

DELINEATION OF A POTENTIAL GASEOUS ELEMENTAL MERCURY EMISSIONS SOURCE IN NORTHEASTERN NEW JERSEY

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Summary

In order to assist in the identification of a potential mercury (Hg) emissions source in northeastern New Jersey, this project was undertaken to delineate geographic areas in which this source may be located. To this end, the objectives of this project were to collect gaseous elemental mercury (GEM) concentrations at a site to the east of the putative emissions source on Staten Island, New York and analyze these results with those collected by the NJDEP at their air monitoring site (ELAB) in Elizabeth, New Jersey. Additional local measurements were to be obtained near possible emissions sources. From September 30, 2011 to September 3, 2012, more than 200 days of GEM data were collected at the Staten Island site and together with wind speed and direction data from the NOAA Bergen Point West Reach naval observatory station were analyzed for source trajectories of GEM in the region. GEM concentration and wind direction data from the ELAB site for the same period of time were also analyzed. Local scale measurements of GEM were limited by access to appropriate sites and difficulties maintaining stable calibration of the portable Hg analyzer.

Directional analysis of the number frequency and concentration-weighted distributions of GEM peaks with concentrations $>4 \text{ ng m}^{-3}$ at the ELAB site revealed a single source direction just east of south (bearing 173.4°). Similar analysis of GEM results for the Staten Island site revealed two source directions, one to the west (bearing 280.5°) and a second to the southwest (bearing 213.8°). The intersections of the two Staten Island trajectories with the ELAM trajectory delineate two possible GEM source areas. The first is centered in Rossville, NY on Staten Island (40.5514 N , 74.1947 W) and the second near Pralls Island in the Arthur Kill along the eastern border of Linden, NJ (40.6119 N , 74.2039 W). Temporal analysis of GEM peaks for the three source directions indicates that the eastern Linden source may contribute more GEM to elevated measurements recorded in Elizabeth than the Rossville source.

Background

Monitoring studies of atmospheric mercury in northeastern New Jersey report that gaseous elemental mercury (GEM) concentrations episodically exceed the average ambient level of 2 ng m^{-3} (Goodrow et al., 2005; Aucott et al., 2009). For example, in a study of GEM concentrations at the NJDEP air monitoring site in Bayonne, New Jersey from September 2001 to February 2003, spikes of elevated GEM ($>6 \text{ ng m}^{-3}$) lasting 15 minutes to a few hours and occurring primarily between midnight and 5am were observed. During one such event that occurred between 2am and 5am on June 30, 2002, GEM levels sustained an hourly averaged concentration of 26 ng m^{-3} and a peak level of 54 ng m^{-3} (Goodrow et al., 2005). Similar spikes in atmospheric mercury, including gaseous elemental mercury, reactive gaseous mercury, and

particulate mercury, were observed at Elizabeth and New Brunswick, New Jersey during the period of 2003 to 2006 (Aucott et al., 2009).

No relationship between the occurrence of episodic spikes in atmospheric mercury and meteorological conditions (season, temperature, precipitation) has been found, and such events are likely related to local industrial activities. Similar spikes observed in New York City were also attributed to local anthropogenic sources (Carpi and Chen 2002). In Elizabeth, elevated atmospheric mercury concentrations appear to correlate with a southerly wind direction (Aucott et al., 2009), indicating a possible location of a mercury point source, but no known source of mercury emissions lies to the south of the Elizabeth monitoring site. An analysis of Elizabeth monitoring results from 2010 again showed a source to the south. A similar directionality was also apparent in the 2002-2003 Bayonne dataset.

Methods

Gaseous elemental mercury (GEM) monitoring and data analysis

Gaseous elemental mercury (GEM) concentrations were measured using a Tekran 2537A continuous mercury vapor analyzer on the campus of the College of Staten Island (CSI). The intake line for the GEM analyzer (¼" Teflon tubing) was held open end down in a plastic rain shield attached to an existing meteorological tower on the roof of the CSI gymnasium (40.6040° N, 74.1484° W) approximate 22 m above the land surface. GEM concentrations were measured nearly continuously at CSI between September 30, 2011 and September 3, 2012 with breaks due to power outages or analyzer malfunction. Due to the malfunction of the Tekran's air pump, no measurements were collected in March 2012. Meteorology data corresponding to the CSI measurements were obtained from the NOAA Bergen Point West Reach naval observatory station (ID: 8519483, 40° 38.2' N, 74° 8.5' W). GEM concentrations and meteorology data for the ELAB site in Elizabeth, New Jersey (40.6414° N, 74.2084° W) were obtained from the NJDEP.

Elevated GEM concentrations that formed recognizable peaks were selected for source direction analysis. Recognizable peaks were defined as those with GEM concentrations $>4 \text{ ng m}^{-3}$. The lower limit of 4 ng m^{-3} was chosen since it exceeded the upper 95% confidence limit of all GEM concentrations measured at CSI (mean = 1.66, SD = 1.16, upper 95% limit = 3.94 ng m^{-3}). In order to exclude possibly spurious measurements of GEM concentrations $>4 \text{ ng m}^{-3}$, an additional criteria for selecting peaks was the occurrence of at least three measurements above the local baseline.

Wind speed and direction data were interpolated to GEM peak times. For ELAB results, 1 min. wind speed and direction data were averaged over the 5 min. GEM sampling times. For CSI results, 6 min. wind speed and direction data were averaged over 18 min. and interpolated to GEM sampling times.

