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A comparison of speciated atmospheric mercury at an urban center and an upwind rural location†

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Gaseous elemental mercury (GEM), particulate mercury (PHg) and reactive gaseous mercury (RGM) were measured every other hour at a rural location in south central Wisconsin (Devil's Lake State Park, WI, USA) between April 2003 and March 2004, and at a predominantly downwind urban site in southeastern Wisconsin (Milwaukee, WI, USA) between June 2004 and May 2005. Annual averages of GEM, PHg, and RGM at the urban site were statistically higher than those measured at the rural site. Pollution roses of GEM and reactive mercury (RM; sum of PHg and RGM) at the rural and urban sites revealed the influences of point source emissions in surrounding counties that were consistent with the US EPA 1999 National Emission Inventory and the 2003–2005 US EPA Toxics Release Inventory. Source-receptor relationships at both sites were studied by quantifying the impacts of point sources on mercury concentrations. Time series of GEM, PHg, and RGM concentrations were sorted into two categories; time periods dominated by impacts from point sources, and time periods dominated by mercury from non-point sources. The analysis revealed average point source contributions to GEM, PHg, and RGM concentration measurements to be significant over the year long studies. At the rural site, contributions to annual average concentrations were: GEM (2%; 0.04 ng m^{-3}); and, RM (48%; 5.7 pg m^{-3}). At the urban site, contributions to annual average concentrations were: GEM (33%; 0.81 ng m^{-3}); and, RM (64%; 13.8 pg m^{-3}).

Introduction

Identifying the sources of atmospheric mercury to sensitive receptor sites is critical to developing effective emission control policies. Understanding a source–receptor relationship for atmospheric mercury lies in a sound knowledge of: (i) the spatial and chemical characteristics of emissions; (ii) the transport, transformation and fate of atmospheric mercury, and (iii) and the bioaccumulation of mercury in the food web once it has been deposited. Present research efforts to understand mercury source–receptor relationships are motivated by the desire to mitigate the anthropogenic mercury contamination of aquatic food webs with methyl mercury. Successful mitigation would help protect the health of prenatal children of mothers who consume fish during pregnancy,[1–3] and avert sub-clinical health effects in individuals with high sport and commercial fish consumptions.[4–6]

Mercury exists as three different species in the atmosphere, each with very different chemical and physical properties, and therefore different source–receptor relationships. Gaseous elemental mercury (GEM) is resistant to chemical oxidation, which means that it has an atmospheric residence time between 0.5–2 years,[7–9] allowing it to become intrahemispherically well mixed. Oxidized mercury (II) compounds are operationally defined as particulate mercury (PHg) and reactive gaseous mercury (RGM), which can be considered subclasses of reactive mercury (RM; sum of RGM and PHg).[10,11] RM compounds are more water soluble and less volatile than GEM, resulting in more rapid removal from the atmosphere.[7,8] The difference in time scales of removal between GEM and RM means that RM will be deposited within a few tens to hundreds of kilometres from the source, whereas GEM will be mixed into the global pool before it is oxidized to RM and subsequently deposited,[7,8,12] a process which is very slow and spatially diffuse. The contrast between the GEM and RM source–receptor relationships means that reductions in RM emissions have the potential to yield large reductions in regions of mercury deposition at short and medium distances from point source emissions, whereas reductions in GEM emissions from point sources would yield much smaller decreases at short and medium distances from the emission sources.

The ability to quantify the impact of point sources on local mercury deposition is essential in formulating mercury emission control policies. Several studies have been conducted to assess the origins and magnitudes of mercury point source impacts at US receptors. The studies fall into two categories:

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(i) modeling studies;[9,12–14] and (ii) field measurements,[15–23] often combined with source apportionment and back trajectory models.[24–35] Modeling studies typically provide estimates of average source impacts over large areas, but impacts of point sources on individual receptors short distances downwind are not as well represented, due to the size of the model grid scales and uncertainties in the mercury emission inventory.[9,12,36] These limitations reduce the usefulness of modeling studies to air quality managers and regulators interested in protecting specific water bodies. Field studies can provide a higher spatial and temporal resolution that is very specific to a receptor site, and therefore are potentially powerful tools for defining source–receptor relationships.

