

Supplementary for

Long-Term Observations of Atmospheric Speciated Mercury at a Coastal Site in the Northern Gulf of Mexico during 2007–2018

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1. Time Series of Yearly Mercury Concentrations

The time series of yearly concentrations of gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate bound mercury (PBM) are shown in Figure S1.

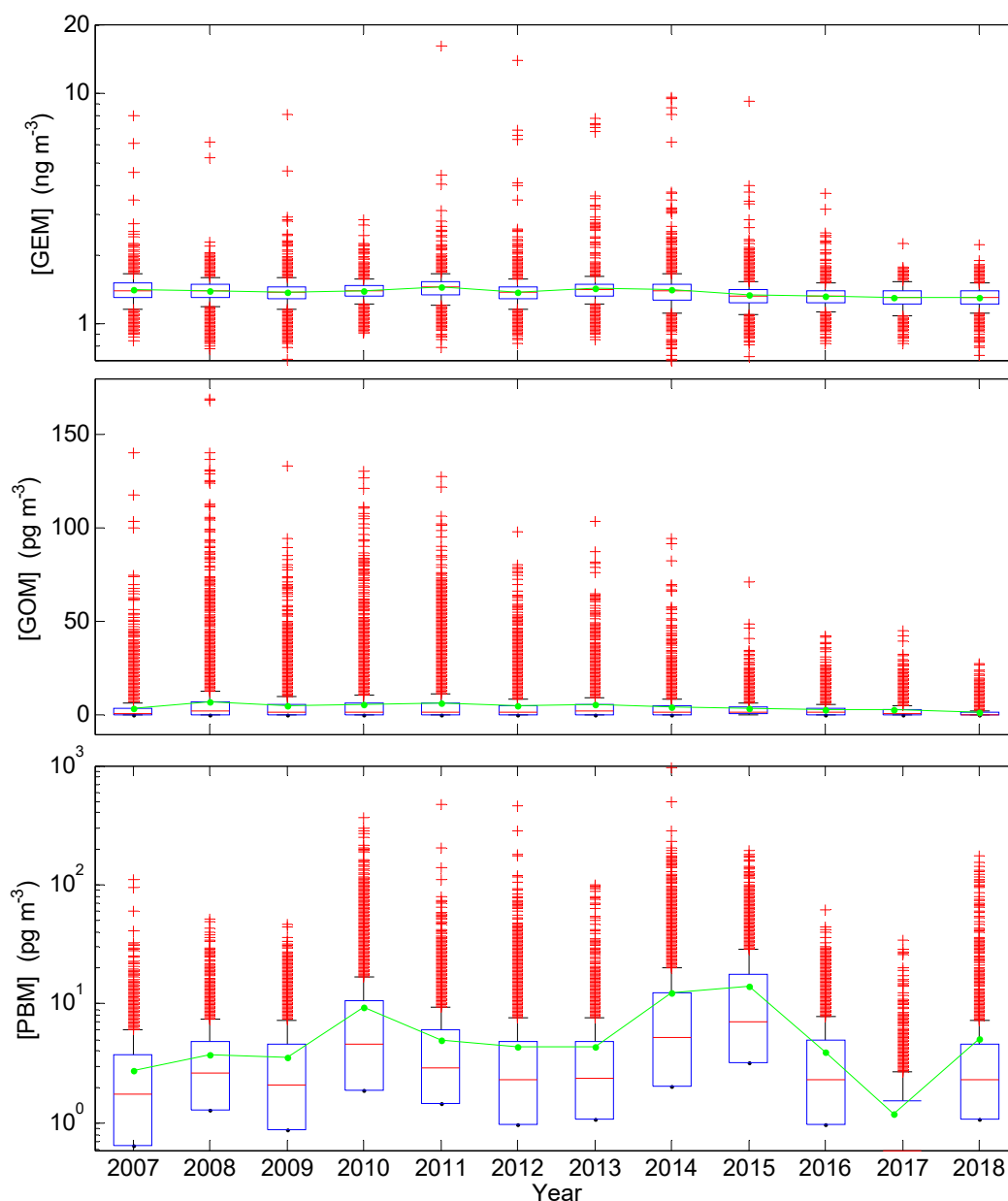


Figure S1. Boxplots showing yearly variations of GEM, GOM, and PBM measured at the Grand Bay site from 2007 to 2018. On each box, the central mark is the median, the edges of the box are the 25th and 75th percentiles, the whiskers extend to the 5th and 95th percentiles, and outliers are plotted individually as crosses. The linked green circles are yearly means.

