

ATMOSPHERIC SOURCES, TRANSPORT AND DEPOSITION OF MERCURY IN MICHIGAN: TWO YEARS OF EVENT PRECIPITATION

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Abstract. To assess the sources, transport and deposition of atmospheric mercury (Hg) in Michigan, a multi-site network was implemented in which Hg concentrations in event precipitation and ambient samples (vapor and particulate phases) were determined. Results from the analysis of 2 years of event precipitation samples for Hg are reported here. The volume-weighted average Hg concentration in precipitation was 7.9, 10.8 and 10.2 ng/L for the Pellston, South Haven and Dexter sites, respectively. Yearly wet deposition of Hg for 1992-93 and 1993-94 was 5.8 and 5.5 $\mu\text{g}/\text{m}^2$ at Pellston, 9.5 and 12.7 $\mu\text{g}/\text{m}^2$ at South Haven and 8.7 and 9.1 $\mu\text{g}/\text{m}^2$ at Dexter. A spatial gradient in both the Hg concentration and wet deposition was observed. Northern Michigan received almost half the deposition of Hg recorded at the southern Michigan sites. The concentration of Hg in precipitation exhibited a strong seasonal behavior with low values of 1.0 to 2.0 ng/L in winter and maximum values greater than 40 ng/L in summer. The spring, summer and autumn precipitation accounted for 89 to 91% of the total yearly Hg deposition. Mixed-layer back trajectories were calculated for each precipitation event to investigate the meteorological history and transport from potential Hg source regions. Elevated Hg concentrations were observed with air mass transport from the west, southwest, south, and southeast. At each of the sites precipitation events for which the Hg concentration was in the 90th and 10th percentile were analyzed for trace elements by ICP-MS to investigate source impacts.

Keywords: Mercury, wet deposition, precipitation, trace elements, regional transport, trajectories

1. Introduction

Atmospheric transport and deposition of Hg has been recognized as an important link in the cycling of Hg in the environment (Lindqvist, 1985; Lindberg *et al.*, 1991). Anthropogenic Hg sources in the U.S. and initial estimates of their annual emissions into the atmosphere have recently been reported (U.S. EPA, 1994). Since anthropogenic sources for Hg in the U.S. are numerous and generally not well characterized, an accurate emissions database which includes both anthropogenic and natural sources is not yet available. This fact coupled with an incomplete understanding of atmospheric processes for Hg limit the applicability of deterministic models in predicting the atmospheric behavior and deposition of Hg over short temporal and large spatial scales. To investigate the local and regional transport and deposition of Hg, accurate, long-term measurements at multiple receptor locations should be obtained. To attempt to diagnose the source(s) and source regions culpable for the observed Hg, concentrations of Hg in ambient air (both vapor and particle phases), and in event precipitation should be measured together with complementary chemical and meteorological parameters at multiple sites over time periods longer than one year.

Research conducted in Scandinavia (Brosset, 1987; Iverfeldt, 1991) identified a spatial gradient in the concentration of Hg in precipitation from northern to southern Sweden. Meteorological analysis indicated that the elevated levels of Hg observed in precipitation were associated with transport from the heavily industrialized region to the south and southeast of Sweden. In addition, Hg concentrations were correlated with concentrations of sulfate, Cd and Pb in precipitation, which suggested a connection

between the Hg found in precipitation and anthropogenic emissions upwind (Iverfeldt, 1991).

In the Great Lakes, indirect evidence suggesting a gradient in the deposition of Hg has been reported by Nater and Grigal (1992). A two fold increase of Hg content in surficial soils was observed from western Minnesota to northeastern Michigan. To investigate these findings and to determine the sources of the Hg deposition in the Great Lakes region, sampling for Hg and other chemical and meteorological parameters was conducted in Michigan from 1992 to 1994. Measurements included event precipitation for the two years of the study, and vapor and particulate phase Hg, as well as other atmospheric constituents on an every 6th day basis for the second year of the study. Data reported in this paper are limited to Hg, major anions and trace elements. Sites were chosen to provide an adequate spatial resolution to characterize regional transport and to document spatial differences in the deposition of Hg in Michigan. In addition, source regions and source types culpable for the Hg measured were investigated using meteorological, elemental, and chemical data for precipitation events with elevated Hg.