In this study, we present a methodology for identifying source regions and calculating an estimate of source impacts on atmospheric concentrations of GEM and RM at a receptor site. The methodology used semi-continuous real time GEM, PHg and RGM measurements made at a rural site for one year and at a predominantly downwind urban site during the following year. The objectives of the study were achieved by: (i) understanding the differences between rural and urban GEM, PHg and RGM concentration measurements; (ii) quantifying the local and regional impact of GEM, PHg and RGM point sources; and (iii) understanding the seasonality of GEM, PHg and RGM concentrations at both sites.

Experimental

Measurement locations

Speciated ambient mercury measurements were made at a rural location (Devil's Lake State Park (DLSP), WI, USA) and at an urban location (Milwaukee, WI, USA) 160 kilometres east of the rural location. During the summer months, prevailing winds in Wisconsin were from the west, meaning that the rural measurement site was located upwind of the urban measurement site (Fig. S1† in the ESI). During the winter and spring months, easterly wind patterns were often observed and were predominant throughout April and May 2003, reversing the upwind–downwind relationship of the sites. The rural site was located at DLSP (43° 26′ 05″N, 89° 40′ 48″W) between April 2003 and March 2004. The urban site was located north of downtown, 0.5 to 1 mile from Lake Michigan (2114 E. Kenwood Blvd, Milwaukee, WI, USA; 43° 06′ 29″N, 87° 53′ 02″W) between June 2004 and May 2005. The ambient mercury analyzer used to make the measurements (see next section) was mounted on top of a portable cabin at DLSP, and on the roof of a two-storey building in Milwaukee. At both sites, the sample inlet was 1.5 m above the nearest surface. Although the data sets were collected during consecutive years, the comparison of the measurements provided powerful insight into the differences in RM concentrations at the rural and urban measurement locations.

Real time ambient mercury analyzer

A real time *in situ* ambient mercury analyzer (Tekran, Inc., Toronto, Ontario, Canada) was used to measure RGM, PHg, and GEM at both measurement locations. Ambient air was introduced into the instrument at 10 L min^{-1} for one hour through a heated elutriator that removed particles larger than 2.5 μm in diameter using an impactor. RGM was collected onto a KCl coated annular denuder, and PHg was collected on a quartz fiber filter located after the RGM denuder in the sampling train. The RGM denuder and PHg filter were located in protective cases that remained outdoors. The cases were heated internally to 50 °C to prevent condensation from damaging the KCl coating on the denuder, or collecting in the sample lines during high relative humidity events. Ambient GEM was collected over 5 min periods during the hour when RGM and PHg were also being collected. Ambient GEM was passed through the RGM and PHg compartments and collected onto gold granules in the mercury analyzer. During analysis, the GEM was thermally extracted from the gold granules and measured using cold vapor atomic fluorescence spectroscopy (CVAFS). During the hour following sample collection, the instrument went off-line and the PHg and RGM samples were analyzed. The PHg collected on the filter was thermally reduced to GEM and desorbed from the filter at 800 °C. The GEM and any unconverted RM compounds were evaporated and heated further using quartz chips at a temperature of 800 °C to ensure that any remaining RM compounds were converted to GEM. The evolved GEM was preconcentrated on the gold granules and then measured with CVAFS.[37] The RGM collected by the denuder was reduced to GEM by heating to 500 °C. The gases evolved during the heating of the RGM denuder were treated and analyzed in the same way as the gases evolved from the PHg filter. The Tekran ambient mercury analyzer has been described in more detail in previous publications.[10,11,38–40]

Meteorological data

Wind direction and atmospheric temperature data at DLSP were obtained from the US EPA Air Quality System website.[41] Wind direction and temperature data at Milwaukee were purchased from the National Environmental Satellite, Data and Information Service (NESDIS), operated by the National Oceanic and Atmospheric Association (NOAA). Measurements at DLSP were made with a Model 2020 Wind Vane and a Model 4480 Thermistor Temperature Probe (All Weather Inc., Sacramento, CA, USA). At Milwaukee, data were collected using a Model 020C Wind Direction Sensor and a Model 060A Air Temperature Sensor (Met One, Grants Pass, OR, USA). Hourly data for both parameters were available at both sites.