2. Cumulative Mercury Concentration as a Function of Concentration rank

One method to show how important episodic peaks are to the overall average concentrations of GEM, GOM, and PBM at the site is to use the cumulative concentration as a function of concentration rank:

- Sort all mercury values (GEM, GOM, and PBM, respectively) from high to low.
- Assign a rank for each mercury concentration (GEM, GOM, and PBM, respectively), from the highest value (rank = 1) to the lowest value (rank = N), where N is the total number of valid measurements.
- Assign the cumulative concentration total, after each mercury concentration is added. If, for example, the highest GOM concentration is 250 pg m^{-3} , then the entry in the cumulative column for that highest value is just 250 pg m^{-3} . If the 2nd highest concentrations is 100 pg m^{-3} , then the entry in the cumulative column for that 2nd highest values is $350 (= 250 + 100)$. And so on. The

entry in the cumulative column for the last row (lowest concentration value) should be equal to the sum of all of the concentrations.

- Normalize the ranks by dividing each rank by N (the total number of measurements) so that the normalized ranks have values between just above 0 and 1.
- Normalized the cumulative concentrations by dividing each cumulative concentration by the total cumulative concentration so that the normalized cumulative concentrations should have values from a small fraction up to 1.
- Plot the normalized cumulative concentration against the normalized rank.

The normalized cumulative concentration as a function of the normalized rank for GEM, GOM, and PBM for each year from 2007 – 2018 is shown in Figures S2-S4

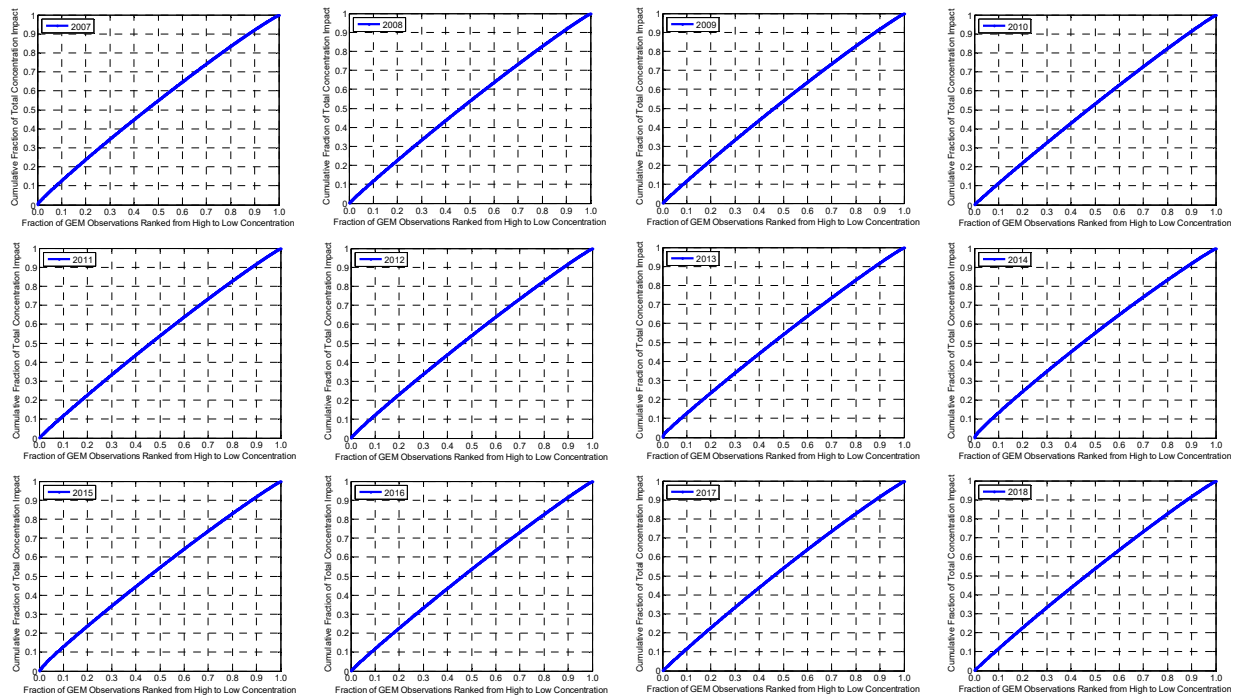


Figure S2. The normalized cumulative concentration as a function of the normalized rank for GEM for each year from 2007 - 2018.

