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Mercury deposition in Southern New Hampshire, 2006–2009

M. A. S. Lombard¹, J. G. Bryce¹, H. Mao², and R. Talbot³

¹Department of Earth Sciences, University of New Hampshire, Durham, NH, 03824, USA

²Department of Chemistry, State University of New York's College of Environmental Science and Forestry, Syracuse, NY, 13210, USA

³Department of Earth and Atmospheric Sciences, University of Houston, Houston, TX, 77204, USA

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Abstract. The atmospheric deposition of mercury (Hg) occurs via several mechanisms including wet, dry, and occult processes. In an effort to understand the atmospheric cycling and seasonal depositional characteristics of Hg, event-based wet deposition samples and reactive gaseous Hg (RGM) measurements were collected for approximately 3 years at Thompson Farm (TF), a near-coastal rural site in Durham, NH, part of the University of New Hampshire AIRMAP Observing Network. Total aqueous mercury exhibited seasonal patterns in Hg wet deposition at TF. The lowest Hg wet deposition was measured in the winter with an average total seasonal deposition of $1.56 \mu\text{g m}^{-2}$ compared to the summer average of $4.71 \mu\text{g m}^{-2}$. Inter-annual differences in total wet deposition are generally linked with precipitation volume, with the greatest deposition occurring in the wettest year. Relationships between surface level RGM and Hg wet deposition were also investigated based on continuous RGM measurements at TF from November 2006 to September 2009. No correlations were observed between RGM mixing ratios and Hg wet deposition, however the ineffective scavenging of RGM during winter precipitation events was evidenced by the less frequent depletion of RGM below the detection level. Seasonal dry deposition of reactive gaseous Hg (RGM) was estimated using an order-of-magnitude approach. RGM mixing ratios and dry deposition estimates were greatest during the winter and spring. The seasonal ratios of Hg wet deposition to RGM dry deposition vary by up to a factor of 80.

1 Introduction

Mercury (Hg) is a naturally occurring contaminant of global concern due to its toxicity and ubiquitous presence in the atmosphere. It exists in diverse chemical forms comprised of gaseous elemental mercury (Hg^0), reactive gaseous mercury ($\text{RGM} = \text{HgCl}_2 + \text{HgBr}_2 + \text{HgOBr} + \dots$), and particulate mercury (Hg^P). Deposition of atmospheric Hg, mainly the more soluble forms of RGM and Hg^P , is an important source of Hg to terrestrial (Rea et al., 2002; Bushey et al., 2008; Choi et al., 2008; Selvendiran et al., 2008) and aquatic ecosystems (Landis and Keeler, 2002; Ariya et al., 2004). Methylated forms of Hg bioaccumulate in fish, and their consumption is the major exposure route of Hg to humans (Downs et al., 2007).

Previous studies suggest that the magnitude of Hg wet deposition varies geographically and seasonally due to climatic conditions, atmospheric chemistry, and human influences (VanArsdale et al., 2005; Selin and Jacob, 2008; Prestbo and Gay, 2009). In North America seasonal patterns in wet deposition are observed in both depositional flux and concentration with the highest values in the summer and lowest values in the winter (Sorensen et al., 1994; Mason et al., 2000; Guentzel et al., 2001; Keeler et al., 2005; VanArsdale et al., 2005; Choi et al., 2008; Prestbo and Gay, 2009). Explanations for this observation include more effective Hg scavenging by rain compared to snow (Sorensen et al., 1994; Mason et al., 2000; Keeler et al., 2005; Selin and Jacob, 2008), a greater availability of soluble Hg due to convective transport in summer events (Guentzel et al., 2001; Keeler et al., 2005), and a summer increase in Hg-containing soil derived particles in the atmosphere (Sorensen et al., 1994).

Geographic differences in Hg wet deposition may be explained in part by the proximity to atmospheric sources. Results from the National Atmospheric Deposition Program's



Correspondence to: M. A. S. Lombard
(mas3@cisunix.unh.edu)

(NADP) Mercury Deposition Network (MDN) sites in the Northeastern United States exhibit a geographic trend with southern and coastal sites receiving higher Hg concentrations and depositional fluxes (VanArsdale et al., 2005; Prestbo and Gay, 2009). The sites with elevated Hg deposition are nearer to the East coast megalopolis and downwind of anthropogenic emission sources such as coal burning power plants and waste incinerators. Inconsistent results are reported in studies comparing Hg wet deposition fluxes and/or concentrations between rural and urban sites. Some report elevated annual fluxes (Mason et al., 2000) and concentrations (Steding and Flegal, 2002; Engle et al., 2010) at urban locations while others report no significant differences in mean concentrations (Sorensen et al., 1994; Guentzel et al., 2001; Hall et al., 2005). Gaseous evasion of Hg^0 from marine waters is a significant global source of atmospheric Hg and may also contribute to elevated depositional fluxes in coastal regions (Mason and Sheu, 2002). Holmes et al. (2009) suggest that elevated levels of Br in the marine boundary layer are important in transforming Hg^0 to RGM, the more readily deposited gaseous form of Hg.

Like many areas in New England, New Hampshire (NH) air quality is adversely affected by large power plants in the Midwest as well as urban areas located to the south along the East coast of the United States (NHDES, 2004). Two coal combustion power plants are also located in the southern portion of NH and are likely contributors to the local atmospheric load of Hg. Within the waterways of the Northeastern United States, including NH, biological species have been identified as containing elevated Hg levels (Chen et al., 2005; Evers et al., 2007) with atmospheric deposition considered the dominant source in undisturbed watersheds (Chen et al., 2005). Three MDN sites were previously located in NH with sample collection lasting from 7 to 16 months and the most recent sampling terminated in 2005. This lack of Hg wet deposition information was filled using measurements conducted by the AIRMAP program (<http://airmap.unh.edu>) at the University of New Hampshire (UNH). Event-based wet deposition samples were collected over a 36-month time period from July 2006–August 2009. In this study, seasonal and annual variations of Hg wet deposition and concentration from a site in Southern NH are compiled and compared to contemporaneous results from MDN sites in the adjacent state of Maine (ME) and data from three MDN sites previously located in New Hampshire. The purpose of this study is to provide an overview of the seasonal Hg wet deposition patterns at TF, briefly examine meteorological conditions and gas phase indicators of anthropogenic air mass sources in relation to Hg wet deposition, and compare RGM measurements and estimated RGM dry deposition to Hg wet deposition.