2. Materials and Methods

Event precipitation was collected for two years (March 1992 - March 1994) at three sites in Michigan: Pellston, South Haven and Dexter (Figure 1). The site near Pellston located at the University of Michigan Biological Station is also a National Atmospheric Deposition Program (NADP) site. The region around Pellston is mixed forest with areas of low intensity farming. The site in South Haven was located in a rural agricultural area 3 km east of Lake Michigan. The Hg monitoring at that site was collocated with atmospheric measurements as part of several studies investigating the loading of Hg and other toxic compounds to the Great Lakes. The Dexter site, located 25 miles northwest of Ann Arbor, MI is a U.S. EPA National Dry Deposition Network (NDDN) Site.

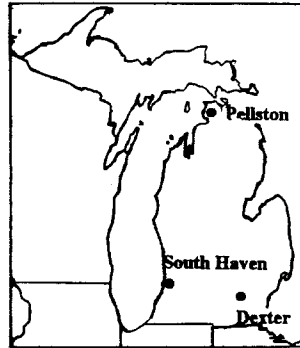


Fig. 1. Hg Measurement sites in Michigan

Precipitation samples were collected into a 10 L borosilicate glass (BSG) vessel using an MIC-B automatic collector (MIC Co., Richmond Hill, Ontario) with a Teflon-coated funnel. After each precipitation event, the sample was transferred from the 10 L collection vessel to a 1 L BSG bottle. With a funnel inlet area of 0.2 m^2 , precipitation events of 0.13 cm or greater provided sufficient volume for analysis. The 10 L vessel was rinsed with ultra-pure water (containing $<0.2 \text{ ng Hg/L}$) after collection of each sample and was replaced biweekly. A field blank was performed biweekly to ensure that the system was contaminant-free. A Teflon-covered pad was installed on the underside

of each funnel cover to shield the funnel from windblown dust and other particulate matter.

All sample bottles and glassware used in sample analysis were rigorously cleaned in an 11-day procedure involving an acetone rinse, detergent wash, multiple rinses with ultra-pure water, heating in 3M HCl at 80°C, and two separate soaking periods in 0.6M HNO₃ followed by extensive rinsing in ultra-pure water in a Class 100 clean room (Rossmann and Barres, 1991). Ultra-clean techniques were used in all phases of sample collection and analysis. Samples were shipped by overnight mail to the University of Michigan Air Quality Lab (UMAQL) in Ann Arbor, MI. A Belfort rain gauge located at each site was used to determine the volume and time of each precipitation event.

When samples were received at UMAQL a subsample was poured off and acidified for analysis of trace metals by inductively coupled plasma mass spectrometry (ICP-MS). A separate subsample was poured off for analysis of pH and major ions by ion chromatography. The remaining sample was oxidized with bromine monochloride to a 1% solution for analysis of total Hg by cold vapor atomic fluorescence spectrometry (Fitzgerald and Gill, 1979). At the start of the study, the detection limit for total Hg, defined as 3σ of the reagent blank, was 0.095 ng/100 mL sample. The detection limit was reduced to the current level of 0.015 ng/100 mL sample with the use of more purified HCl and bromine salts. All samples were initially analyzed in duplicate. After the first 6 months through the end of the study 50% of the samples were analyzed in duplicate. The average analytical precision over the duration of this study was 6%.

Procedures required to implement and operate a multi-site network for collection and analysis of event precipitation for Hg are described in Hoyer and Keeler (1994a). The manuscript details experiments including collocated manual and automatic collectors, interlaboratory comparisons and investigation of the stability of Hg in precipitation. Results from the first year of network operation are reported elsewhere (Hoyer *et al.*, 1993).

Three-day backward mixed-layer trajectories were calculated for each day on which precipitation was collected. Air mass trajectories define the most probable path of an air mass before arriving at a specified receptor site. Trajectories were calculated from upper air data collected at National Weather Service monitoring stations (Heffter, 1980).

3. Results and Discussion

3.1 Hg CONCENTRATION IN PRECIPITATION

During the two years of event precipitation collection, a total of 153 samples were collected at Pellston, 188 at South Haven and 181 at Dexter. The two year mean Hg concentration in precipitation was 7.9 ng/L at Pellston, 10.8 ng/L at South Haven and 10.2 ng/L at Dexter (Table I).

Variability in the precipitation Hg concentration between the sites was clearly evident. The maximum concentration observed at the northern Michigan site in Pellston was approximately 20% lower than the maximum at the southwestern Michigan site in South Haven and the southeastern Michigan site in Dexter. Precipitation events with elevated Hg were also less frequent at the Pellston site than at the two southern Michigan sites. The standard deviation of the Hg concentration in precipitation at South Haven was

higher than at the other two sites largely due to the elevated Hg concentrations in several summer samples collected at the site.