Wind directions associated with all GEM peaks for both sites were sorted in 12 degree bins to establish dominant source directions. In addition, maximum GEM concentrations associated with all peaks for each site were sorted by wind direction in 12 degree bins to assess the strength of identified source directions. The distributions of peak frequency clusters associated with dominant wind directions were fit to a Gaussian model in order to determine the central direction and variance associated with these source directions. Potential emissions source areas were delineated by determining the intersections of the central bearing and 95% confidence

intervals bounding each GEM source direction. Finally, GEM peaks were sorted by time of day into 1 hour bins to assess temporal patterns.

Local scale GEM measurements

Methods were developed for computer control and data logging for the Lumex gaseous mercury analyzer and a rain-sheltered air intake support for field use was constructed. Access was sought and granted to the Graselli Point Industries property in Linden, New Jersey. Short term monitoring was carried out at the at the Graselli Point property on November 10-11, 2011. Local scale measurements of GEM were also conducted in western Staten Island on July 23, 2012.

Results

Gaseous elemental mercury peaks at the College of Staten Island and Elizabeth, New Jersey

During the period of monitoring at the College of Staten Island (CSI), 124 GEM peaks with concentrations $>4 \text{ ng m}^{-3}$ were recorded (Fig. 1). Over the same time period, 171 GEM peaks were measured at the ELAB site. For the CSI site, most of the GEM peaks were recorded between April and August, 2012 (Fig. 2) with the highest maximum concentrations associated with peaks recorded in July and August 2012 (Fig. 1). At the ELAB site, GEM peaks were observed throughout the year with the highest maximum concentrations occurring in peaks during May and July.

Analysis of wind directions associated with GEM peaks at CSI and ELAB

The association of GEM peaks and cumulative GEM concentrations measured at the CSI and ELAB sites with wind direction are shown in Figures 3 and 4. These plots show a dominant GEM source for the ELAB site associated winds from just east of south and two source directions for the CSI site, one to the southwest and a second to the west.

The Gaussian fit to the cluster of peaks measured during south winds at the ELAB site had an R^2 of 0.985 and a central value of 173.4° (Fig. 5). Based on the range of values within the 95% confidence limits of the Gaussian distribution, the direction from the ELAB site to the possible GEM emissions source was between 164.0° and 182.7° . For the CSI results, Gaussian fits to the two clusters of peaks for west and southwest winds had R^2 values of 0.962 and 0.919 and central values of 280.5° and 213.8° , respectively (Fig. 6). For these fits, 95% confidence limits place possible GEM emissions sources at bearings of 206.0° to 221.5° and 275.1° to 286.0° with respect to the CSI site, respectively. The highest 7 GEM peaks measured at the CSI site that were associated with southwest winds had an average duration of 2.5 ± 1.6 hours. The highest 7 GEM peaks associated with west winds had an average duration of 0.8 ± 0.3 hours.

GEM peaks associated with south winds at the ELAB site occurred in the early morning (before 6am) or in the evening (after 7pm) (Fig. 7). Peak GEM concentrations at the CSI site associated with southwest winds occurred primarily in the mid-morning (7 to 8am) while those associated with west winds occurred in the early morning and evening (Fig. 8). Distributions of GEM concentrations among hours of the day matched hourly peak frequencies for all peak clusters at both the ELAB and CSI sites (Fig. 9). Background concentrations of GEM (1 to 4 ng m^{-3}) at the CSI site measured during southwest winds occurred most frequently during overnight and morning hours (11pm to 7am) and somewhat less frequently from 11am to 10pm (Fig. 10). Background concentrations of GEM associated with west winds were recorded with relatively constant frequency during the day.