Event-based precipitation sampling is necessary to elucidate relationships with meteorological and atmospheric chemical conditions. MDN sites predominantly collect weekly samples, not individual event samples. Results in-

dicate single weekly samples contribute significantly to the annual Hg load (VanArsdale et al., 2005). Collecting samples over pre-determined time intervals can obscure the contribution of single events and relationships with other factors. An event-based sampling site in Underhill, VT (MDN site VT99) reports discrete precipitation events can contribute between 5–17% of the total annual wet deposition (Keeler et al., 2005). The event-based sampling at TF provides the opportunity to evaluate relationships between Hg wet deposition, meteorological conditions and gas phase species.

The more soluble gaseous species, RGM, is thought to be the predominant source of Hg in wet deposition with minor contributions from washout of Hg^P (Schroeder and Munthe, 1998; Guentzel et al., 2001; Sakata and Asakura, 2007; Kieber et al., 2008). Simultaneous measurements of gas phase Hg species and wet deposition offer the opportunity for a more thorough understanding of processes affecting Hg deposition and more accurate estimates of wet and dry deposition. Long-term Hg wet deposition measurements exist at many locations within the United States and Canada as part of the MDN; however, long-term contemporaneous Hg gas phase and Hg wet deposition measurements are lacking (Lindberg et al., 2007; Selin, 2009; Zhang et al., 2009). A recent study (Engle et al., 2010) reports Hg gas phase speciation data, Hg^P , and Hg wet deposition fluxes at nine sites located in the central and eastern United States and Puerto Rico, none of which had data for more than one year. Zhang et al. (2009) provide an overview of the current knowledge regarding the dry deposition of Hg including Hg^0 , RGM, and Hg^P . The limited measurement data that are available for RGM deposition have large uncertainties due to the very low ambient concentration and instrument detection limits, the frequent use of surrogate surfaces in measurement techniques, the small vertical gradients in RGM concentration, and the effects of fast chemical reactions and advection from local sources (Zhang et al., 2009). In this study we use automated continuous RGM measurements over a 35-month time period to generate an order of magnitude estimate of the RGM deposition velocity (V_d) and RGM dry deposition. This is the first multi-year comparison of Hg wet deposition and RGM and provides insights into seasonal variations in Hg deposition pathways.

2 Sample collection and analysis

Precipitation samples were collected at Thompson Farm (TF) (43.11° N, –70.95° W, 24 m elevation) located in Durham, New Hampshire, USA (Fig. 1). The sample site is situated in a rural, residential and agricultural setting immediately surrounded by agricultural fields and mixed hardwood and pine forests. It is approximately 25 km from the Gulf of Maine and 110 km north of the city of Boston. The UNH AIRMAP program maintains and collects numerous atmospheric chemistry measurements at TF (Mao and

Talbot, 2004; Chen et al., 2007; Darby et al., 2007; Mao et al., 2008; Sigler et al., 2009b). Meteorological data used in this study (temperature, solar radiation, precipitation amount) are from the NOAA Climate Reference Network (CRN) site co-located at TF. Information about CRN data measurement and collection techniques is available at <http://www.ncdc.noaa.gov/crn/instrdoc.html>.

Wet deposition samples were collected using a modified Aerochem automated precipitation sampler, identical to samplers used in the MDN. Sample collection bottles were manually changed on a primarily event-based schedule. Trace metal sampling techniques were followed in accordance with EPA method 1669. The sampling train consisted of acid washed polyethylene funnels placed directly into pre-acidified and acid washed fluorinated ethylene propylene (FEP) bottles. Prior to sample deployment, bottle blanks were collected and sample bottles were treated with 1.25 mL of 6 N HCl for sample preservation.

Upon collection, samples were preserved in the original collection bottle with the addition of trace metal grade hydrochloric acid and bromine monochloride to a final concentration of 0.5 %. Samples were analyzed with a Tekran model 2600 dual amalgamation cold vapor atomic fluorescence spectrometer following a modified version of EPA method 1631 recommended in the Tekran user's guide. The average system blank value over all sample analyses was 0.45 ng l^{-1} and the average method detection limit was 0.08 ng l^{-1} as determined by three times the standard deviation of the system blank. The average bottle blank abundance was 0.09 ng. ORMS-3 and ORMS-4 (National Research Council, Canada) were used as external standards and results were within range of the accepted values. Final concentration values were corrected for system and bottle blanks. Precipitation samples with a collected volume of less than 20 ml are excluded from this data set ($n = 21$). The Hg wet deposition data discussed in this study include 162 wet-only samples collected from 21 July 2006 to 30 August 2009.

RGM has been measured at TF since November 2006 using a KCl-coated denuder module attached to a cold vapor atomic fluorescence spectrometer (Tekran model 2537A; for details see Sigler et al., 2009b). The RGM sampling interval was 2 h followed by a 30 min flush with zero air and heating cycle to desorb the RGM and allow for quantification as Hg^0 by the Tekran 2537A unit. Due to the addition of in-line Hg^P measurements in February 2009, the desorption interval increased to 60 min. Following this change the zero flushes showed no evidence of contamination, and there were no significant differences in Hg^0 and RGM levels. The limit of detection for RGM determined from three times the standard deviation of the average blank was approximately 0.1 ppqv.

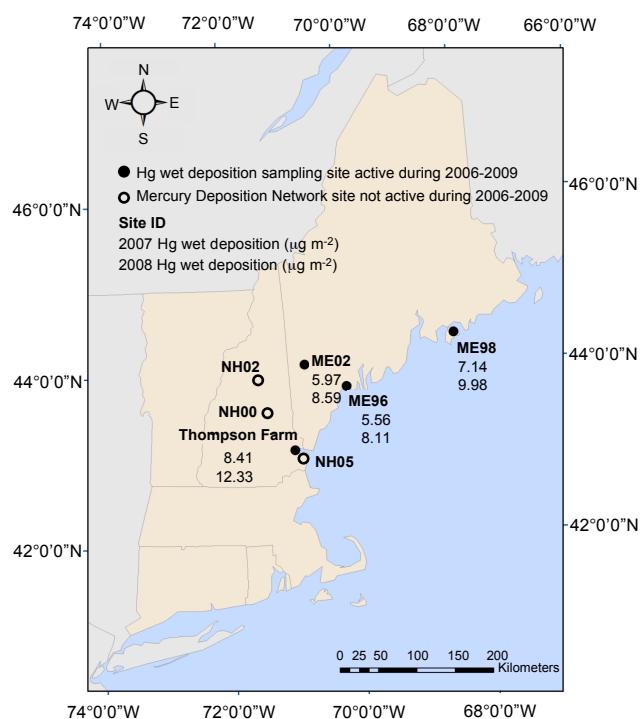


Fig. 1. Thompson Farm location and Mercury Deposition Network locations in Maine and New Hampshire.

