions but a portion is re-emitted back to the atmosphere. GOM dry deposits rapidly, especially near emission sources, as does coarse PBM, while fine particulate PBM deposits more slowly. These chemical forms of Hg also inter-convert as discussed in Chap. 3 (this volume) and thus the deposition of Hg is dependent on emissions and concentrations of all forms of Hg. While most atmospheric deposition of Hg is due to the inorganic chemical forms discussed above, there is also a relatively small fraction of methylmercury (MeHg) present in wet deposition.

The atmospheric deposition flux of Hg in southern Florida is among the highest in the United States (see next section). Several measurement campaigns and other studies have increased our knowledge of the deposition of atmospheric Hg to the Everglades. The findings from these networks and studies are discussed below in the context of wet and dry deposition flux of Hg to the Everglades together with a comparison to deposition in other geographic regions, in Florida and elsewhere in the United States.

4.2 Wet Deposition

Hg in precipitation has been measured at monitoring stations in the Mercury Deposition Network (MDN) in North America for over 20 years. A component of the National Atmospheric Deposition Program (NADP), the MDN is the only network providing a long-term record of Hg rain concentration and wet deposition in the U.S. and Canada (NADP 2017). All MDN sites follow standard procedures and have uniform precipitation chemistry collectors and gauges with weekly samples. The network has over 100 monitoring stations in the U.S and Canada (Fig. 4.1) with five stations in Florida active in 2016 including the following three located within or near the Everglades:

- Everglades National Park – Beard Research Center (MDN site ID FL11), active since March 1996 and operated by the Everglades National Park.
- Everglades – Western Broward County (MDN site ID FL97), active since November 2006 and operated by the South Florida Water Management District.
- Everglades – South Palm Beach County (MDN site ID FL95), active since April 2015 and operated by the South Florida Water Management District.

The measured annual Hg wet deposition fluxes in 2016 at MDN stations across the U.S. and Canada are shown in Fig. 4.2. The map contour surface represents a gridded interpolation developed by the NADP (2017) using discrete MDN wet deposition measurements, terrain, a precipitation grid derived from NADP precipitation observations, and a national precipitation grid developed by the PRISM (Parameter-elevation Regressions on Independent Slopes Model) Climate Group (PRISM 2017). The three stations near the Everglades experience much higher Hg

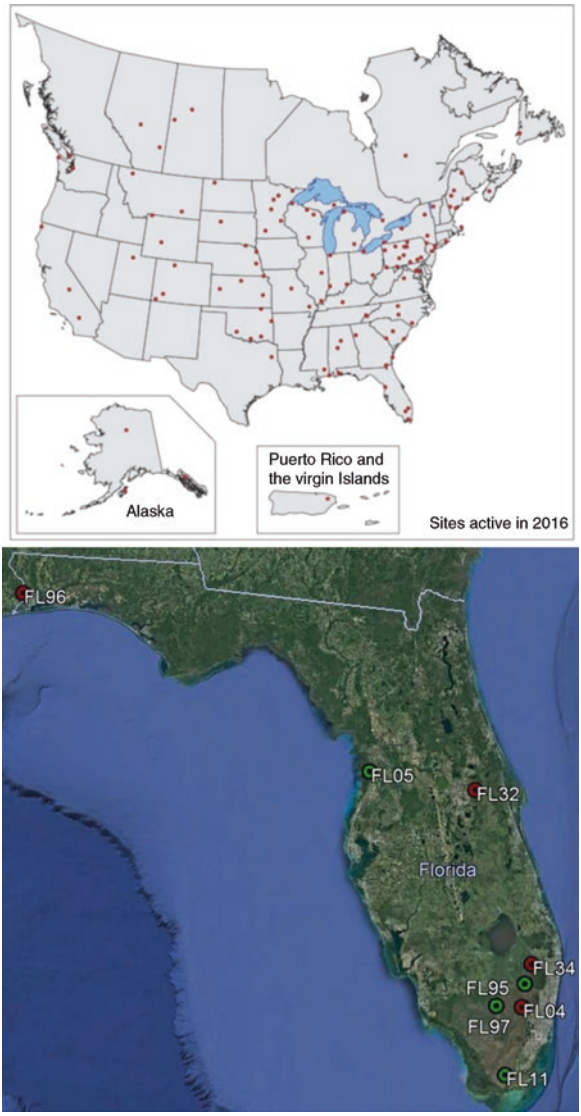

Fig. 4.1 Locations of active MDN Hg wet deposition monitoring stations in 2016 (top) and of all MDN stations historically operated in Florida (bottom) (in bottom panel, green circles = active stations and red circles = inactive stations). (From NADP 2017)