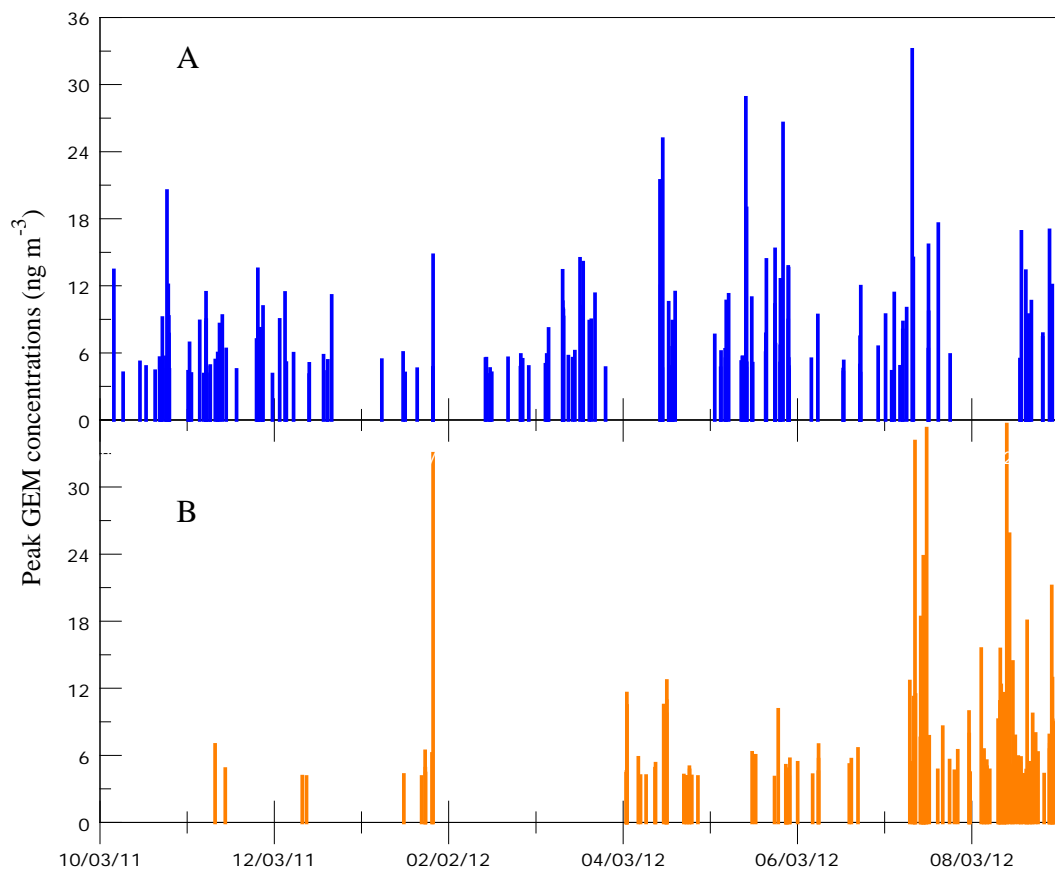


Figure 1. Maximum concentrations of GEM peaks ($>4 \text{ ng m}^{-3}$) measured at the NJDEP site in Elizabeth, New Jersey (A) and the College of Staten Island, New York (B) from October, 2011 to September, 2012. Note that GEM was not measured at CSI during March 2012.

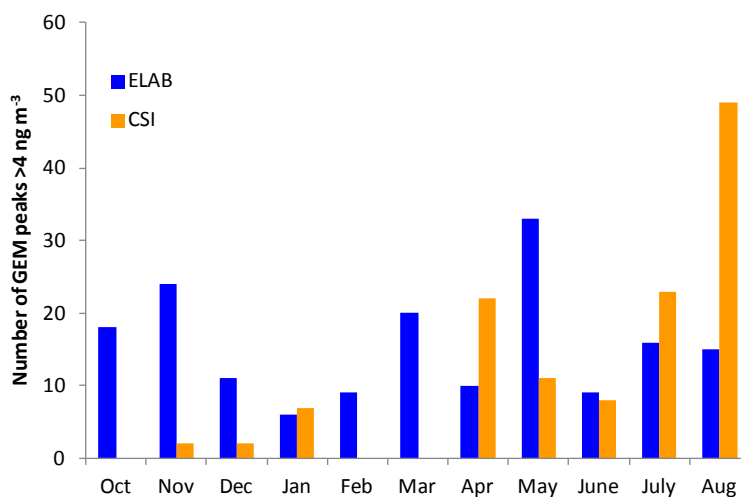


Figure 2. Monthly frequencies of GEM peaks ($>4 \text{ ng m}^{-3}$) measured at the NJDEP site in Elizabeth, New Jersey (ELAB) and the College of Staten Island, New York (CSI) from October, 2011 to August, 2012. Note that GEM was not measured at CSI during March 2012 and two peaks measured at CSI between September 1 and 3, 2012 are included with results for August.

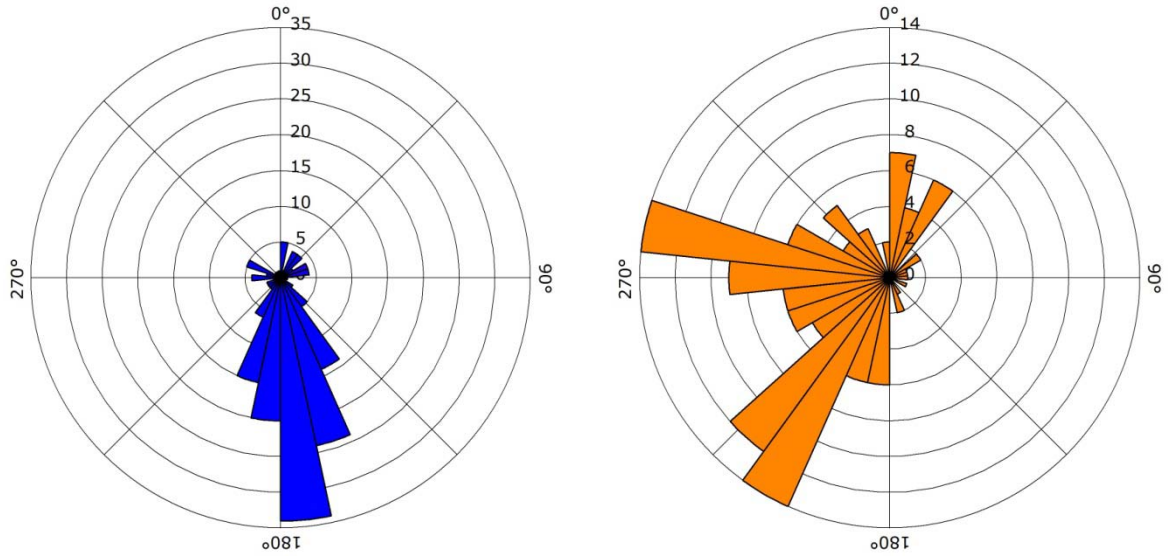


Figure 3. Frequencies of GEM peaks ($>4 \text{ ng m}^{-3}$) by wind direction (12 degree resolution) measured in Elizabeth, New Jersey (left) and at the College of Staten Island, New York (right).