3 Hg wet deposition seasonal patterns and inter-annual variability

Wet-only samples were collected at TF from 21 July 2006 to 30 August 2009 and represent 260 precipitation events. In this study, we define a precipitation event as a period of precipitation bordered by a twelve-hour time interval of no precipitation. An in-depth analysis of the meteorological conditions resulting in precipitation was not conducted as part of this study, therefore the potential exists that our definition of an event could include the passage of two different storm fronts within 12 h of each other. Ninety-seven samples (60 %) represent single events and 45 samples (28 %) represent two precipitation events. Figure 2a–c shows the measured concentration, calculated deposition, and total precipitation for each sample in the study period. The maximum Hg concentration was 65.09 ng l^{-1} occurring on 12 July 2007. The maximum single event deposition was $1.74 \mu\text{g m}^{-2}$ and occurred from 23 July to 24 July 2008. This single precipitation event constituted almost 6 % of the total wet deposition at TF during this three-year study and 14 % of the annual load for 2008. As shown in Fig. 2b, single precipitation events with elevated Hg deposition levels can account for a substantial portion of the total deposition. Similarly, Keeler et al. (2005) also report a single event contributing approximately 17 % to the annual Hg wet deposition load from event-based sampling in Underhill, VT.

Table 1. Seasonal and annual total precipitation, Hg wet deposition, and concentration summary statistics for Thompson Farm. Spring and summer are shaded for easier visual comparison by season.

Season	Precipitation total (cm)	Deposition ($\mu\text{g m}^{-2}$)				Concentration (ng l^{-1})			VWM concentration (ng l^{-1})
		Total	Mean	Median	Range	Mean	Median	Range	
21 Jul 2006 to 20 Sep 2006	22.46	1.16	0.116	0.089	0.027–0.263	6.16	5.05	1.39–12.51	5.23
Fall 2006	42.02	2.85	0.190	0.139	0.058–0.600	9.63	8.10	2.28–23.06	6.71
Winter 2006–2007	24.58	1.12	0.125	0.121	0.016–0.274	10.50	5.90	0.96–47.50	4.76
Spring 2007	40.13	3.39	0.339	0.379	0.030–0.561	18.14	10.57	0.99–47.89	8.69
Summer 2007	20.59	3.02	0.275	0.234	0.090–0.548	22.84	14.24	4.24–65.09	14.85
Fall 2007	30.26	0.99	0.083	0.061	0.023–0.231	3.39	2.71	0.75–8.94	3.67
Winter 2007–2008	47.53	2.17	0.135	0.117	0.055–0.399	5.79	5.36	1.41–10.88	4.33
Spring 2008	19.97	1.79	0.162	0.107	0.066–0.553	12.49	8.64	3.48–25.81	8.84
Summer 2008	52.52	6.37	0.354	0.112	0.015–1.737	15.29	14.74	4.21–37.72	12.48
Fall 2008	37.00	1.76	0.125	0.114	0.039–0.256	7.55	6.77	2.24–19.21	4.60
Winter 2008–2009	29.86	1.49	0.149	0.133	0.050–0.339	9.04	4.50	2.72–34.83	5.67
Spring 2009	27.37	2.23	0.172	0.137	0.020–0.452	9.52	7.86	3.57–17.76	8.18
21 Jun 2009 to 30 Aug 2009	37.36	2.62	0.202	0.168	0.042–0.565	9.94	9.70	3.34–20.62	7.02
Year 2007	114.1	8.41	0.205	0.155	0.016–0.561	13.68	6.88	0.75–65.09	7.97
Year 2008	160.3	12.33	0.209	0.115	0.015–1.74	10.41	8.41	1.66–37.72	8.09

During the 37-month sampling period at TF, the cumulative Hg wet deposition was $30.78 \mu\text{g m}^{-2}$ and the total precipitation depth was 4.28 m. The seasonal and annual variations in Hg concentration and wet deposition are summarized in Table 1. In this study, seasons are delineated according to the calendar definition. In general, the summer and spring exhibited elevated Hg concentrations and wet deposition with an unusually large wet deposition value in summer 2008.

The seasonal volume weighted mean (VWM) concentrations of Hg in precipitation at TF are shown in Fig. 3a and listed in Table 1. The VWM concentrations are elevated during the spring and summer seasons in comparison to the fall and winter seasons. These seasonal variations in VWM Hg concentrations are annually repeatable. The greatest seasonal VWM Hg concentrations at TF occurred in both summer seasons (summer 2007 = 14.85 ng l^{-1} ; summer 2008 = 12.48 ng l^{-1}), with the second highest seasonal concentrations occurring in the spring seasons of each year. The summer VWM Hg concentrations are 2.2–3.4 times greater than the fall and winter values. There is little variability in the VWM concentrations at TF for the same season from year-to-year. These seasonal variations are similar to previously reported patterns at MDN sites within northeastern North America (Keeler et al., 2005; VanArsdale et al., 2005; Prestbo and Gay, 2009).

Total seasonal Hg wet deposition at TF is shown in Fig. 3b and listed in Table 1. The Hg wet deposition is calculated as the product of the event concentration and amount of precipitation (Fig. 3c). Patterns in seasonal Hg wet deposition are less consistent than the VWM concentrations and are linked more closely to precipitation totals. In 2007 the highest seasonal deposition, $3.39 \mu\text{g m}^{-2}$, occurred in the spring,

while in 2008 it was observed in the summer with a value of $6.39 \mu\text{g m}^{-2}$. The large deposition in summer 2008 reflects the combination of typically greater summer Hg concentrations and the above normal precipitation for that season (Fig. 3c). The total amount of precipitation received in summer 2008 was 180 % above the 30-year summer average in New Hampshire (<http://www.nrcc.cornell.edu>). Similarly, the elevated deposition at TF during the 2007–2008 winter, compared to other winters, is most likely due to the elevated amount of precipitation, which was 154 % above the 30 year winter average (<http://www.nrcc.cornell.edu>).