wet deposition (ranging from 17.6 to 25.8 μg/m^2-yr) than most parts of the U.S. with deposition more than double that observed in the western and northeastern U.S. This spatial pattern has been observed for more than 20 years across the MDN network (e.g., Prestbo and Gay 2009). In particular, for the past 20 years, Hg wet deposition has been high at MDN stations in southern Florida with values regularly near or exceeding 18 μg/m^2-yr (Fig. 4.3). In general, some of the highest wet deposition in the country occurs along the Gulf of Mexico; this phenomenon has been attributed to a combination of regional emissions (FDEP 2003) as well as long-

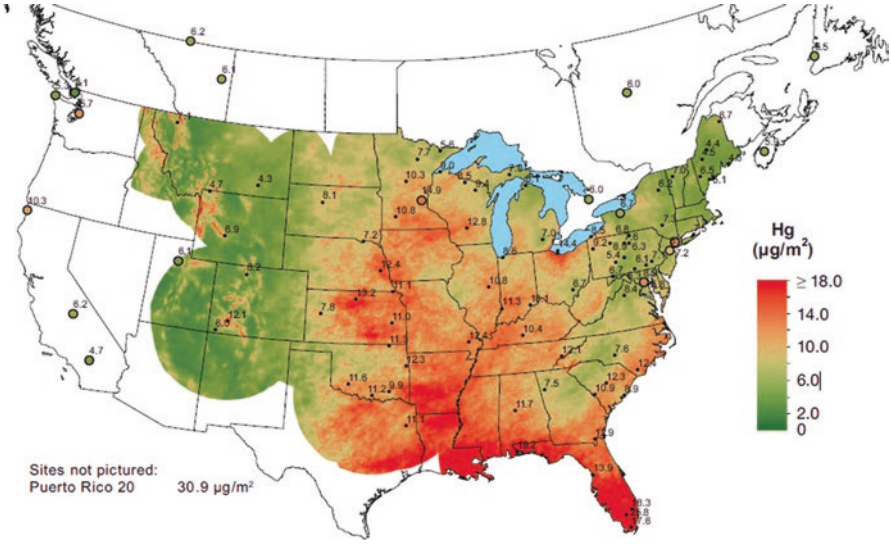

Fig. 4.2 Annual measured Hg wet deposition flux in 2016. (From NADP 2017)

range transport of Hg, high thunderstorm frequency and total rainfall, along with high atmospheric convection during thunderstorms and scavenging of GOM from the free troposphere where high atmospheric concentrations of GOM are evident (FDEP 2013; Nair et al. 2013; Holmes et al. 2016), as well as subsidence of Hg containing air from the free troposphere (Gustin et al. 2012; Holmes et al. 2016; Huang et al. 2017). The high frequency of convective storms (thunderstorms) and the large size of these weather systems in south Florida (storms that can climb in excess of 16 km) allows stripping of atmospheric constituents, including GOM, from these great vertical columns. The idea that convective storm scavenging of high concentrations of GOM in the free troposphere could at least in part be responsible for the fact that wet deposition in south Florida was both elevated and showed no significant east-west gradient moving downwind from the urban fringe along the eastern Florida coast was first proposed by Guentzel et al. (2001). Guentzel et al. predicted that it would only take about 50 pg/m^3 of GOM in the free troposphere upwind from southern Florida to account for the rainfall Hg deposition they observed across southern Florida. This hypothesis subsequently was validated by measurements of GOM obtained by aircraft sampling over the Bahamas which found up to 350 pg/m^3, more than enough to account for the rainfall Hg across the Everglades (Landis et al. 2005).