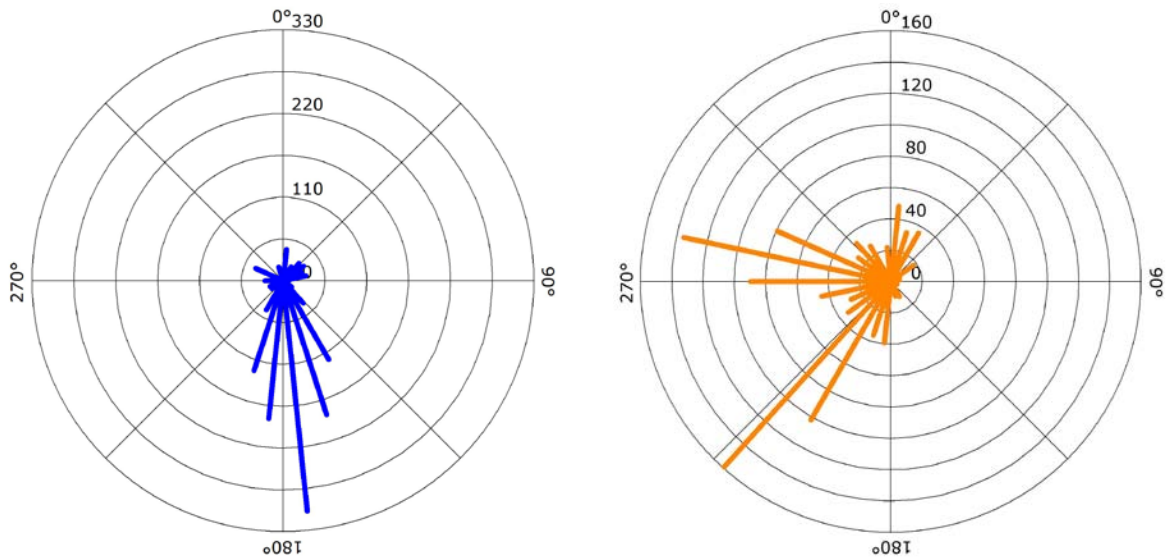


Figure 4. Distributions of cumulative GEM concentrations (ng m^{-3}) by wind direction (12 degree resolution) for peak values ($>4 \text{ ng m}^{-3}$) measured at the ELAB site in Elizabeth, New Jersey (left) and at the College of Staten Island, New York (right).

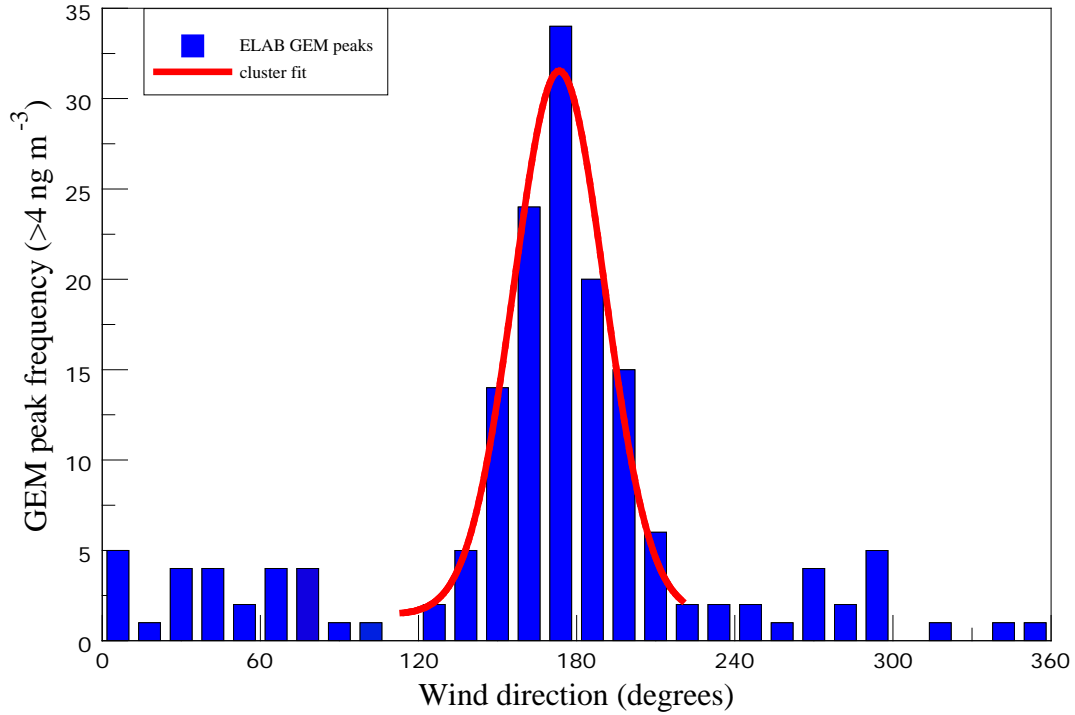


Figure 5. Model fit to the cluster of GEM peaks associated with winds from the south in Elizabeth, New Jersey.