Results and discussion

Fig. 1a and b and Fig. 2a and b show the annual and monthly averages of GEM, PHg and RGM that were measured at DLSP and Milwaukee. The annual average concentrations of all species measured at DLSP (1.61 ± 0.01 ng m^{-3}; 9.1 ± 0.1 pg m^{-3}; 5.3 ± 0.2 pg m^{-3}) were considerably lower than the annual average concentrations measured at Milwaukee (2.48 ± 0.02 ng m^{-3}; 11.8 ± 0.3 pg m^{-3};10.3 ± 0.2 pg m^{-3}). These results agreed well with published studies conducted in rural and urban locations.[16,42] The higher monthly averages at Milwaukee indicated that the source–receptor relationships at the rural and urban sites were different. The National

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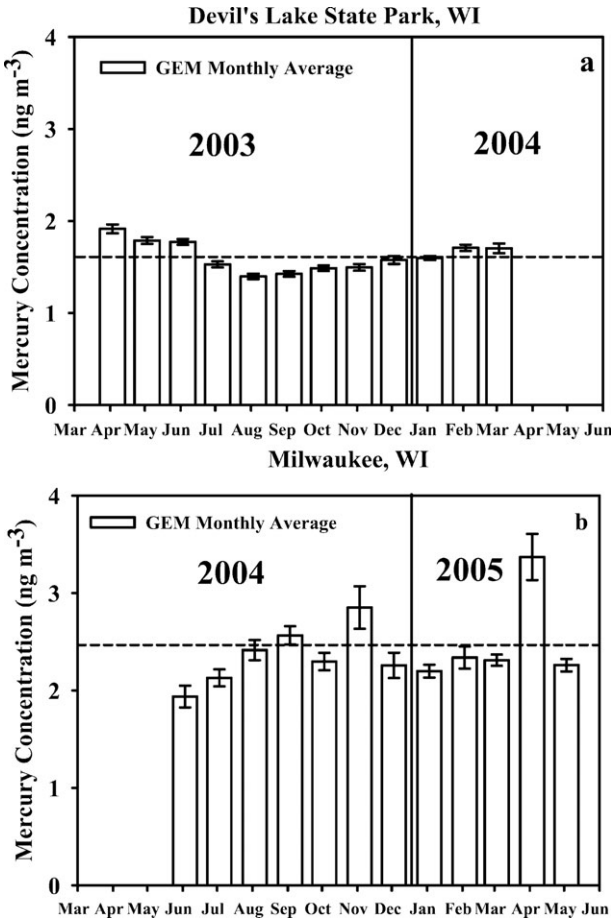

Fig. 1 Monthly averages of gaseous elemental mercury (GEM) concentrations at Devil's Lake State Park, WI, between April 2003 and March 2004 (a) and Milwaukee, WI, between July 2004 and May 2005 (b). The dashed lines represent the annual GEM averages of 1.61 ± 0.01 ng m^{-3} (a) and 2.48 ± 0.02 ng m^{-3} (b). All uncertainties are represented with standard errors of the averages.