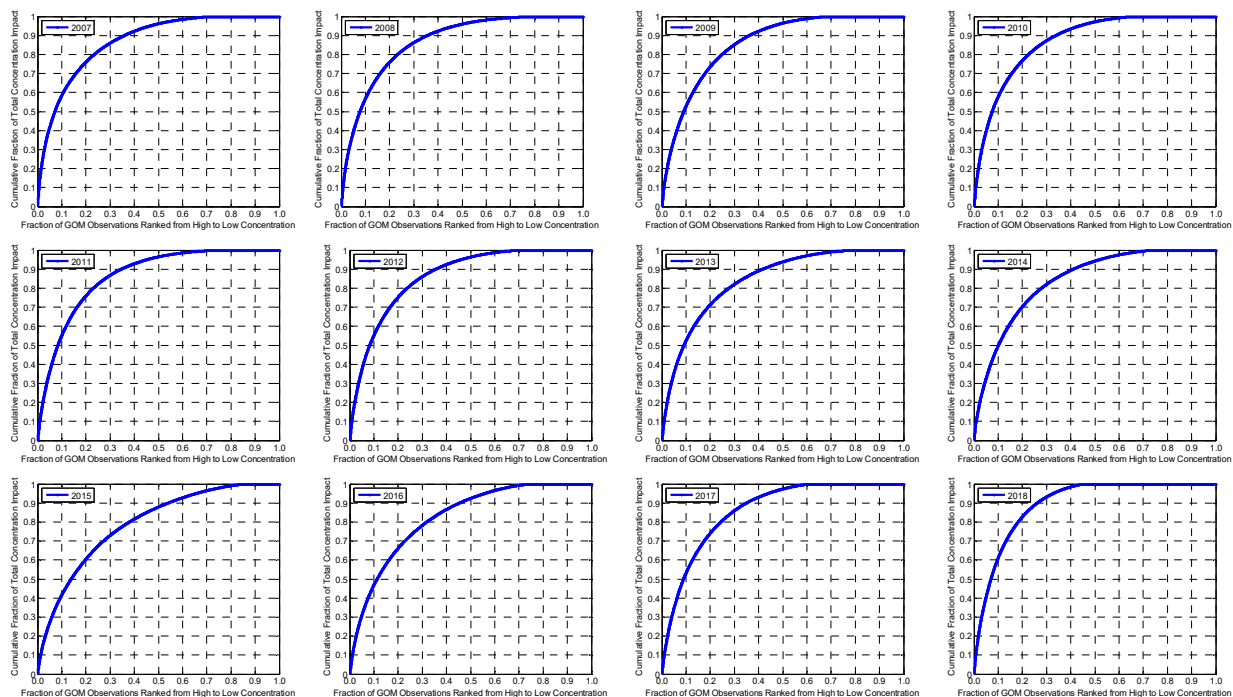


Figure S3. The normalized cumulative concentration as a function of the normalized rank for GOM for each year from 2007 - 2018.