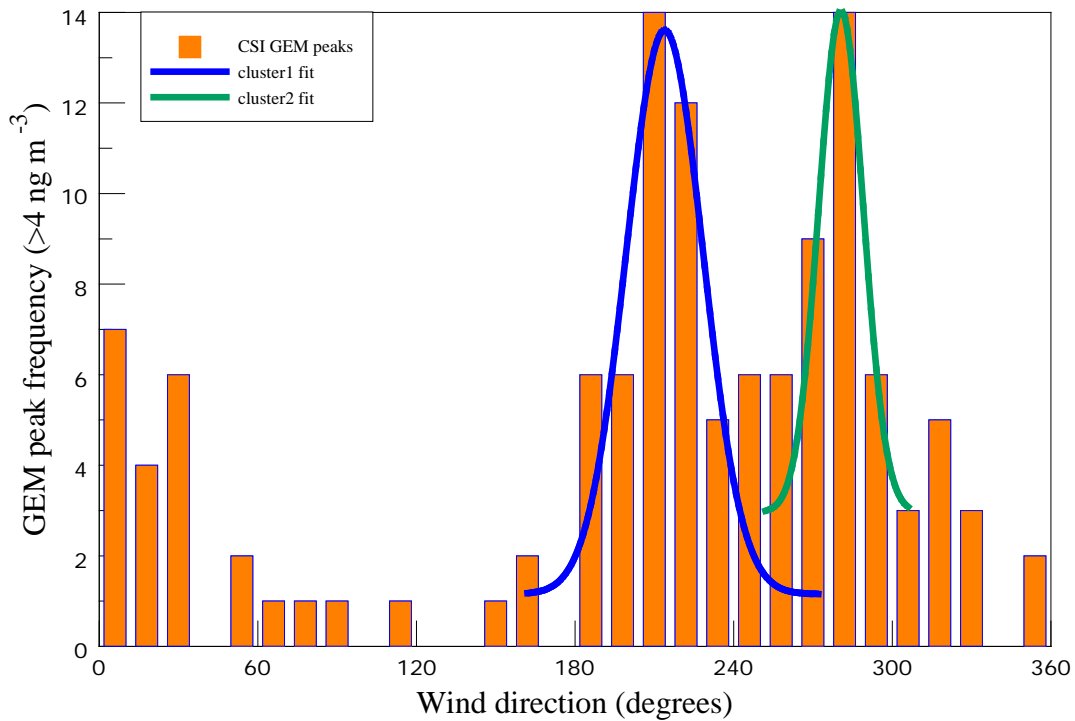


Figure 6. Model fits to the clusters of GEM peaks associated with winds from the southwest (blue line) and west (green line) at the College of Staten Island, New York.

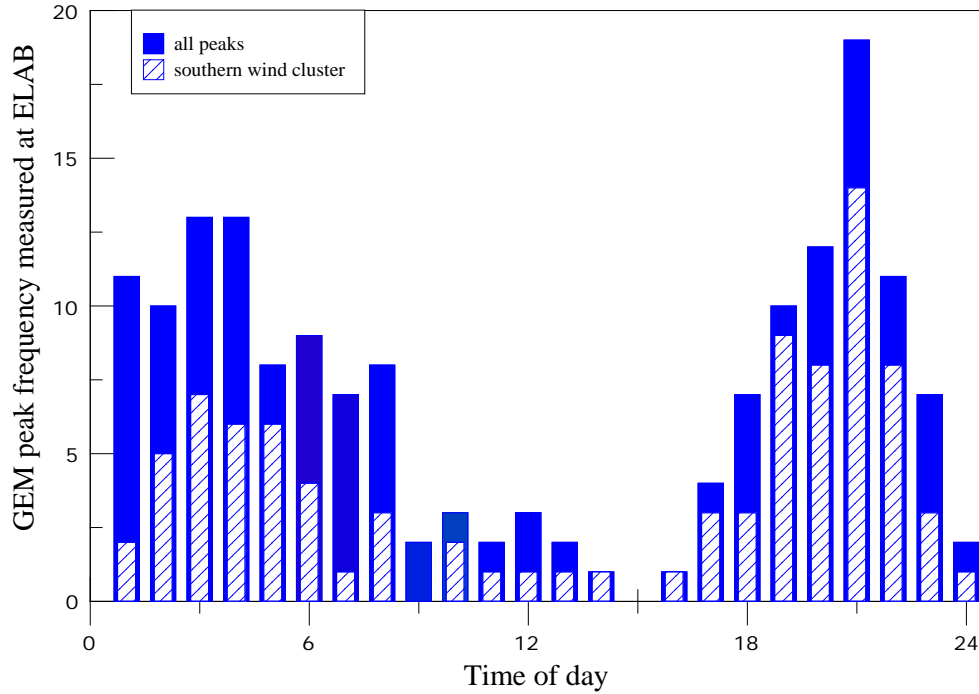


Figure 7. Frequency of GEM peaks by time of day in Elizabeth, New Jersey for all peaks (solid bars) and for the cluster of peaks associated with winds from the south (hatched bars).

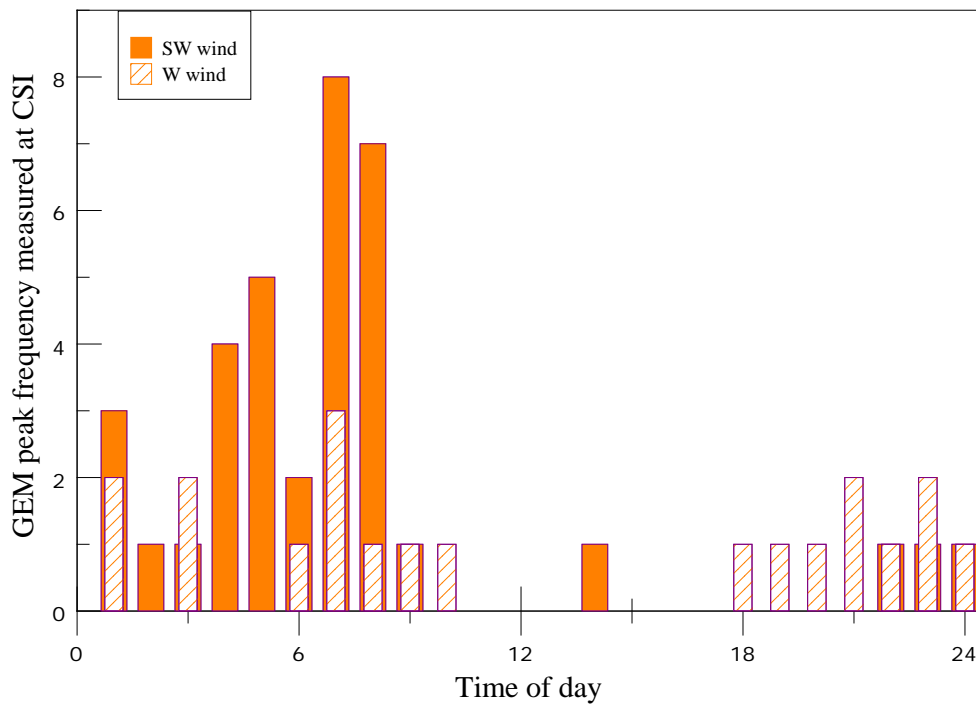


Figure 8. Frequency of GEM peaks by time of day in Staten Island, New York for the cluster of peaks associated with winds from the southwest (solid bars) and the cluster of peaks associated with winds from the west (hatched bars).