The prevalence of Hg wet deposition in the Everglades and elsewhere in southern Florida has been investigated in several studies. The Florida Hg Pilot Total Maximum Daily Load (TMDL) study (FDEP 2003) was conducted to evaluate the technical feasibility of calculating an atmospherically driven TMDL for Hg for the Florida Everglades, and the analysis was conducted for a site within the Water Conservation Area 3A (site 3A-15) portion of the Everglades considered to be a “hot-spot” with

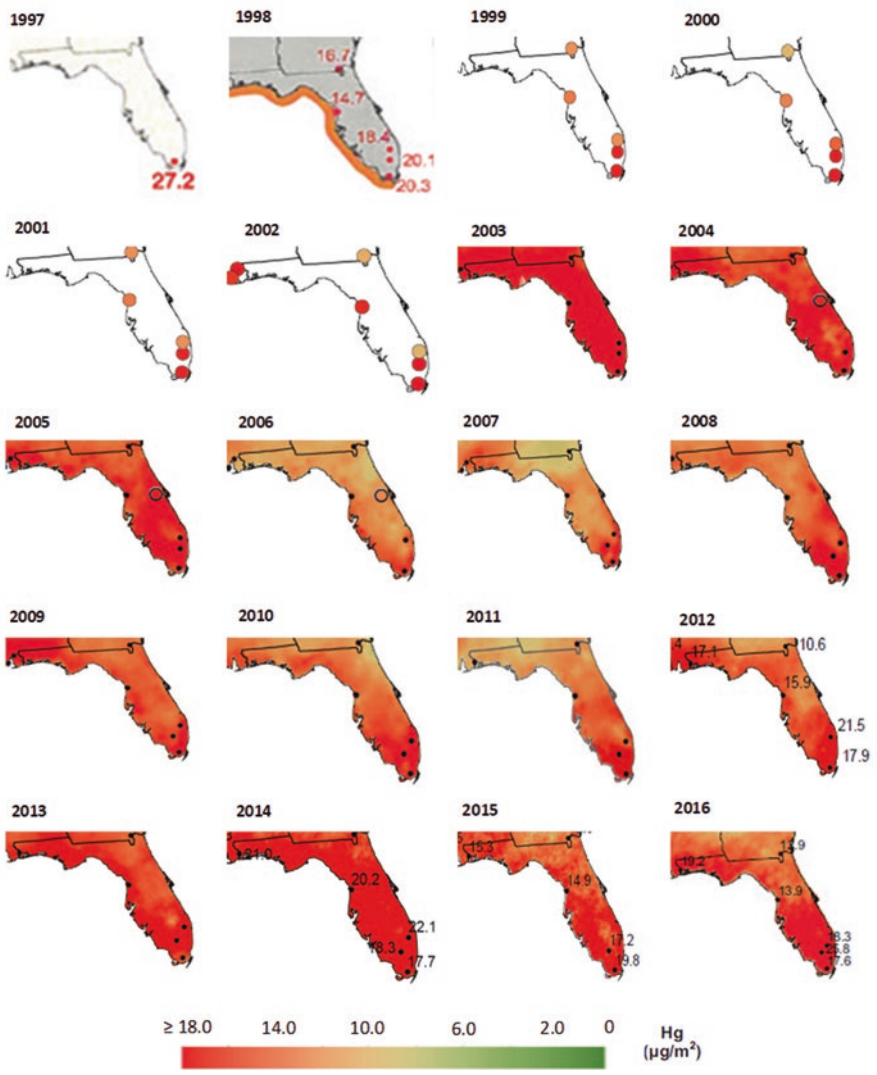

Fig. 4.3 Annual Hg wet deposition flux in Florida from 1997–2016. (From NADP 2017)

respect to fish Hg concentrations. The study incorporated extensive field data into a framework combining atmospheric mercury deposition and aquatic mercury cycling models to demonstrate the feasibility of the approach. The goal was to simulate how changes in local atmospheric mercury emissions in south Florida would influence mercury concentrations in top predator fish, thus demonstrating the potential of combining air and water modeling approaches in TMDLs involving contaminants with an important atmospheric cycling and deposition component such as Hg.