TABLE I
Total Hg in precipitation in Michigan (ng/L), Mar 1992-Mar 1994

SITE	n	Volume-Weighted			90th Percentile
		Average	Std Dev	Range	
Pellston, MI	153	7.9	7.4	1.4 - 47.6	16.6
South Haven, MI	188	10.8	11.2	1.2 - 59.5	25.7
Dexter, MI	181	10.2	9.8	1.8 - 55.2	19.6

The concentration of Hg in precipitation varied seasonally at each of the sites (Figure 2). Average Hg concentrations for the spring and summer were a factor of two greater than those in winter, except at the Dexter site where two high concentration events occurred in winter 1994 which elevated the mean. Year-to-year differences in the seasonal average Hg concentration in precipitation are evident (Figure 2). The average Hg concentration was 14% and 25% lower in the summer of 1992 at South Haven and Dexter, respectively, than in summer of 1993. This may have resulted from lower than average ambient temperatures during the summer 1992, but more years of sampling would be required to adequately characterize seasonal differences on a year-to-year basis. The average Hg concentration in precipitation in winter 1994 at Dexter was higher than that in winter 1993. This was uncharacteristic when compared to measurements at the other sites. This elevated average was due to one event in Feb 1994 and two events in Mar 1994 for which the concentration of Hg was greater than 20 ng/L. These storms deposited precipitation with slow transport from the east (Detroit metropolitan area).

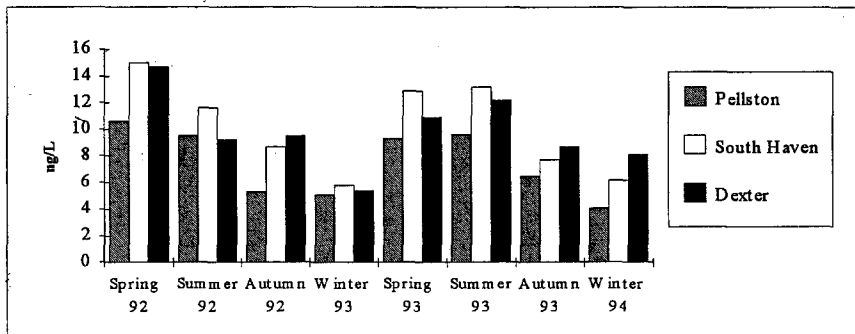


Fig. 2. Volume-weighted average Hg concentration in precipitation 1992 - 1994.

Many factors, both chemical and physical, may influence the seasonal variation in the concentration of Hg in precipitation. Those factors include precipitation type, source influences, atmospheric oxidant concentration (Munthe *et al.*, 1991), in-cloud processes (Borys *et al.*, 1988), meteorology and chemical/physical aspects of the aerosol population. Borys *et al.* (1988) reported that the concentration of constituents in frozen precipitation may be strongly influenced by in-cloud growth processes (accretion and vapor deposition). Several in-cloud processes are potentially important factors which

may control the concentration of Hg in cloud droplets and subsequently in precipitation. These include in-cloud temperature, turbulence, aerosol size distribution, and chemical properties of the atmospheric aerosol incorporated into the cloud system. Chemical parameters of importance include oxidant concentrations and the particulate and vapor species in the air which feeds the precipitating cloud system (Munthe *et al.*, 1991).

3.2 Hg WET DEPOSITION

During the first year of measurement, the total Hg wet deposition observed at South Haven was $9.45 \mu\text{g}/\text{m}^2$ while at Pellston and Dexter the wet deposition was 5.79 and $8.66 \mu\text{g}/\text{m}^2$, respectively (Table II). The wet deposition of Hg was substantially higher in the second year of sampling at South Haven, but nearly identical for both years at Pellston and Dexter. The increase in deposition measured at South Haven resulted from an increased precipitation rate. Event Hg wet deposition measurements in Underhill, VT made from Dec 1992-'93 also demonstrate the importance of precipitation amount on yearly loading (Burke *et al.*, this volume). During this one year period, the average volume-weighted Hg concentration at the site in Vermont ($8.3 \text{ ng}/\text{L}$) was similar to that observed at Pellston ($7.9 \text{ ng}/\text{L}$). However, Hg deposition observed at the Vermont site was $9.26 \mu\text{g}/\text{m}^2$, substantially higher than that measured at Pellston. The measured values at the Pellston site are also lower than values estimated from measurements in northern Wisconsin at similar latitudes (Fitzgerald *et al.*, 1991), and from measurements of weekly precipitation collected in Ontario (Mierle, 1990). These differences may be due to sampling frequency, precipitation amount and/or differences in source impacts.