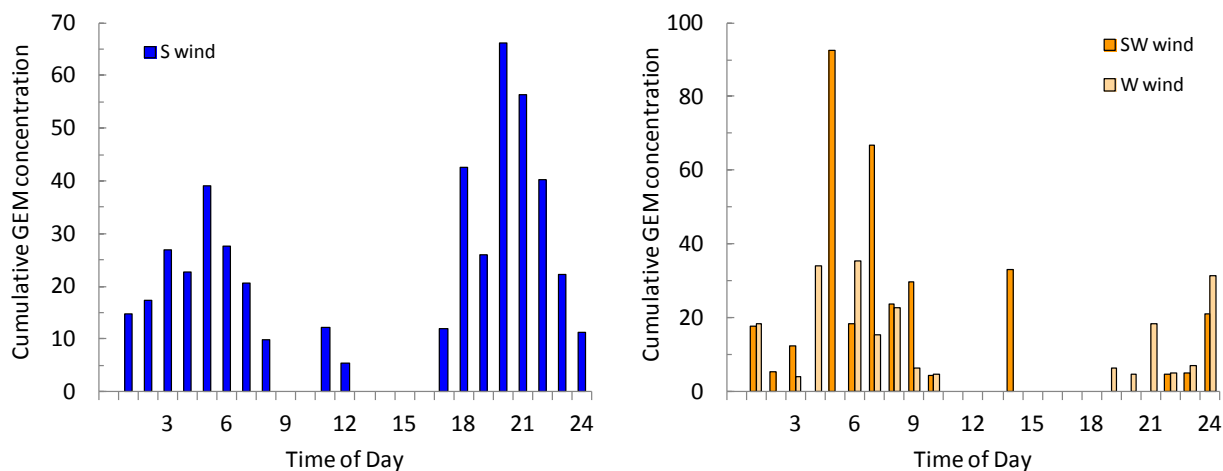


Figure 9. Distributions of cumulative GEM concentrations (ng m^{-3}) by time of day in Elizabeth, New Jersey (left) and Staten Island, New York (right). Elizabeth data are for the cluster of peaks associated with winds from the south. Staten Island data are for the clusters of peaks associated with southwest (orange) and west (light orange) winds.

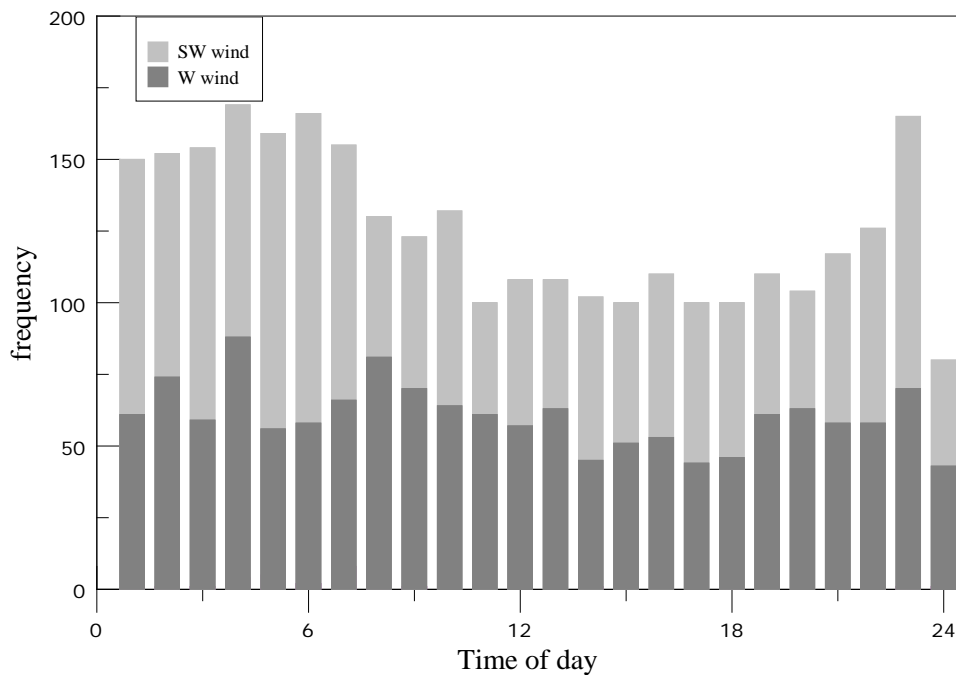


Figure 10. Frequency of background GEM concentrations (1 to 4 ng m^{-3}) by time of day in Staten Island, New York for measurements associated with winds from the southwest (light grey) and the west (dark grey).