Annual Hg wet deposition varied over the duration of this study and was strongly linked to annual precipitation totals. During the calendar years 2007 and 2008 the Hg wet deposition at TF was $8.41 \mu\text{g m}^{-2} \text{ yr}^{-1}$ and $12.33 \mu\text{g m}^{-2} \text{ yr}^{-1}$, respectively with corresponding precipitation totals of 114.1 cm and 160.3 cm. During this two year interval the amount of precipitation increased by 40 % and the annual Hg wet deposition increased by 47 %. These increases are similar in magnitude, indicating that the large annual Hg wet deposition for 2008 is primarily a consequence of enhanced precipitation. The amount of precipitation in New Hampshire during 2008 was 43 % above the 30 year normal and the highest annual amount of precipitation based on a 114-year record (<http://www.nrcc.cornell.edu>). In contrast, the amount of precipitation at TF during 2007 was only 11 % above the normal. To put the annual Hg wet deposition in context, the typical annual fluxes reported for MDN sites in the northeastern United States (NY, NJ, and New England) and eastern Canada from 1996–2005 were $4\text{--}8 \mu\text{g m}^{-2} \text{ yr}^{-1}$ (Prestbo and Gay, 2009). The Hg annual wet deposition at TF for 2007 is slightly above this range, whereas the annual deposition for 2008 is >50 % higher. This comparison in

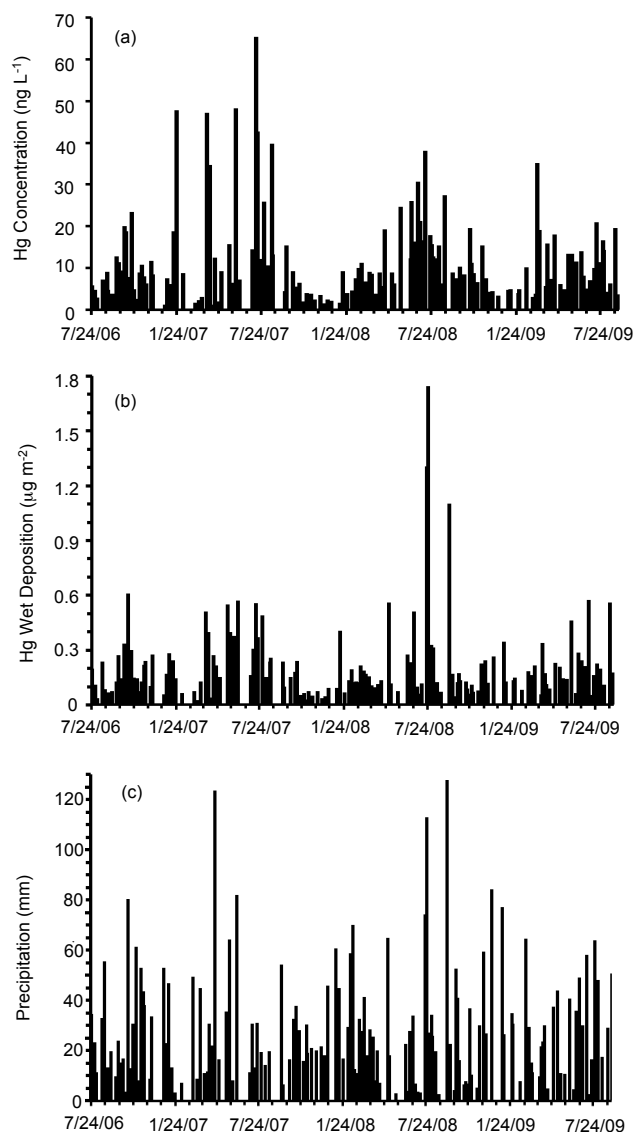


Fig. 2. Time series of wet deposition samples from Thompson Farm; (a) Hg concentration, (b) Hg wet deposition, (c) precipitation amount.

annual Hg wet deposition is made to emphasize the elevated deposition measured at TF during 2008. Comparisons between different time periods and locations should be made with caution due to the varying conditions such as the proximity and output of emission sources that may affect deposition and change with time and location.

4 Comparison with MDN sites

The wet only results from TF are compared to samples collected during the same time period at MDN sites located in Maine (Fig. 3a–c) (NADP/MDN, 2001–2009). These MDN sites were chosen for comparative purposes due to their prox-