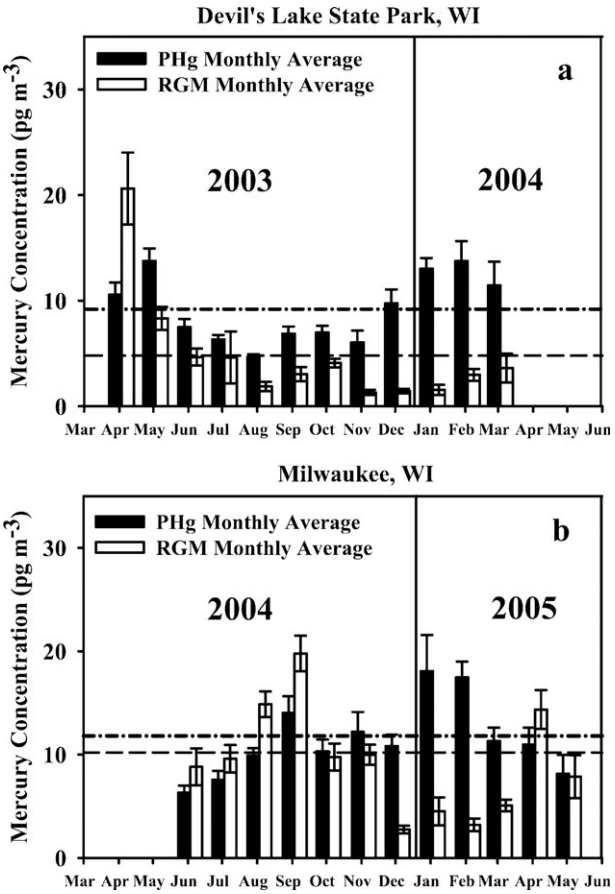

Fig. 2 Monthly averages of particulate mercury (PHg) and reactive gaseous mercury (RGM) concentrations at Devil's Lake State Park, WI, between April 2003 and March 2004 (a), and Milwaukee, WI, between July 2004 and May 2005 (b). The dashed line represents the PHg and RGM annual averages of: (i) 9.1 ± 0.1 pg m^{-3}, and 5.3 ± 0.2 pg m^{-3} (a); and (ii) 11.8 ± 0.3 pg m^{-3}, and 10.3 ± 0.2 pg m^{-3} (b). All uncertainties are represented with standard errors of the averages.

Emissions Inventory (NEI)[43] and Toxics Release Inventory (TRI)[44] revealed that total mercury emissions were higher, more spatially dense, and closer to the site in Milwaukee than in DLSP: total mercury (sum of GEM, PHg and RGM) emissions were approximately 4 times higher in Milwaukee Co. and surrounding counties (Milwaukee-Chicago area; $\sim$5000 km^2), than in Sauk Co. and surrounding counties (DLSP area; $\sim$225 000 km^2).

Fig. 2a and b exhibited seasonal variations in the relative distribution of RM between RGM and PHg. This was caused by the effect of ambient temperature on the vapor pressure of RM, which is semi-volatile.[8,45,46] More reactive mercury is associated with atmospheric particles during the winter months (80–90%), when the temperature is colder and the vapor pressure of RM is lower, than in the warmer summer months (40–60%). This phenomenon was observed at both the rural and the urban sites.

Fig. 3a and b show time series excerpts from RM concentration measurements made at DLSP between April 2003 and March 2004 (Fig. 3a), and Milwaukee between June 2004 and May 2005 (Fig. 3b). Fig. S2a–h† in the ESI show complete time series of GEM, PHg, RGM, and RM measurements made at both sites. Short-lived concentration increases in all of the three mercury species were interpreted as the impacts from point sources because they could not be explained by atmospheric oxidation of GEM to RM,[7,32,33,47,48] or the re-emission of previously deposited mercury.[49] The data presented in Fig. 3a and b, and Fig. S2a–h† revealed that increases in all of the mercury species at DLSP and Milwaukee did not fit the diurnal pattern that would be expected with re-emission of previously deposited RM and GEM.[49–54] Furthermore, correlations between reactive mercury and ozone were very weak at both DLSP ($r^2 < 0.02$) and Milwaukee ($r^2 < 0.05$) before and after point source impacts were removed, indicating that at these locations, rapid GEM oxidation by ozone was not a strong contributor to short-lived increases in RM concentrations. Therefore, the assertion that short-lived increases in RM at DLSP and Milwaukee were predominantly caused by point sources was considered to be valid. Fig. S3–S5† in the ESI contain illustrative examples of individual short-lived increases linked through back trajectories to point sources listed in the EPA NEI.[43] Forty-eight hour back trajectories were calculated for these point source impacts using the NOAA web based HYSPLIT model (v.4.8)[55] and

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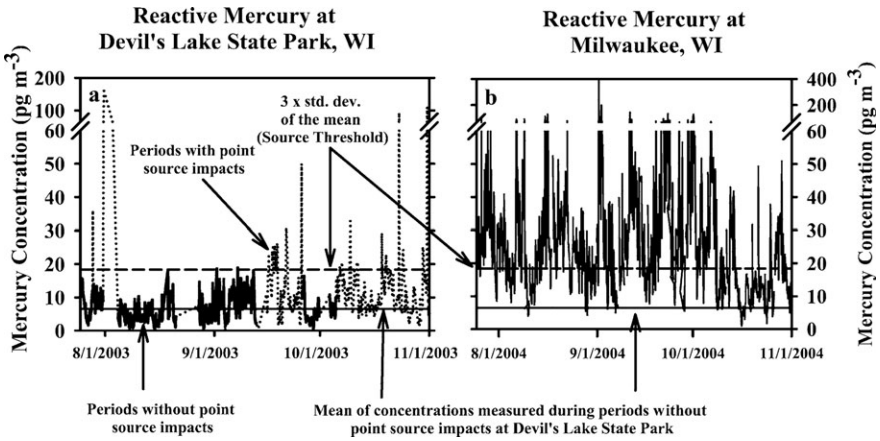