The number and intensity of GEM peaks associated with south winds at the ELAB site was highest on Sunday through Wednesday, but decreased from Wednesday to Saturday (Fig. 11A,C). At the CSI site, GEM peaks associated with southwest winds occurred primarily on Wednesdays, Thursdays and Fridays with lower frequencies on Tuesdays, Saturdays and Sundays and no occurrences on Monday (Fig. 11B,D). In contrast, the number of GEM peaks observed at the CSI site associated with west winds decreased from Monday to Thursday, were zero on Thursday, Friday, and Saturday, and picked up on Sunday. The similarity of temporal patterns for the cluster of GEM peaks at CSI associated with west winds and the cluster of GEM peaks at ELAB associated with south winds indicates that they may have a common source. Background GEM (1 to 4 ng m⁻³) frequencies and cumulative GEM concentrations were evenly distributed among days of the week at the CSI site for both southwest and west winds (Fig. 12).

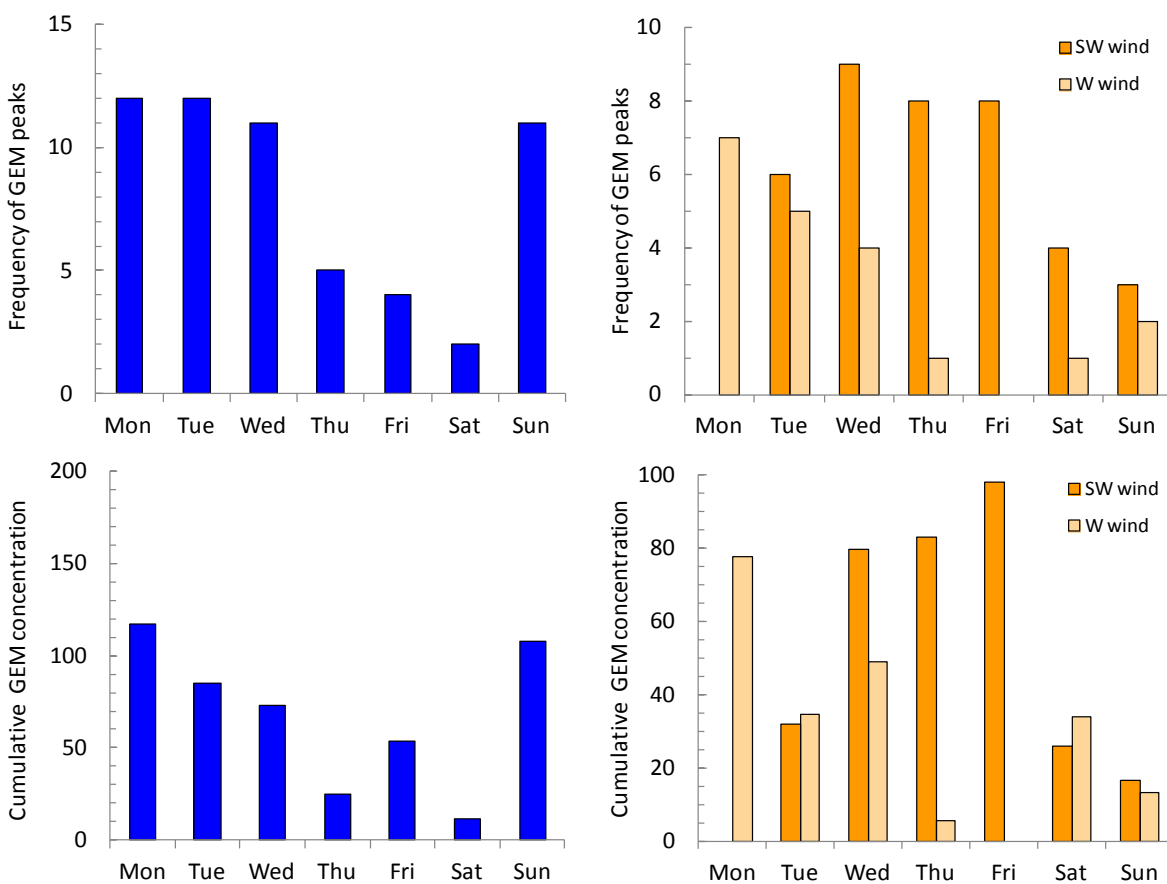


Figure 11. Frequencies of GEM peaks ($>4 \text{ ng m}^{-3}$) and distributions of cumulative GEM concentrations (ng m^{-3}) among days of the week. Values are for the cluster of GEM peaks measured in Elizabeth, New Jersey during south winds (A, C) and for clusters of GEM peaks measured in Staten Island, New York during southwest winds (dark orange) and during west winds (light orange).

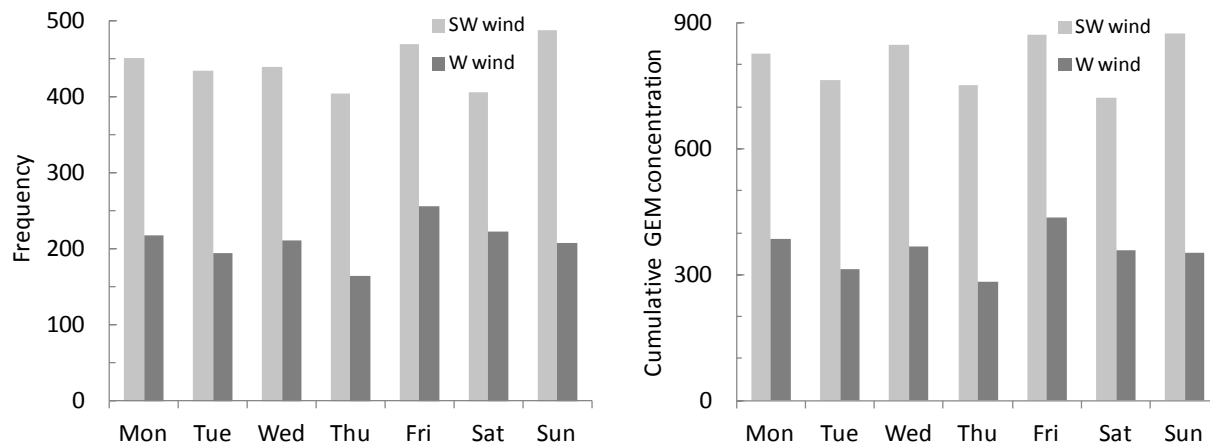


Figure 12. Distributions of background GEM (1 to 4 ng m⁻³) frequencies and cumulative GEM concentrations among days of the week in Staten Island, New York for measurements associated with winds from the southwest (light grey) and the west (dark grey).