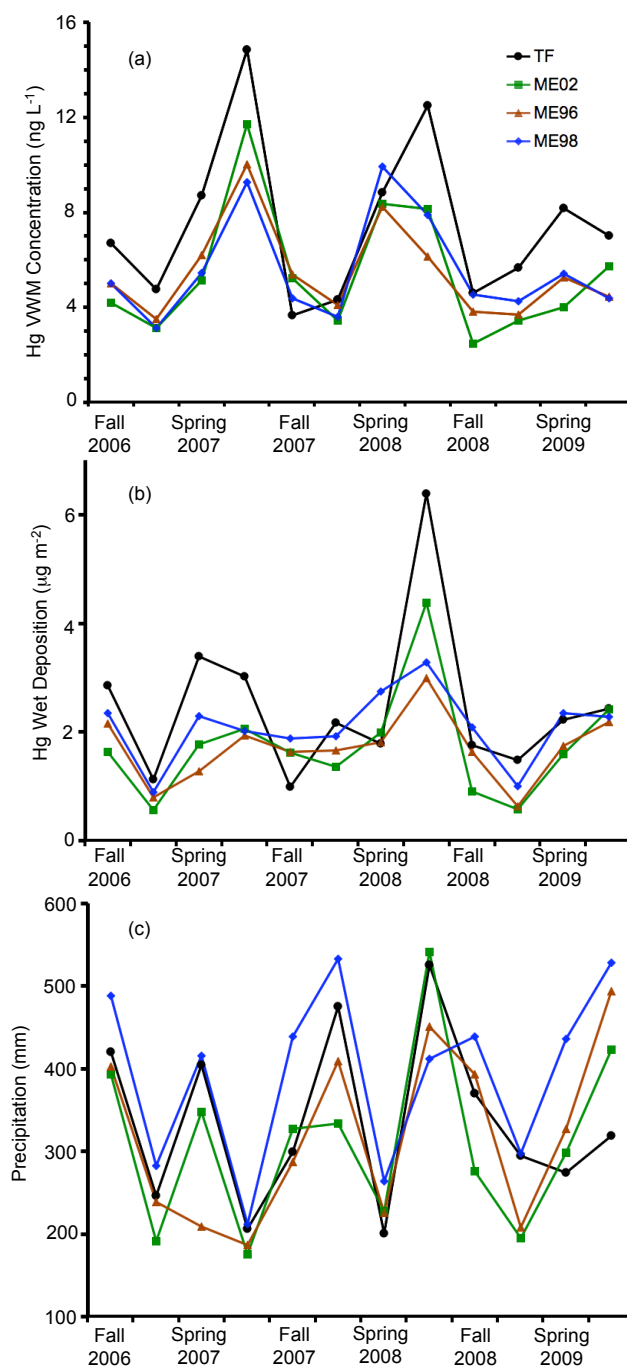


Fig. 3. Seasonal Hg volume weighted mean concentrations (a), Hg wet deposition (b), and precipitation amount (c), at Thompson Farm and Mercury Deposition Network sites located in Maine.

imity to TF, the coastal locations of ME96 and ME98 and locations downwind of the city of Boston. Patterns in seasonal VWM concentrations and Hg wet deposition are generally consistent between TF and the Maine MDN sites (Fig. 3a and b) with elevated levels during spring and summer seasons. The greatest seasonal VWM concentration during this

Table 2. Kendall's τ correlation co-efficients for Hg wet deposition and Hg concentration with meteorological conditions and gas phase measurements at Thompson Farm. Asterisks indicate $p < 0.05$.

	Hg wet deposition τ	Hg concentration τ
Daily average temperature	0.07	0.23*
Daily total solar radiation	−0.02	0.29*
Daily average CO	−0.01	0.00
Daily average NO _y	−0.09	−0.11
Daily maximum RGM	0.07	0.09
RGM depletion during pre- cipitation event	0.10	−0.02

sampling period occurred at all locations for summer 2007. The 2006–2007 winter had the lowest seasonal VWM Hg concentration at TF and all Maine MDN sites with the exception of ME02. Similarly, all sites had the highest total seasonal Hg wet deposition in summer 2008 and low wet deposition totals during the winter seasons.

The seasonal VWM Hg concentrations and seasonal wet deposition at TF are typically greater than the Maine MDN sites (Fig. 3a and b), possibly due to a combination of elevated Hg concentrations and precipitation. TF is the most southerly of the sites resulting in slightly warmer temperatures compared to the MDN sites and is also located nearer large urban pollution sources such as Boston and New York City. Mao and Talbot (2004) indicate TF can be influenced by transport of polluted air masses from the Boston and Mid-Atlantic States region. Thus it is reasonable to hypothesize that TF receives more Hg due to the proximity of anthropogenic emissions. Also the amount of precipitation recorded at TF is consistently second highest amongst these sites with MDN site ME98 regularly receiving the most precipitation. In-depth studies are warranted to understand the causes for such geographic differences in Hg wet deposition.

For an historical perspective, results from this study are briefly compared to the three MDN sites previously located in New Hampshire (Fig. 1) (NADP/MDN, 2001–2009). The only historical site with results for four complete seasons is NH05 with data available from March 2001 to June 2002. At NH05, summer 2001 had the highest VWM concentration (11.51 ng l^{-1}) and spring 2002 had the greatest seasonal Hg deposition and precipitation totaling $2.59 \mu\text{g m}^{-2}$ and 37.3 cm, respectively. Hg wet deposition data is available for NH02 from February 2004 to February 2005. For the seasons with complete data available, spring 2004 had the highest VWM Hg concentration (9.02 ng l^{-1}). Summer 2004 had the greatest wet deposition and precipitation totaling $3.47 \mu\text{g m}^{-2}$ and 46.3 cm, respectively. At NH00 data are only available for seven months from May 2001 through De-

cember 2001. The summer had greater Hg wet deposition and VWM concentration than the fall. The seasonal variations in the data collected from the MDN sites previously located in NH are consistent with our findings at TF. The spring and summer have elevated VWM concentrations and Hg wet deposition in comparison to the fall and winter.

5 Influence of meteorological conditions and other trace gases on Hg wet deposition

Relationships were examined between Hg wet deposition, Hg concentration, and meteorological parameters including temperature and solar radiation at the TF site. Non-parametric Kendall's τ was calculated to determine correlations between these parameters. Only precipitation samples representative of single events are included in this analysis. Table 2 summarizes these statistical results.

Previous studies attribute regional and seasonal differences in Hg wet deposition to temperature differences (Keeler et al., 2005). On an event basis there is weak correlation between the average daily temperature and Hg concentration ($\tau = 0.23$, $p < 0.05$). The correlation between average daily temperature and Hg wet deposition is very minor and not statistically significant. Additionally, studies suggest photochemistry is important in the production of RGM (Lin and Pehkonen, 1999; Sigler et al., 2009b) implying a relationship with Hg wet deposition (Selin and Jacob, 2008). In this study we looked into relationships between solar radiation and Hg wet deposition. At TF, Hg concentration is correlated with total daily solar radiation ($\tau = 0.29$, $p < 0.05$). The lack of strong correlations on an event basis between temperature, solar radiation and Hg wet deposition and concentrations indicates that effects from these parameters are not directly related to Hg wet deposition.

To investigate anthropogenic contributions to Hg wet deposition, we examined links with Hg wet deposition and gas phase concentrations of carbon monoxide (CO), and total reactive nitrogen (NO_y), commonly used indicators for anthropogenic influence (Mao et al., 2008). CO is emitted mainly from mobile combustion sources while NO_y includes compounds emitted directly from fossil-fuel combustion and oxidation products of such compounds. This initial investigation of relationships between CO, NO_y, and Hg concentration in precipitation and wet deposition does not suggest strong or statistically significant correlations ($p < 0.05$). An in-depth analysis of individual events with identified air mass source regions may provide more information on the lack of influence of these trace gases on Hg wet deposition.