A spatial gradient in the deposition of Hg was observed in Michigan in this study. South Haven received 1.6 and 2.3 times more Hg deposition than Pellston in the two respective years of sampling. While the spatial differences in Hg wet deposition are clearly a function of the different amounts of precipitation received, the difference in Hg concentration at the sites also contributes substantially to the regional gradient in wet deposition of Hg. In 1992-'93 South Haven received 1.2 times more precipitation than Pellston and 1.6 times more Hg was deposited in South Haven. In 1993-'94 South Haven received 1.6 times more precipitation than Pellston, but 2.3 times the Hg wet deposition. The yearly precipitation totals in 1992-'94 were within 10% of the 30-year climatological average for these regions in Michigan (NOAA, 1992). However, the precipitation received at South Haven in 1993-'94 was 24% higher than the 30-year average.

Seasonal variation in the wet deposition of Hg was also observed in both years of measurement with summer storms delivering 3 to 5 times more Hg than winter storms (Figure 3). Note that while the volume-weighted average concentration of Hg in precipitation collected at Dexter was elevated in winter 1994 compared to winter 1993, the deposition for this season was similar in the two years of study. Hg wet deposition occurring during winter of 1992-'93 accounted for only 6% of the total Hg deposition at Pellston, 7% at South Haven and 15% at Dexter. Wintertime Hg wet deposition in 1993-'94 contributed 7% of the yearly total at both Pellston and Dexter and 8% at South Haven. The largest portion of the annual Hg wet deposition occurred in the spring and summer months at each of the sites with 34% of the total being deposited during those seasons. Hg wet deposition during the autumn periods averaged 22% of the total annual wet deposition at the three sites.

Table II
Deposition of Hg in precipitation ($\mu\text{g}/\text{m}^2$), Mar 1992-Mar 1994

SITE	Event Avg	Event Max	1992-'93 Deposition	1993-'94 Deposition	1992-'93 Precip Amnt, cm	1993-'94 Precip Amnt, cm
Pellston, MI	0.07	0.51	5.79	5.54	73	71
South Haven, MI	0.12	0.85	9.45	12.67	89	116
Dexter, MI	0.10	0.98	8.66	9.11	87	88

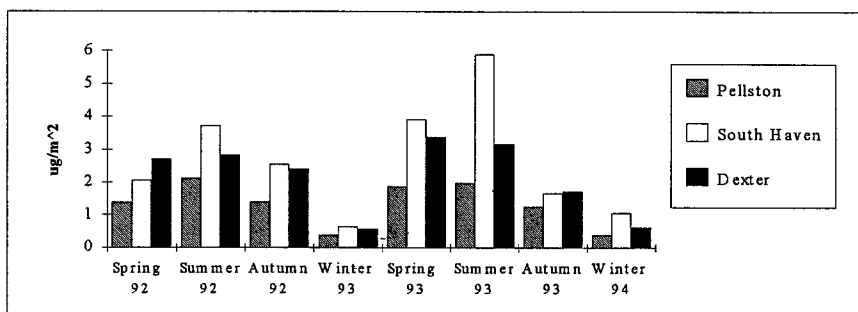


Fig. 3. Mercury wet deposition by season in Michigan 1992 - 1994.

Individual precipitation events resulted in Hg wet deposition from $0.002 \mu\text{g}/\text{m}^2$ to a high of $0.984 \mu\text{g}/\text{m}^2$ measured at Dexter in 1992-'93 which represented 10.8% of the total annual deposition. Elevated Hg deposition events (90th percentile by site) were generally the result of large volume precipitation events with average or above average Hg concentration. At all sites precipitation events in the 90th percentile for Hg concentration accounted for 18 to 22% of the cumulative Hg wet deposition for the two-year period while the 10th percentile events accounted for only 3 to 6% of the Hg wet deposition.

For this study, precipitation events which accumulated less than 0.13 cm did not produce sufficient volume for a complete chemical and elemental analysis. The contribution of these events was estimated using the average concentration in events which accumulated 0.13 to 0.25 cm for the month during which the low-volume event occurred. This estimation procedure was used because of the seasonal variation in Hg concentration and the relationship between precipitation and concentration amount (lower volume events have a higher Hg concentration, Hoyer and Keeler, 1994b). The estimated deposition from these events was not included in Table II. These small events were estimated to have contributed 4% of the two year total at Pellston, 2% at South Haven and 3% at Dexter. These data indicate that collection of events less than 0.13 cm is not important in accurately assessing wet deposition when event samples are collected throughout the year.