Estimates of atmospheric loading rates of Hg via wet deposition in the Pilot TMDL were obtained from three sources:

1. Measurements of wet deposition at three sites in the Everglades region obtained during the Florida Atmospheric Mercury Study (FAMS) monitoring program conducted between 1993 and 1996 (Guentzel et al. 1995, 2001; Landing et al. 1995; Pollman et al. 1995).
2. Measurements of wet deposition at three MDN sites in south Florida between late 1995 and 2003 (sites FL11, FL04 (Andytown) and FL97 shown in Fig. 4.1).
3. Modeled estimates of wet deposition derived from source-receptor modeling conducted by the University of Michigan Air Quality Laboratory (UMAQL) for the FAMS monitoring period.

During the FAMS program, measurements were made for bulk and wet Hg deposition, particulate-associated mercury and total gas-phase mercury (TGM) at four sites near the Everglades and five other sites in Florida (the panhandle, north-central Florida, a marine background site and two sites in the southwestern peninsula). The FAMS sites used for analysis in the Pilot TMDL were those at Tamiami Trail Ranger Station and Andytown in Florida.

FDEP (2003) combined the Hg wet deposition fluxes, rainfall depth and rain concentrations from the MDN monitor at FL11 from 1996 to 2002 and the equivalent monitor in the FAMS study during 1993–1996 to form a period of record of eight full years. By definition, wet deposition flux is very closely related to rainfall depth and most of the variance in the deposition flux is related to the variance in rainfall depth. Nonetheless, an analysis of volume weighted mean (VWM) concentrations in precipitation revealed that these concentrations declined by approximately 25% (3 ng/L) between 1994 and 2002 due to factors other than seasonal dynamics and precipitation. FDEP (2003) concluded that the relatively modest decreases in VWM concentrations agrees reasonably well with the Hg emissions declines in Dade, Broward and Palm Beach counties from 1993 to 2000.

Axelrad et al. (2005, 2007) subsequently examined whether there had been a continuing decline in atmospheric deposition of mercury in the Everglades beyond 2002 by extending the period of record through 2004. Their analysis showed that an increase in mercury wet deposition and annual VWM concentrations from early 2003 through mid-2004 essentially negated the overall declines that had been observed previously from late 1993 through 2002. Axelrad et al. (2007) also concluded that there was no statistically significant trend in atmospheric deposition during 1994–2005 in south Florida. In a study of regional trends in Hg wet deposition at MDN stations from 1998–2005, Butler et al. (2008) concluded that there was no statistically significant trend in Hg wet deposition or VWM concentration at the FL11 Everglades monitoring station during this period. Similarly, using the nonparametric seasonal Kendall trend test, Prestbo and Gay (2009) determined that there was no statistically significant trend in Hg wet deposition or VWM concentration between 1996 and 2005 at the two stations near the Everglades (FL11 and

FL04). Chapter 1 of Volume III re-examines the issue of recent trends in wet deposition in the Everglades between November 1993 through June 2016 based on FAMS data for FL11 coupled with MDN data. That analysis indicates that the Hg signal declined from late 1993 through mid-2013 followed by a relatively sharp increase through mid-2016.

One feature of the weekly mercury deposition record at the MDN sites near the Everglades is the occurrence of individual weeks with unusually high Hg deposition (Prestbo and Gay 2009). Among all MDN sites in the country, the Everglades station at FL11 shows the highest percent (7%) of its weeks with heavy deposition (with heavy deposition defined here as having annualized wet deposition fluxes greater than 1.5 μg/m^2-yr), reflecting the strong seasonal convective precipitation in this region.

As part of the state-wide Florida mercury TMDL study conducted in 2009 and 2010 (FDEP 2013), the UMAQL measured Hg wet deposition at six monitoring sites across Florida (Fig. 4.4) including one at the ENP at the Beard Research Center (lat. 25.3898, long. -80.6803). Operational logistics of the station were coordinated with ENP staff, with precipitation monitoring beginning in November 2008 and continuing through the end of August 2010. Strong seasonal variability was observed in VWM concentrations as well as Hg wet deposition with increases during summer months (Fig. 4.5), with the sharply accentuated summertime depositional fluxes reflecting higher precipitation volumes coupled with higher VWM concentrations. The annual Hg wet deposition measured at ENP in 2009 was 25 μg/m^2-yr, higher by 2–70% than the other five TMDL sites in Florida.