The intersections of the two Staten Island GEM wind trajectories with the ELAM trajectory delineate two possible GEM source areas (Fig. 13). The first is centered in Rossville, NY on Staten Island (40.5514 N, 74.1947 W) and the second near Pralls Island in the Arthur Kill along the eastern border of Linden, NJ (40.6119 N, 74.2039 W). Potential specific Hg emissions sources within these areas should be identified and investigated further.

Local scale GEM measurements

Local scale measurements of GEM concentrations were deemed unreliable since values appeared to drift and did not match those measured at the CSI or ELAB sites in concentration or trend.

Acknowledgements

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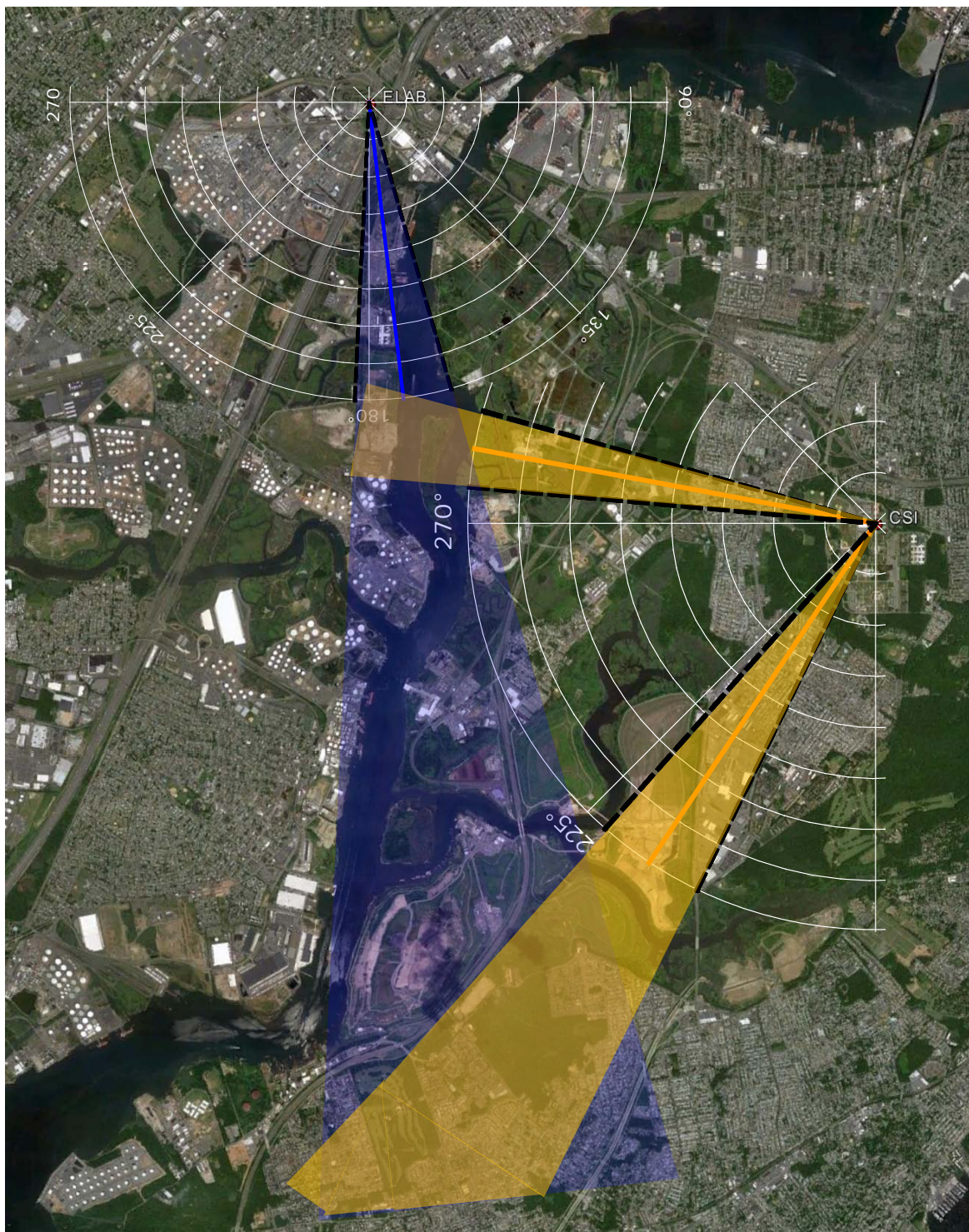


Figure 13. Directional bearings of possible GEM emission sources to monitoring sites in Elizabeth, New Jersey (blue cone) and at the College of Staten Island, New York (orange cone). Directional bearings were defined by the mean (solid lines) and 95% confidence intervals (dashed black lines) of the Gaussian model fits for each GEM peak cluster.

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