6 Linkage between RGM and Hg wet deposition

RGM is more soluble than Hg⁰ and therefore important in contributing to both the wet and dry deposition of Hg (Schroeder and Munthe, 1998; Selin, 2009). However, few

studies report long-term concurrent measurements of RGM and Hg wet deposition (Engle et al., 2010). RGM has been measured at TF (Sigler et al., 2009b; Mao et al., 2011) and we compare these measurements with Hg wet deposition measurements during the nearly three-year period from November 2006 through August 2009.

Elevated RGM mixing ratios typically occur in winter and spring at TF (Fig. 4), and the typical diurnal cycle for RGM is a minimum at night with a rapid increase during the morning to peak levels at midday (Sigler et al., 2009b; Mao et al., 2011). Based on relationships of RGM with trace gases such as CO, CO₂, and SO₂, and meteorological conditions at TF, Sigler et al. (2009a, b) suggest the elevated RGM mixing ratios during winter months may be due to local emissions from heating sources and slower RGM removal processes. The elevated spring RGM mixing ratios at TF are attributed to photochemical production and high biogenic emissions of Hg⁰.

Scavenging of RGM during precipitation events

RGM mixing ratios typically decline during precipitation events at TF. Sigler et al. (2009a, b) and Mao and Talbot (2011) observed RGM depletion during precipitation events at this site and others have made similar observations at diverse locations (Lindberg and Stratton, 1998; Yatavelli et al., 2006; Laurier and Mason, 2007). Despite this indication of RGM scavenging during precipitation events at TF, correlations between Hg wet deposition and Hg concentration in precipitation versus daily maximum RGM and RGM depletion during precipitation events were not statistically significant (Table 2). Our results demonstrate an indirect relationship between seasonal surface level RGM mixing ratios and total aqueous Hg in wet deposition. RGM mixing ratios are greatest during the winter, yet Hg concentrations in precipitation and wet deposition are lowest during the winter. Possible explanations for the low Hg wet deposition in winter are the underestimation of wet deposition due to inefficient snow collection and/or less effective scavenging of RGM by snow.

A comparison between the collected sample volume and precipitation amount reveals that lower sampling efficiencies occur most frequently during winter precipitation events. Based on the surface area of the funnel used in our sampling train, 1 mm of precipitation should result in 12 ml of collected sample. A linear regression between the actual amount of sample collected and amount of precipitation during the non-winter seasons at TF reveals the same result (i.e. 1 ml of precipitation $\sim$ 12 ml of sample, $r^2 = 0.99$). Not all winter precipitation events are under sampled, however 13 of a total 16 precipitation events with a sampling efficiency of less than 80 % occur during the winter. It is not known how the inefficient collection of snow affects the measured Hg concentration at TF, however based on a limited study at a nearby MDN site, we infer the lower sampling efficiency may result in low Hg concentrations. Nelson et al. (2008) compare

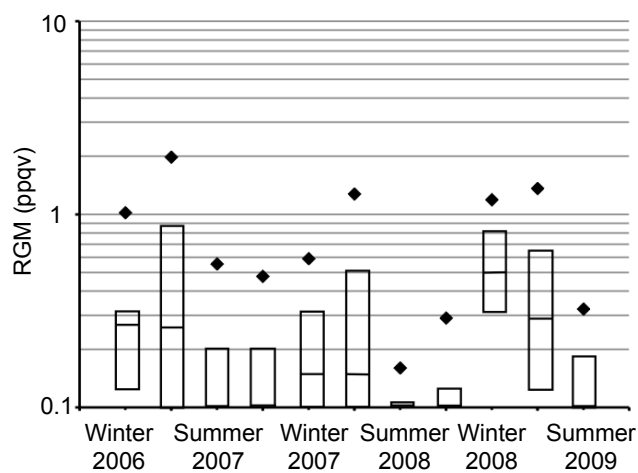


Fig. 4. Seasonal variations in RGM at TF. Each box encompasses the 25th to 75th percentiles and the solid horizontal line within each box represents the median value. The black diamonds indicate the 90th percentile.

event-based snow sampling techniques at MDN site ME98. Their results show greater snow water equivalents (i.e. collection efficiency) and Hg snowfall concentrations in samples collected using a collection method different than the MDN.

In this study, ineffective scavenging of RGM by snow is evidenced by the less frequent depletion of RGM below the limit of detection (LOD, 0.1 ppqv) during winter precipitation events at TF. Seven of 19 winter precipitation events (37 %) result in RGM mixing ratios below the LOD. RGM mixing ratios during summer precipitation events dip below the LOD at a much higher frequency; 17 of 20 events (85 %). These seasonal variations in RGM removal efficiencies substantiate the hypothesis that seasonal variations in Hg wet deposition are due in part to less effective scavenging of gas phase Hg by snow (Keeler et al., 2005; Selin and Jacob, 2008).

7 Estimation of RGM dry deposition

It is important to gauge the relative contribution of Hg wet deposition in comparison with other atmospheric Hg sinks such as RGM dry deposition. To accomplish this we performed an order-of-magnitude estimate for RGM dry deposition using long-term continuous measurements of RGM mixing ratios. Estimates of RGM dry deposition velocity and deposition at TF were calculated based on nighttime depletion events, which are most common during warm season (May to September) nocturnal inversions in the planetary boundary layer. The method has been employed in Talbot et al. (2005), Mao et al. (2008), and Sigler et al. (2009b), and the step-by-step estimate is elucidated in Russo et al. (2010). A brief explanation of this method is given here. Nocturnal

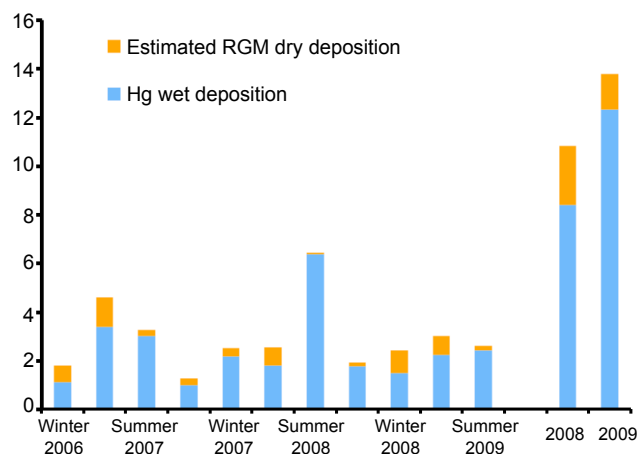


Fig. 5. Seasonal and annual Hg wet deposition and estimated RGM dry deposition at TF.