Fig. 3 Excerpts of reactive mercury concentration measurements made at (a) Devil's Lake State Park, WI, USA (DLSP) and (b) Milwaukee, WI, USA. The bold solid lines in (a) depict sections of the DSLP data sets during which no point source impacts were observed, that were used to calculate a reference mean and a point source threshold (shown in (a) and (b)). The reference mean and point source threshold values were then used to estimate point source impacts at DLSP and Milwaukee.

are also presented in Fig. S3–S5.† Absences in short-lived concentration increases at DLSP were defined as periods during which point sources did not impact the measurement site. These periods are shown in Fig. 3a as the sections of data represented with bold solid lines.

In order to estimate the contributions of point sources to atmospheric mercury concentrations measured at DLSP and Milwaukee, a simple data analysis method was developed around this approach to identify point source impacts. Mean GEM and RM concentrations during periods without point source impacts were calculated for southern Wisconsin using the bold sections of the DLSP data presented in Fig. 3a and S2a–d† of the ESI: (i) GEM = 1.5 ± 0.2 ng m^{-3} (1 std. dev.; n = 1474); and, (ii) RM = 6.5 ± 4.0 pg m^{-3} (1 std. dev.; n = 1203). These mean concentrations will be hereafter referred to as the reference means. A threshold three times the standard deviation above the reference mean was used to sort the GEM and RM concentrations from both DLSP (Fig. 3a) and Milwaukee (Fig. 3b) into two classes: (i) periods when the receptor site was impacted by emissions from a point source; and (ii) periods of time when no impacts from point sources were observed. The concentration data were sorted using the histogram analysis tool available in Microsoft Excel®. Any concentrations higher than the point source thresholds (*ST*; *i.e.* for GEM, $ST = 1.5 + (3 \times 0.2)$ ng m^{-3} = 2.1 ng m^{-3}) were considered to be impacts from point source emissions.

Once the concentrations had been sorted into the point source and non-point source classes, the following calculations were performed:

$$NI = \frac{\sum [C_{\leq ST} \times t]}{n} \quad (1)$$

$$SI = \frac{\sum [(C_{>ST} - R) \times t]}{n} \quad (2)$$

where NI is the impact of the non-point source atmospheric mercury measured at the receptor, SI is the impact of a point source on the receptor, C is the concentration of GEM or RM measured by the Tekran during a measurement cycle, t is the time over which C was measured (1 h), R is the reference mean, ST is the point source threshold, and n is the number of hours of concentration measurements made throughout the year. An important point to remember is that any concentration measured that was higher than the ST due to a point source impact was made up of the reference mean with the point source impact added to it. Therefore, all of the subtractions of R made in eqn (2) are added to the $C_{\leq ST}$ term in eqn (1). Both the rural and urban data sets were analyzed with this method.

The results of the point source impact calculations are presented in Table 1 as the contributions of point sources to annual average GEM and RM concentrations measured at DLSP and Milwaukee. Table 1 shows that point sources have a lower impact on GEM concentrations than non-point sources at both DLSP and Milwaukee. At DLSP, the impact on the annual average GEM concentrations was 2.4%, while at Milwaukee the average contribution was 33%. Non-point sources contributed the majority of GEM at both sites: 98% at

Table 1 Contributions of point sources and non-point sources to the annual averages of gaseous elemental mercury and reactive mercury at Devil's Lake State Park and Milwaukee, WI, USA

	Devil's Lake State Park, WI, USA					Milwaukee, WI, USA				
	Annual average conc.	Contribution to annual average conc.				Annual average conc.	Contribution to annual average conc.			
		Non-point source	%	Point source	%		Non-point source	%	Point source	%
GEM/ng m^{-3}	1.62	1.58	98	0.04	2	2.48	1.67	67	0.81	33
RM/pg m^{-3}	11.8	6.0	52	5.7	48	21.7	7.9	36	13.8	64