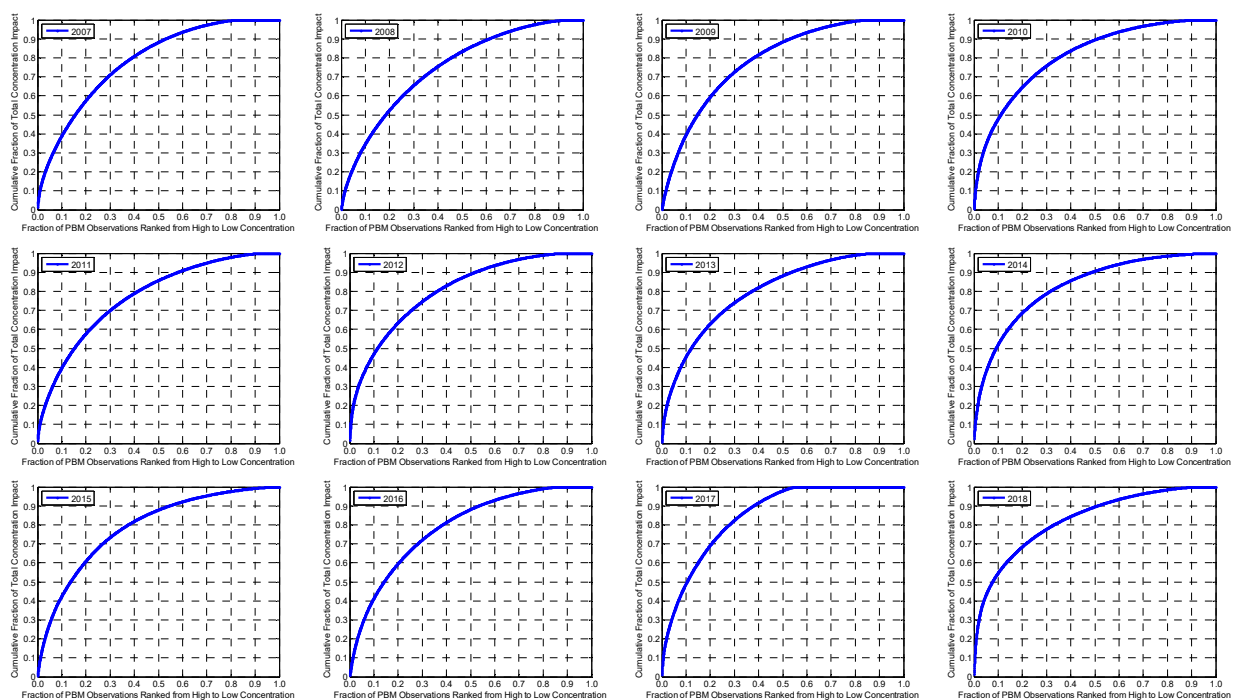


Figure S4. The normalized cumulative concentration as a function of the normalized rank for PBM for each year from 2007 - 2018.

3. Diurnal of Total Reactive Mercury

The diurnal profile of total reactive mercury (TRM = GOM + PBM) show that it peaks during the day with a mean maximum of 21 pg m^{-3} . Nighttime [TRM] remained constant with a mean concentration of $\sim 6 \text{ pg m}^{-3}$ (Figure S5).

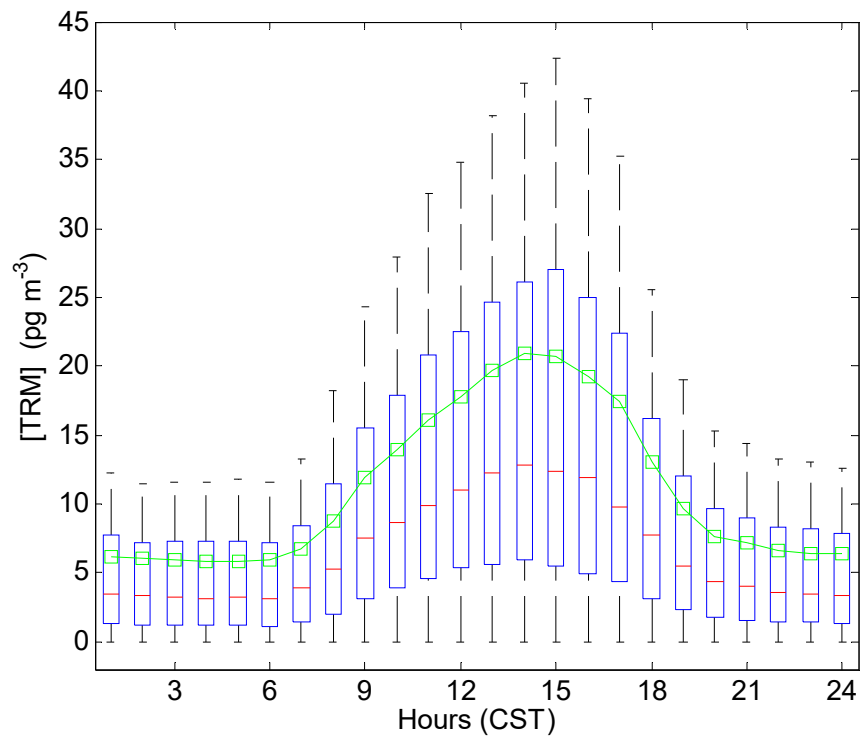


Figure S5. Boxplots showing diurnal variations of total reactive mercury (TRM) measured at the Grand Bay site from 2007 to 2018. On each box, the central mark is the median, the edges of the box are the 25th and 75th percentiles, and the whiskers extend to the 5th and 95th percentiles. The linked green circles are hourly means. Times are Central Standard Time (CST).