3.3 TRANSPORT AND SOURCE CHARACTERISTICS

Air mass trajectories are a useful tool in the analysis of air and precipitation chemistry measurements (Pierson *et al.*, 1986; Keeler *et al.*, 1990; Moody and Galloway, 1988). However, interpretation of trajectory measurements should not be undertaken without full knowledge of the associated uncertainties and the limitations inherent in the meteorological measurements on which they are based (Kahl, 1993). An initial investigation of the meteorological associations and potential sources influencing the Hg concentrations measured in event precipitation samples was performed. Mixed-layer back trajectories were plotted for each event and a suite of trace elements in samples with elevated Hg were analyzed. The relative abundance and combination of the elements quantified were used to characterize potential sources. Previous work has demonstrated that trace element components of a transported aerosol mass and metal ratios reveal specific source influences (Rahn and Lowenthal, 1984; Keeler and Samson, 1989).

The highest Hg concentrations (90th percentile) were typically associated with air mass transport from the west, southwest, south and southeast of the sites in Michigan (Figures 4a-b). Although less frequently, events elevated in Hg were also observed with transport from the north and east to each of the sites. Upwind stagnation was evident for a few of the 90th percentile events which has been previously shown to result in elevated anion levels in precipitation (Pierson *et al.*, 1989; Moody and Samson, 1989). Trajectories plotted for 10th percentile events indicated that transport from the north, northwest, and east (frequently accompanied by rapid advection) often resulted in low Hg concentration in precipitation.

Trace element concentrations in precipitation events with high Hg at the South Haven site were investigated for potential source markers. On 11 May 94 and 24 May 94 transport to South Haven was from the northwest and these events had elevated concentrations of Hg, V, Pb, Fe, Zn, Mn, Cu, NO_3^- and SO_4^{2-} (elevated in comparison to concentrations in other samples in the 90th and 10th percentile for Hg). The sample collected on 24 May 94 also contained elevated levels of As and Se and especially elevated NO_3^- and SO_4^{2-} concentrations. These elemental concentrations indicate potential contributions from metals processing, iron and steel manufacturing, and oil and coal combustion.

During the summer months, with prevailing winds from the southwest, the South Haven site received storms with feed air from one of the largest Hg emission source areas in the region. According to the draft EPA Emissions Inventory (U.S. EPA, 1994), Cook County and the three counties surrounding it at the southern tip of Lake Michigan emit more than 5.0 tons of Hg/year. The number and variety of Hg and other trace element source types are diverse in this region so that characterization of Hg sources using measurements at receptor sites requires analysis of the intersection of a large number of precipitation events representing different meteorological conditions. Events elevated in Hg concentration arriving at the South Haven site from the southwest all had elevated concentrations of Pb, Fe, Zn, Mn, As, Cd, Se, V, NO_3^- and SO_4^{2-} reflecting a variety of possible source influences including coal combustion, oil combustion, metals processing (ferrous and non-ferrous) and incineration.

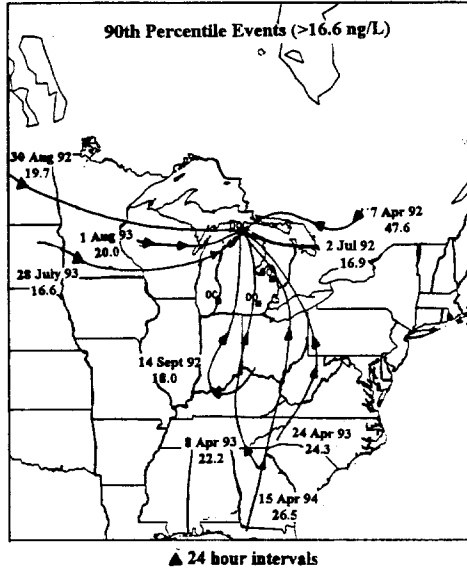


Fig. 4a. Pellston Mixed-Layer Trajectories

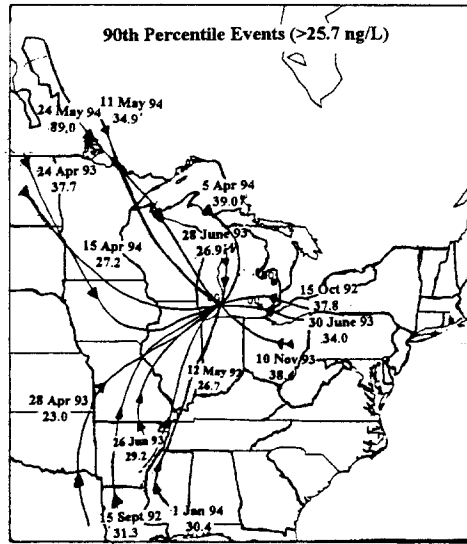


Fig. 4b. South Haven Mixed-Layer Trajectories

Each trajectory is labeled with the date of precipitation and concentration of Hg in ng/L.