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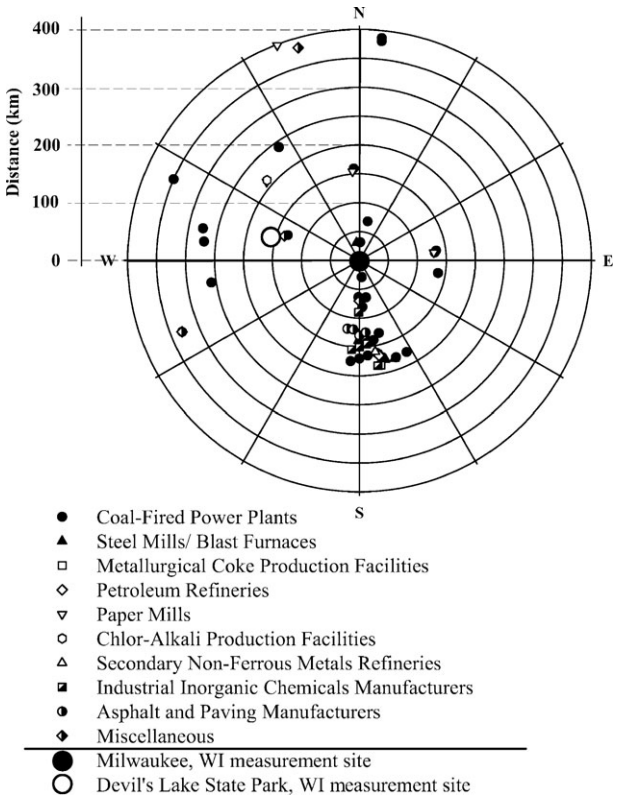

Fig. 4 A diagram that shows the locations of mercury point sources (>50 lbs yr^{-1}) listed in the US EPA National Emission Inventory and Toxics Release Inventory. Also shown are the rural and urban measurement sites at Devil's Lake State Park and Milwaukee, WI.

DLSP and 67% at Milwaukee. In contrast, the contributions of point sources to RM concentrations measured at DLSP and Milwaukee were much higher than contributions of point sources to GEM concentrations. Contributions of point sources to annual average RM concentrations were 48% at DLSP and 64% at Milwaukee, while non-point sources contributed 52% and 36%, respectively.

In order to determine the source–receptor relationships at DLSP and Milwaukee, a knowledge of the types of nearby points sources and their locations was necessary. Fig. 4 is a wind rose of all of the large mercury emitting point sources (defined in this study as a source that emits more than 50 lbs yr^{-1}) listed by either the NEI[43] or the TRI.[44] The Milwaukee and DLSP measurement sites are shown as a black circle at the origin, and a white circle to the west, respectively. The sources listed in the inventories were sorted into ten categories: coal fired power plants; industrial inorganic chemical manufacturers; chlor-alkali production facilities; metallurgical coke production facilities; steel mills/blast furnaces; asphalt and paving manufacturers; secondary non-ferrous metals refineries; petroleum refineries; paper mills; and miscellaneous, which included a crematory, a battery processing plant, a corn processing plant and a waste-to-energy facility. The mercury emissions (not speciated into GEM, PHg and RGM) from each of these categories are presented in Table 2. Coal fired power plants contributed 54% of the mercury emissions in the area, making them the highest emitting source category of mercury in the region. Industrial inorganic chemical manufacturers are the next largest emitting category (17%), followed by

Table 2 Annual emissions of mercury compounds from facilities surrounding the Devil's Lake State Park and Milwaukee, WI, USA, measurement sites in Southern Wisconsin, Eastern Iowa, Northern Illinois, and Western Michigan

Type of facility	Emissions/lbs yr^{-1}	%
Coal fired power plants	6918	54
Industrial inorganic chemical manufacturers	2122	17
Chlor-alkali plants	1082	8
Coke production facilities	650	5
Steel mills/blast furnaces	565	4
Asphalt and paving	443	3
Secondary non-ferrous metal refineries	215	2
Petroleum refineries	159	1
Paper mills	154	1
Miscellaneous	438	3
Total	12 747	

the remaining categories, each of which emit less than 8% of the total. Neither the NEI nor the TRI provided mercury speciation information for any of the facilities in the inventories. The 2002 NEI provided a table of nominal speciation information for a wide variety of process categories, but gave no assurance of the accuracy of these values for any particular facility. The speciation of mercury point source emissions controls how close to the emission source deposition occurs. For example, it is clear from Table 1 and Fig. S2a–h† that RM concentrations in both DLSP and Milwaukee were impacted by point sources emitting substantial amounts of reactive mercury. In order for environmental impact assessments to be successful, it is important that accurate speciation information be provided to air quality professionals for all sources emitting mercury.