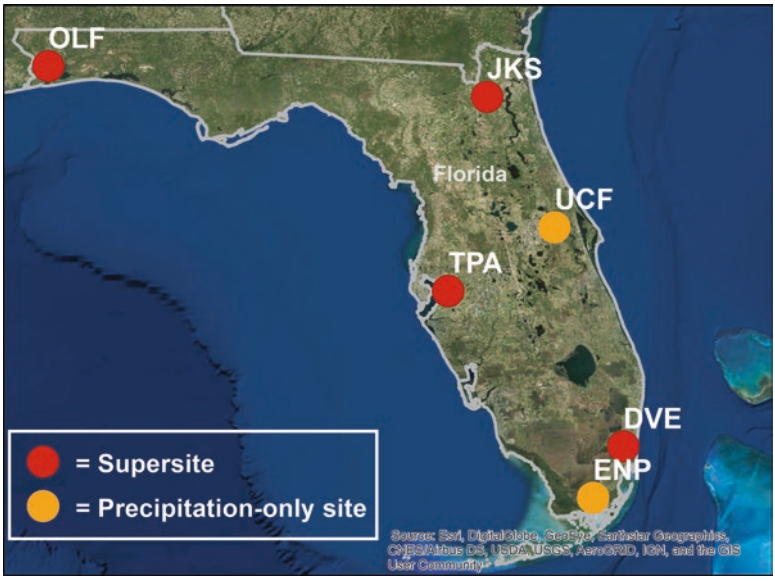

Fig. 4.4 Locations of Hg wet deposition monitoring stations during the 2009 Florida mercury TMDL study. (From FDEP 2013)

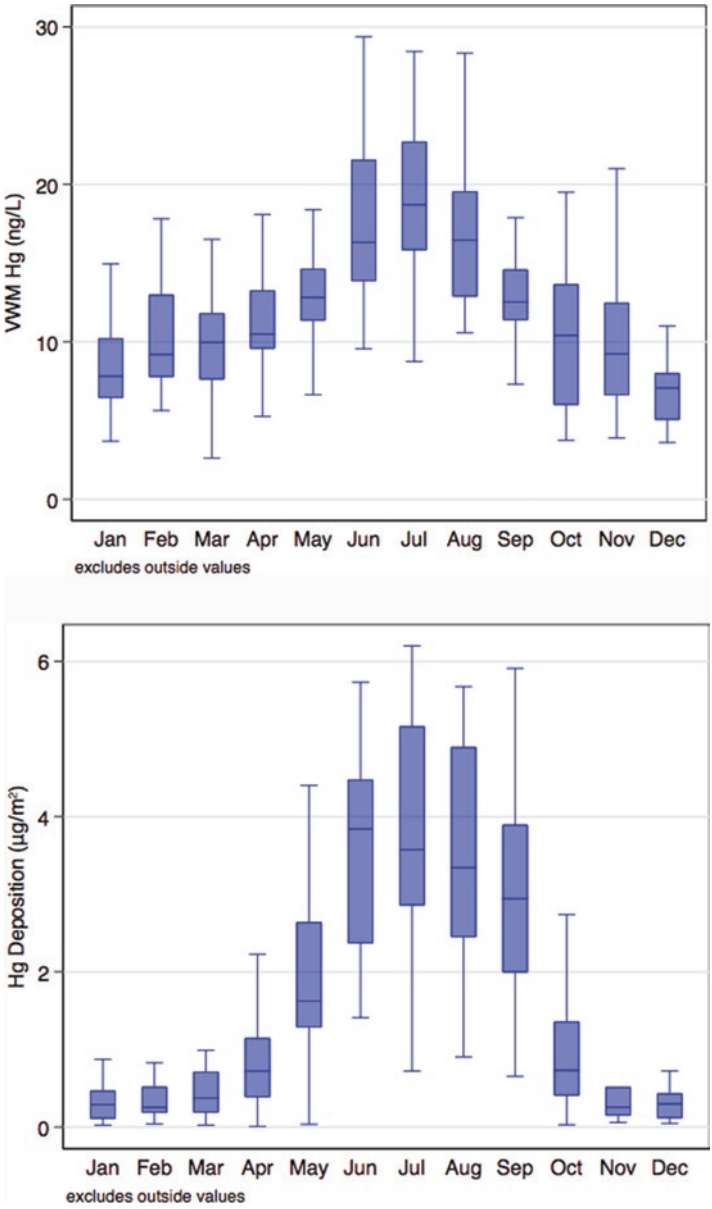

Fig. 4.5 Box plots of the distribution of monthly volume weighted mean Hg concentration in wet deposition (top) and Hg wet deposition flux (bottom) at the Everglades (ENP) monitoring site, November 1993 through June 2016. (Data from FAMS (Guentzel et al. 2001; NADP 2017))