inversions at TF are evidenced by the depletion (<5 ppbv) of atmospheric ozone and Hg^0 (Mao et al., 2008). Concurrent depletions were also observed in RGM. To obtain a robust estimate we used the diurnal cycle average over all days from the warm season with the occurrence of nocturnal inversions. The average rates of RGM depletion and RGM concentration during these inversions were calculated to solve for the deposition velocity in the following equation:

$$V_d = \frac{dC}{dt} \cdot \frac{H}{\bar{C}} \quad (1)$$

where V_d is the deposition velocity, dC/dt is the rate of change in RGM concentration from the average diurnal cycle in RGM over all inversion events, $\bar{C}$ is the average RGM concentration over the depletion period, and H is the boundary layer height. In these calculations a constant boundary layer height of 125 m is applied (Talbot et al., 2005; Mao et al., 2008; Russo et al., 2010). This calculation also assumes that during the nocturnal inversions dry deposition is the only loss mechanism of RGM and there is no RGM production, therefore the calculated V_d should be considered a maximum due to the potential for RGM loss due to aerosol uptake.

Nocturnal inversion events were identified by the nighttime depletion of ozone to less than 5 ppbv with a corresponding decrease in RGM to less than 0.1 ppqv. The number of inversion events per warm season varied from 17 to 21 during 2007 to 2009. The average RGM concentration over the depletion period varied annually from 0.13 to 0.20 ppqv however, the RGM depletion based on the average diurnal cycle was always complete in the time window of 00:00 to 03:00 UTC. Using Eq. (1) the average RGM dry deposition velocity at TF is estimated to be 2.31 cm s^{-1} . This estimate is within the range of RGM dry deposition velocities reported in the literature (0.5 to 7.6 cm s^{-1}) from a variety of measurement methods, surface compositions, locations, and seasons (Zhang et al., 2009, and references therein).

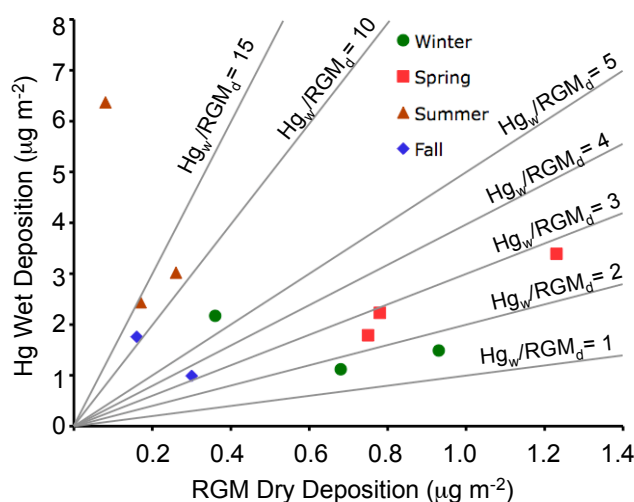


Fig. 6. Seasonal Hg wet deposition fluxes and estimated RGM dry deposition fluxes at TF. Contour lines represent wet to dry deposition (Hg_w/RGM_d) ratios.

RGM dry deposition at TF was estimated using measured RGM mixing ratios and a dry deposition velocity of 2.31 cm s^{-1} . The seasonal and annual estimated RGM dry deposition and comparison to Hg wet deposition is shown in Fig. 5. There is distinct variation in seasonal dry deposition of RGM. The greatest seasonal RGM dry deposition ($>0.6 \mu\text{g m}^{-2}$) occurs in the winter and spring (excluding winter 2007), following the seasonal pattern in RGM mixing ratios. Summer and fall exhibit low RGM dry deposition values, all below $0.4 \mu\text{g m}^{-2}$ (Fig. 6).

8 Comparison between RGM dry deposition and Hg wet deposition

Estimated RGM dry deposition is less than the measured Hg wet deposition for all seasons and on an annual basis (Table 3 and Fig. 5). Our results suggest that the relative contribution of Hg wet deposition and RGM dry deposition to the total Hg deposition flux at TF varies greatly by season and is opposite in phase with ratios of Hg wet deposition to RGM dry deposition ranging from 1.6 in the winter to 80 during summer 2008 (Fig. 6). Large Hg wet deposition and low RGM dry deposition typically occurs in summer. The greatest ratio occurred in summer 2008 reflecting the exceptionally large amount of precipitation and Hg wet deposition and the lowest RGM dry deposition estimate of all summers. On an annual basis the ratios of Hg wet deposition to RGM dry deposition are moderate in comparison to the large seasonal variations at TF. The ratio for annual year 2008 is more than double the ratio for 2007 (8.5 and 3.5, respectively) and the large ratio likely reflects the record amount of precipitation in 2008.

We can compare our calculations to only a few studies from the literature reporting both Hg wet deposition

Table 3. Seasonal and annual Hg wet deposition and estimated RGM dry deposition fluxes, and the sum of Hg wet deposition and estimated RGM dry deposition at TF. The asterisks indicate seasons missing more than 3 days of RGM measurements. The seasonal daily average RGM was used to fill gaps in the data and calculate a total RGM flux. Spring and summer are shaded for easier visual comparison by season.

Season	Hg wet deposition ($\mu\text{g m}^{-2}$)	RGM dry deposition ($\mu\text{g m}^{-2}$)	Wet plus RGM dry deposition ($\mu\text{g m}^{-2}$)
Winter 2006–2007	1.12	0.68	1.80
Spring 2007	3.39	1.23	4.62
Summer 2007	3.02	0.26	3.28
Fall 2007	0.99	0.30	1.29
Winter 2007–2008	2.17	0.36	2.53
Spring 2008	1.79	0.75	2.54
Summer 2008	6.37	0.08*	6.45
Fall 2008	1.76	0.16*	1.92
Winter 2008–2009	1.49	0.93*	2.42
Spring 2009	2.23	0.78	3.01
21 Jun 2009 to 30 Aug 2009	2.44	0.17	2.61
Year 2007	8.41	2.43	10.84
Year 2008	12.33	1.45	13.78

Table 4. A comparison between annual Hg wet deposition and RGM dry deposition values reported in the literature and calculated in this study. Hg wet deposition to RGM dry deposition ratios are calculated from data provided in Engle et al. (2010) and Miller et al. (2005).