In general, when Hg was elevated other trace metals in the sample were also elevated indicating common source(s) and/or source regions. Also, when a sample was low in Hg concentration it was generally accompanied by low elemental and anion levels. However, for samples in the 10th percentile for Hg concentration, there were two events at Pellston, one at South Haven and one at Dexter which had an elevated level of one or more other elements. A snow storm at Pellston on 19 Dec 1993 with Hg concentration of 2.0 ng/L was highly elevated in Ni, Pb, Fe, Zn, As, Cd, Se, Cu and Cr and slightly elevated in Mn and V. The trajectory for this event indicates air mass transport from west-southwest of the site. On a second occasion at Pellston (10 Sept 1993) a precipitation sample had 2.6 ng/L of Hg but contained highly elevated Pb and Cd. Air mass transport for this event was very rapid and directly out of the northwest traveling over potential metals sources in the Upper Peninsula of Michigan. On 19 Feb 1994 a snow sample at Pellston contained 2.8 ng/L Hg and elevated Mn, V and Cr indicating potential input from oil combustion sources and non-ferrous metal manufacture.

At South Haven only one of the 10th percentile events contained elevated levels of trace metals other than Hg. Frozen precipitation that fell on 25 Feb 1994 contained 3.1 ng/L Hg but had elevated Cu and a moderate level of Pb, Zn and Mn. Trajectories plotted for events in the 10th percentile for Hg concentration indicated that a source region contributing to elevated Hg may, on occasion, contribute to very low levels of Hg in precipitation. Clearly, factors other than air mass origin were affecting the constituent load in precipitation. Precipitation type, storm type (convective vs. frontal), wind speed, other boundary layer parameters, and the Hg chemistry itself could have affected the Hg concentration in precipitation from storms which arrive from known source regions.

4. Conclusions

Analysis of two years of event precipitation samples at three sites in Michigan has resulted in several findings to date. A gradient in both the Hg concentration and wet Hg deposition in Michigan was observed with the northern Michigan site receiving less Hg than the southern Michigan sites. The Hg concentration in event precipitation samples varied by season with Hg concentrations two times greater during spring and summer months than during winter. The data suggest that the sources of Hg measured at the sites are of regional origin (within and outside of Michigan) and that proximity to known anthropogenic sources significantly influence the concentration and wet deposition of Hg in the Great Lakes basin. Assessment of event deposition over a two-year period has enabled an accurate measurement of total Hg loading to the lower peninsula of Michigan in the Great Lakes basin. This study provides the first spatially resolved event Hg wet deposition data in the region. These results can be used to improve model estimates of the wet Hg flux in this region.

Several studies have now documented a seasonal trend in the Hg concentration in precipitation and in the wet deposition of Hg (Burke *et al.*, this volume; Glass *et al.*, 1986). This observation is possibly due to combined effects of chemistry and physical processes (e.g. ozone and other in-cloud oxidant concentration, physical factors in-cloud involved in the growth of rain droplets and ice crystals). To elucidate the dominant factors controlling the seasonal variation of Hg in precipitation, further investigations of the concentrations and reaction rates for Hg and other ions and constituents in

precipitation are needed as well as better understanding of Hg-containing aerosols, their size distributions, concentrations, and their role in precipitation formation.

Mixed-layer trajectories indicate that the dominant sources of Hg to the Michigan sampling sites are located to the west, southwest, south and southeast. Investigating the cross-section of several events that contained elevated Hg concentration provides the ability to separate meteorological influences from source effects on precipitation chemistry. Specific source types can also be identified with the use of tracer compounds measured in the precipitation.

In order to investigate sources utilizing the receptor-based approach employed in this study, short duration (event) precipitation samples need to be collected for at least one year in order to assess sample to sample variability, inter-site differences, seasonal variation and meteorological factors which can drastically alter precipitation chemistry.

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