The discussions of Hg wet deposition above have been for inorganic Hg. Measurements of MeHg deposition are much more scant than those of inorganic Hg. MeHg is measured at a few MDN stations none of which are in Florida. The concentration of MeHg in summertime precipitation measured at south Florida sites during FAMS ranged from <0.005 to 0.020 ng/L based on limited wet and bulk deposition measurements conducted during the FAMS study (Guentzel et al. 1995). These values were 2–3 orders of magnitude lower than corresponding inorganic Hg wet deposition. The FAMS study concluded that deposition of MeHg may not be a significant source of MeHg in south Florida's aquatic environments.

4.3 Dry Deposition

While wet deposition can be well quantified through the collection and analysis of precipitation, direct measurements of dry deposition are difficult to make and have large uncertainties (e.g., Lyman et al. 2007; Zhang et al. 2009, 2012; Lai et al. 2011). FDEP (2013) provide a succinct explanation of the importance of dry deposition measurements in Florida that are applicable to the Everglades. "The dry deposition mercury speciation and continuous measures are important in understanding the specifics and dynamics of mercury cycling within Florida. Atmospheric dry mercury is stripped by forests in leaf and needle uptake as well as in resistance knocking *(removing)* mercury from the air to the forest floor". The atmospheric dry deposition of Hg to prairie, shrub, and wetland plants may be a critical avenue of entry into food webs, and a potential first step of Hg being bound to organic matter and eventually entering aquatic systems.

Although measurements of Hg dry deposition are scarce to this day and measurements of wet deposition were uncommon in the Everglades prior to the 1990s, cores of lake sediments and bog peat have been used to document historical changes in atmospheric Hg deposition, both wet and dry. Analyses of sediment records in the Everglades indicate an increasing trend in total deposition up through the decade of the 1980s (Rood et al. 1995; Delfino et al. 1993; see Chap. 1, Vol. III for a review of these studies). Pollman et al. (2007) inferred historic atmospheric fluxes of Hg over a 200-year period from 1800–2000 using sediment cores collected at site F3 in Water Conservation Area (WCA-2A) (Fig. 4.6) using unpublished data from Abelak et al. provided through T. Atkeson (FDEP). There is a clear increasing trend in total deposition over the 200-year period till the 1980s followed by a reduction in total deposition.

A modeled analysis of dry deposition conducted during the Florida Pilot TMDL (FDEP 2003) suggests that dry deposition is an important component, comprising perhaps 34–40% of the total Hg deposition signal. Dry deposition to the Everglades

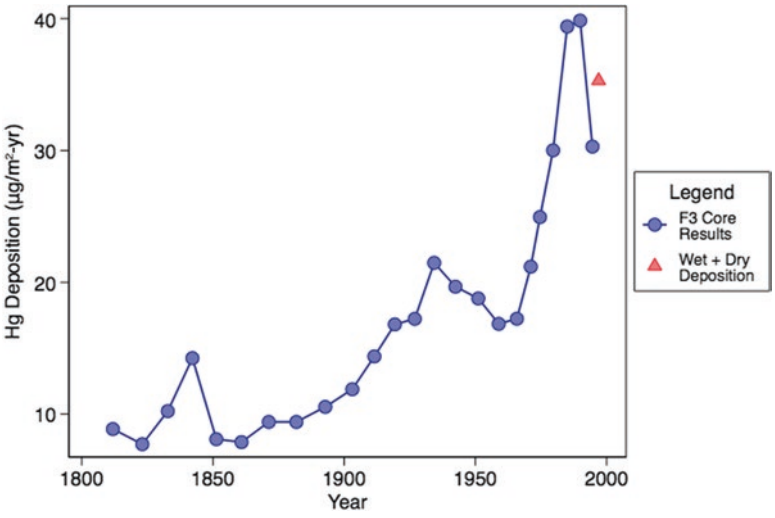

Fig. 4.6 Inferred atmospheric fluxes of Hg from sediment core collected at site F3 in WCA-2A. Red triangle shows for comparison the estimated total deposition flux (wet and dry) of Hg based on FAMS measurements of wet deposition and dry deposition fluxes modeled by Keeler et al. (2000). (Data from Abelak et al. (unpublished data). Redrawn from Pollman et al. (2007))