Location	Dates	Hg wet deposition ($\mu\text{g m}^{-2} \text{ yr}^{-1}$)	RGM dry deposition ($\mu\text{g m}^{-2} \text{ yr}^{-1}$)	Hg wet dep./ RGM dry dep.	Reference
Alabama	12 Apr 2005–11 Apr 2006	10.9	2.2	4.95	Engle et al. (2010)
Illinois	1 Jan 2004–31 Dec 2004	11.0	51.8	0.21	Engle et al. (2010)
Massachusetts	5 Feb 2008–3 Feb 2009	2.9	1.0	2.9	Engle et al. (2010)
New Hampshire	None given	5.8	7.5	0.77	Miller et al. (2005)
New Hampshire	1 Jan 2007–31 Dec 2007	8.41	2.43	3.46	This study
New Hampshire	1 Jan 2008–31 Dec 2008	12.33	1.45	8.50	This study
North Dakota	1 Jan 2004–12 Dec 2004	3.3	1.7	1.94	Engle et al. (2010)
Puerto Rico	1 Jan 2006–31 Dec 2006	29.5	0.5	59	Engle et al. (2010)
South Carolina	23 May 2006–22 May 2007	6.5	1.8	3.61	Engle et al. (2010)
Virginia	1 Jan 2006–12 Dec 2006	9.0	1.4	6.43	Engle et al. (2010)
Wisconsin	28 Jun 2004–6 Jun 2005	6.7	5.3	1.26	Engle et al. (2010)

and RGM dry deposition (Table 4). Published comparisons of Hg wet deposition to total Hg dry deposition ($\text{Hg}^0 + \text{RGM} + \text{Hg}^P$) in New Hampshire are based on modeled results (Miller et al., 2005; Han et al., 2008). Miller et al. (2005) estimate a total Hg flux of $21.1 \mu\text{g m}^{-2} \text{ yr}^{-1}$ in New Hampshire with approximately equal contributions of 7.4 and $7.5 \mu\text{g m}^{-2} \text{ yr}^{-1}$, respectively, from Hg^0 and RGM dry deposition followed by a wet deposition contribution of $5.8 \mu\text{g m}^{-2} \text{ yr}^{-1}$. Minor contributions are attributed to Hg^P and cloud water at 0.38 and $0.058 \mu\text{g m}^{-2} \text{ yr}^{-1}$, respectively. Miller et al. (2005) state that their RGM estimates should be considered within the correct order magnitude but they have low confidence in the exact value due to the lack of measurement data for comparison. Han et al. (2008) simulated

the total atmospheric deposition of RGM plus Hg^P in New Hampshire for the years 1996, 1999, and 2002 based on Hg emission inventories for the state and adjacent areas. Their ratios of annual wet to dry RGM plus Hg^P deposition range from 1.01 to 0.57. RGM deposition ranges from a factor of 6 to 21 times greater than Hg^P deposition. The model used by Han et al. (2008) only considers Hg from local emission sources and does not account for regional and global sources of Hg or atmospheric reactions.

Annual ratios of Hg wet deposition to RGM dry deposition for eight sites located in the eastern United States and Puerto Rico were calculated from data in Engle et al. (2010). Engle et al. (2010) determine RGM dry deposition using continuous RGM concentration measurements and a numerical

resistance-based deposition model. Miller et al. (2005) estimate higher annual fluxes of RGM dry deposition than Hg wet deposition for New Hampshire. In comparison the TF ratio for 2007 is within the range of values from Engle et al. (2010) for rural and coastal sites and the TF ratio for 2008 is slightly greater (excluding Puerto Rico). In contrast to the findings of Miller et al. (2005), results from our study, as well as those of Engle et al. (2010), demonstrate that annual Hg wet deposition fluxes are typically greater than RGM dry deposition fluxes. The observations hold across many different sites despite differences in geographic location and sampling years. The exception to this generalization is one urban site studied in Engle et al. (2010) in East St. Louis, IL, which likely stands out because of its large population (2.8 million) as well as the impact of several local anthropogenic sources.

9 Summary and conclusions

Total aqueous Hg in precipitation samples collected at TF in Durham, NH from July 2006 to September 2009 demonstrate seasonal Hg wet deposition and VWM concentration patterns consistent with previous observations for the north-eastern United States with elevated values during the summer and spring seasons. Wet deposition samples from regional MDN sites collected during the same sampling interval exhibit similar seasonal patterns. Comparisons of the relative Hg precipitation concentrations and wet deposition fluxes between the TF and MDN sites suggest that the proximity to anthropogenic Hg sources may partially explain observed differences.

The quantity of precipitation also contributes to the seasonal and annual variations in Hg wet deposition. As observed at TF, the winter 2007–2008 and summer 2008 had above normal precipitation amounts and high Hg wet deposition fluxes. This relationship is also exhibited on an annual basis with the anomalously high amount of precipitation that fell during 2008 contributing to the very high annual Hg wet deposition flux for the year. While this observation may seem rudimentary (i.e. more precipitation equates to more wet deposition), it warrants noting as observed and predicted increases in precipitation amount and intensity in the mid-latitudes due to climate change (Easterling et al., 2000) imply Hg wet deposition fluxes will also increase.

Our multi-year dataset and event-based sampling of Hg wet deposition and RGM measurements allows for seasonal comparisons. The inefficient scavenging of RGM by snowfall is evidenced by the less frequent depletion of RGM below the LOD during winter months. Although the winter wet deposition values are low, the RGM dry deposition estimates at TF indicate enhanced dry deposition in the winter. These seasonal changes in Hg deposition pathways are reflected in the seasonal ratios of Hg wet deposition to RGM dry deposition. These ratios differ greatly by season and range from a summer value of 79.6 to a winter value of 1.60. In general,

the winter and spring ratios are lowest while the summer ratios are greatest. The elevated amounts of precipitation during 2008 likely influence our ratios of Hg wet to RGM dry deposition. The seasonality in the atmospheric Hg depositional mechanisms (wet vs. dry) may subsequently affect the fate and transport of Hg in aquatic and terrestrial ecosystems. Improved quantification of Hg wet and dry deposition, via long term simultaneous measurements and advances in measurement technology, will lead to a better understanding of the biogeochemical cycle of Hg.

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