Fig. 5a and b show pollution roses for GEM and RM at DLSP. The data presented were greater than the point source threshold (*ST*), so that only point source impacts are shown. Fig. 5a and b revealed that elevated concentrations of GEM and RM came predominantly from the east and southeast, southwest, and northwest. The data in Fig. 5a and b corresponded well with the NEI and TRI source locations presented in Fig. 4. DLSP was clearly impacted by point sources located in counties to the east and southeast (Columbia, WI; Milwaukee, WI; Cook, IL; DuPage, IL; Will, IL; Lake, IL), southwest (Grant, WI; Linn, IA), and northwest (Vernon, WI; Allamakee, IA).

Fig. 6a and b are pollution roses of GEM and RM concentrations measured in Milwaukee that were greater than the *ST*. The figures show that elevated concentrations of GEM and RM came predominantly from 110° to 240° and 0° to 45°. A comparison of the source locations in Fig. 4, with the data presented in Fig. 6a and b, shows that the elevated concentrations arriving at the measurement site from the south are likely due to emissions from point sources in the following counties: Milwaukee (WI), Kenosha (WI), Cook (IL), Lake (IL), DuPage (IL), Will (IL), Lake (IN), Jasper (IN) and Porter (IN).[43] The elevated concentrations that approached the measurement site from the east (the direction of Lake Michigan) and west were probably due to emissions from point sources to the north and south of Milwaukee (Fig. S4)† combined with air recirculation during lake breeze meteorological effects.[56–58] Elevated concentrations from the east were also caused by point sources in Michigan counties Muskegon and Ottawa on the opposite shore of Lake Michigan impacting the site during

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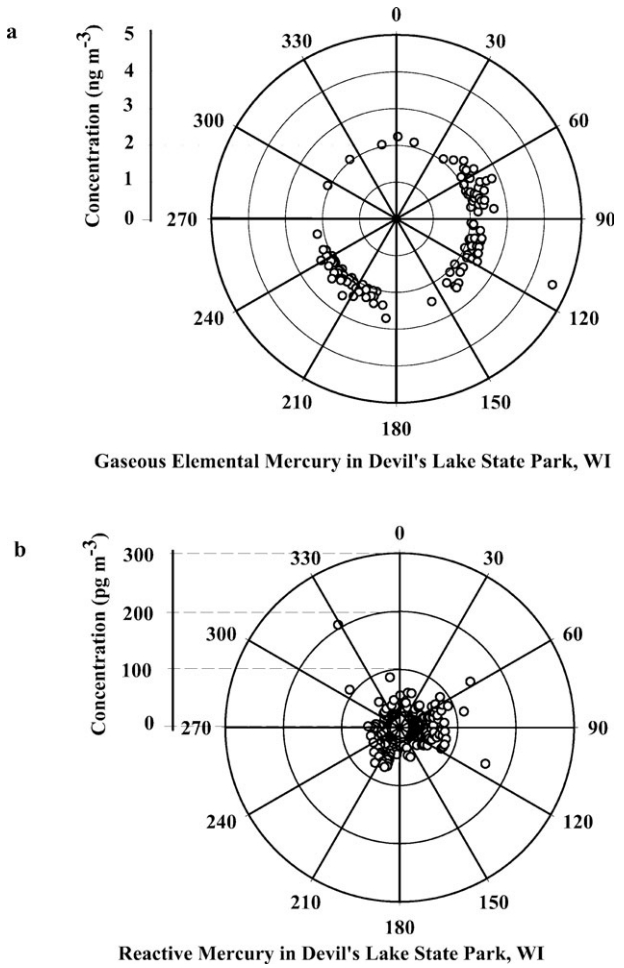

Fig. 5 Pollution roses for atmospheric gaseous elemental mercury (a) and reactive mercury (b) for Devil's Lake State Park, WI, between April 2003 and March 2004. Both pollution roses only show data above the point source threshold.