WCA 3A-15 was estimated using a hybrid model system comprising the Hybrid Single Particle Lagrangian Integrated Trajectories Model, version 4 (HYSPLIT_4) and the Regional Atmospheric Modeling System (RAMS) (FDEP 2003). Dry deposition due to local sources was estimated to be 12.2 ± 2.0 µg/m^2-yr (+1 standard deviation) during a 12-month period from June 1995 to June 1996. While considerable variability was evident in the monthly deposition estimates, on average, dry deposition to the site showed a seasonal trend, with relatively greater deposition occurring during the climatological wet season. As was the case for the wet deposition to WCA 3A-15, dry deposition to this area was dominated by the GOM fraction (FDEP 2003).

Atkeson et al. (2005) combined dry deposition estimates modeled by Keeler et al. (2000) for 22 June 1995 through 21 June 1996 with wet deposition measured at FAMS sites during the same period to obtain a total deposition flux of 35.3 µg/m^2-yr. Pollman et al. (2007) estimated that the dry deposition flux in 2000 at ENP was 10.1 µg/m^2-yr using the measured wet deposition flux of 19.9 µg/m^2-yr at the Beard Research Center (FL11) MDN site in ENP and assuming that the dry:wet deposition ratio of 0.528 used by Atkeson et al. (2005) had remained constant.

During the Florida statewide mercury TMDL study (FDEP 2013), an inferential model was used to estimate Hg dry deposition (Marsik et al. 2011) using surface measurements of ambient speciated Hg and meteorological variables at four "supersites" (Pensacola, Jacksonville, Tampa, and Davie) for 14–18 months from 2009 to

2010. The annual dry deposition flux at Davie was estimated to be 31 μg/m^2-yr, lower than the values in central and northern Florida (FDEP 2013), but higher than the value estimated above for the ENP.

4.4 Summary

The robust long-term record of atmospheric wet deposition of Hg to the Everglades from the NADP Mercury Deposition Network indicates that inorganic Hg wet deposition in this region has continued to be among the highest in the country over the past two decades. In particular, measurements at the Beard Research Center at the Everglades National Park show annual wet deposition exceeding 18 μg/m^2-yr (and often higher than 20 μg/m^2-yr) during most of the time since monitoring began in 1996. These high values have also been confirmed in periodic monitoring campaigns such as the Florida Pilot and State-wide Hg TMDL programs. In contrast, the Hg wet deposition flux over most of the rest of the country is much lower and varies between 5 and 15 μg/m^2-yr. The strong summertime convective precipitation in the Everglades and high concentrations of Hg in precipitation result in individual weeks with unusually high mercury deposition, a characteristic feature of wet deposition dynamics in the Everglades.

Observations of Hg dry deposition are scarce due to challenges in quantifying Hg molecular diffusion and removal; however, the limited measurements (and associated modeling) suggest that the dry deposition signal to the Everglades could be comparable to that of wet deposition. The significantly different deposition characteristics of the different forms of Hg in the atmosphere and the differences in their subsequent methylation characteristics underpin the need to understand not just the magnitude of total deposition to the Everglades but the forms of Hg present in deposition. The dry deposition measurements of Hg are likely an underestimate as they do not typically include dry deposition of GEM that has the potential to be later converted to GOM. Although the deposition of MeHg to the Everglades is likely two orders of magnitude lower than that of inorganic Hg, the organic form needs to be studied as well because it has the potential to directly affect the Everglades without a need for methylation first.

Declines in the wet deposition and Hg concentrations in precipitation in the Everglades in the 1990s appear to be correlated with declines in local Hg emission sources. These trends in Hg deposition are investigated further in Chap. 1, Volume III. The continued high wet deposition to south Florida in the 2000s and 2010s despite significant changes in local and other Florida Hg emissions suggest a strong long-range contribution to Hg deposition in the Everglades. This concept is explored further in Chap. 5 (this volume).

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