4. Correlation between PBM and GOM in Four Seasons

Figure S6 shows the correlation between PBM and GOM in four seasons: spring (March-May), summer (June-August), fall (September-November), and winter (December-February). Very poor correlation between PBM and GOM for all four seasons was observed with all correlation coefficients (r^2) being less than 0.1.

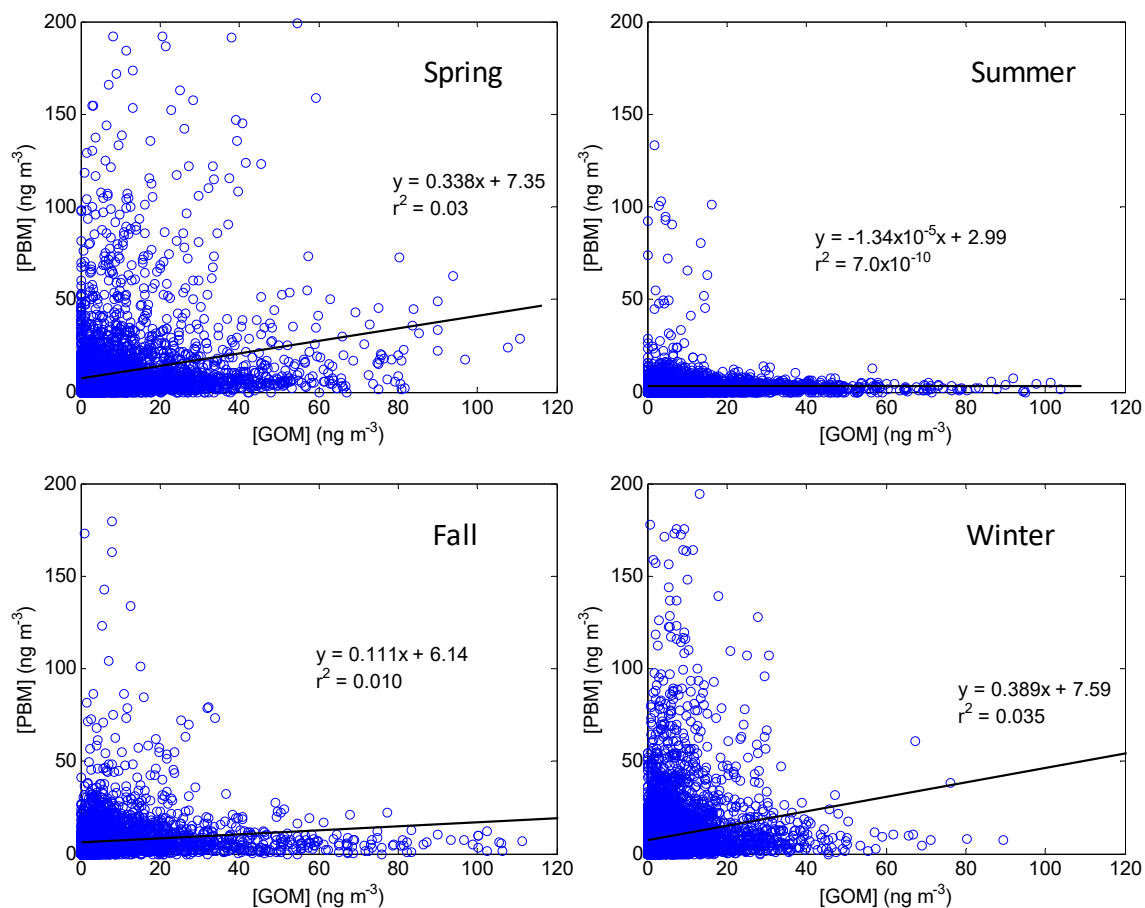


Figure S6. Correlation between PBM and GOM in four seasons. Individual circles are hourly mean measurements at the Grand Bay site from 2007 to 2018 and solid lines are the linear regressions.