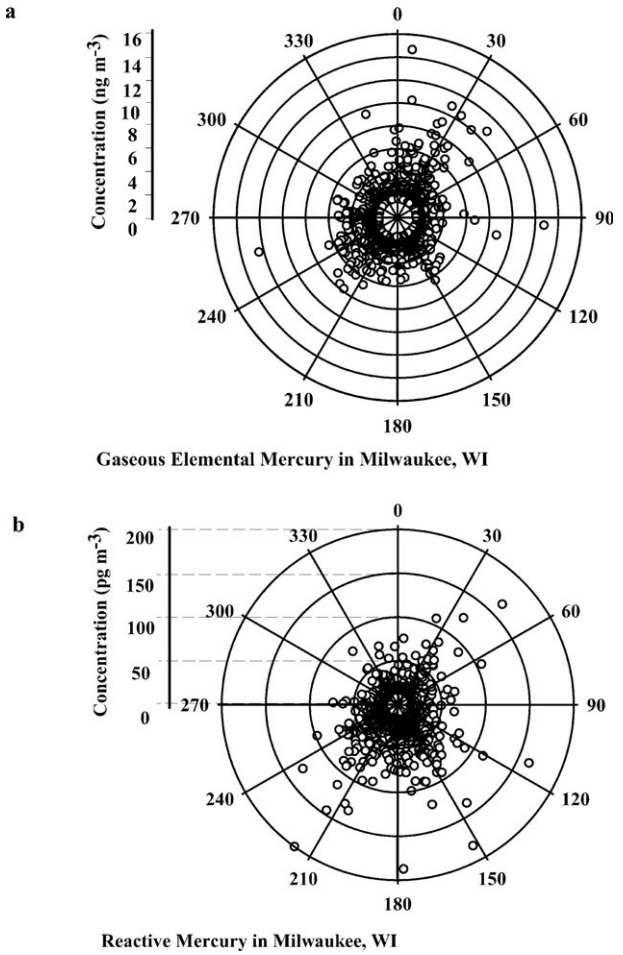

Fig. 6 Pollution roses for atmospheric gaseous elemental mercury (a) and reactive mercury (b) at Milwaukee, WI between July 2004 and May 2005. Both pollution roses only show data above the point source threshold.

easterly wind patterns (Fig. S5).† It is possible that some of the mercury observed from non-point source directions could have been emitted by small point sources (less than 50 lbs yr^{-1}) or unregistered point sources distributed throughout the city. The recirculation of air during lake breeze events made it difficult to distinguish between recirculated emissions from known point sources, and emissions from unregistered point sources. In order for effective air quality management decisions to be made for Milwaukee, the effect of the lake breeze system on the recirculation of known RM and GEM point source emissions will need to be determined.

Conclusions

A significant urban excess of atmospheric mercury was observed at Milwaukee when GEM, PHg, and RGM concentrations were compared with rural measurements made at DLSP. The urban excess indicated that the source–receptor relationships for the two sites were different. The Milwaukee site was impacted by point source emissions more often than the DLSP site, due to the higher number, greater magnitude and closer proximity of point sources surrounding the city,[43] and to a lesser degree by recirculation of emissions caused by lake breeze meteorology.[57,58] The relationship between the point sources and the measurement sites was revealed by pollution rose analyses of GEM, PHg and RGM concentration data and the locations of point sources (>50 lbs yr^{-1}) listed in the NEI.[43] These relationships were supported by HYSPLIT back trajectory models presented in the ESI.† The contributions of point sources to GEM, PHg, and RGM concentrations at Devil's Lake State Park, WI, USA, and Milwaukee, WI, USA, were estimated using a simple data analysis method that sorted the concentration data into point source and non-point source classes. Respective point source contributions to annual average concentrations at DLSP and Milwaukee were: (i) 2.4% and 33% for GEM; and (ii) 48% and 64% for RM. The analysis demonstrated that point sources exerted significant impacts on concentrations measured at receptor sites located tens of kilometres downwind. This means that mitigation of wet and dry mercury deposition to these receptors would be strongly affected by reductions in PHg and RGM emissions from point sources located nearby. In contrast, local reductions in GEM emissions would yield much smaller decreases in mercury deposition from the global pool at short and medium distances from the emission sources. The large difference in source–receptor relationships of

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each of the mercury species must be considered during the development of mercury control policies.

This study demonstrates the value of high time resolved mercury concentration and speciation measurements made over 1 year at receptor sites deemed sensitive to mercury impacts from point sources. From such measurements, it is possible to estimate the point source impacts on atmospheric mercury concentrations, and therefore dry and wet deposition, using the data analysis method described. These impact estimates can then be used to assess the potential success of proposed air quality management schemes on mitigating the deposition of reactive mercury to sensitive receptors.

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