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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characteristics of total gaseous mercury (TGM) concentrations in an industrial complex in southern Korea: Impacts from local sources" *by* Yong-Seok Seo et al.

Anonymous Referee #1

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General comments

This study analyzed seasonal and diurnal variations of TGM at a sampling site in southern Korea. Sources of TGM affecting the sampling site were investigated by correlating TGM with other pollutants and meteorological data and applying several source-receptor methods utilizing wind direction and back trajectory data. A newer method called the conditional bivariate probability function (CBPF) was used in this study to identify sources of TGM. However, I did not find this method very effective at differentiating between ground and stack emission sources. I find that there are many uncertainties in the CBPF results as well as in the back trajectory results that haven't been addressed in this paper. I have concerns about the methodology (insufficient

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Discussion paper

TGM data, selection of trajectory duration) and interpretation of correlation analysis results. A discussion of how the results vary in the different seasons was also lacking in many places of the paper, even though the results are shown in the figures. Overall, I find that a major revision of this paper is necessary.

Specific comments

Line 57 – the correlation coefficient of r = -0.08 is very small. It's more accurate to state there is little correlation between TGM and air temperature

Line 84 – "Atmospheric Hg released from natural and anthropogenic sources leading to enhanced deposition" Please clarify this statement.

Lines 87-89 – Is this sentence about Hg emissions to the atmosphere and the biogeochemical cycling of Hg or the direct release of Hg from industrial effluent?

Lines 94 – "coal combustion and waste incinerators" was already mentioned in this sentence.

Line 108 – use "data" instead of "information"

Line 138 – What temperature was the heated sampling line maintained at? Why is a heated sampling line necessary for sampling TGM?

2.3 QA/QC – the measurements were made for a one week period in each season. How often were the manual injections performed? Was there any maintenance activities performed prior to re-deployment of the instrument each time? These are important QA/QC procedures to mention because the instruments were offline for a long period of time.

Line 168 – This definition of CPF doesn't seem right because it is not exactly the source contribution. You can replace this sentence with the one in line 177.

Lines 173-175 – were the wind data measured every 5 min similar to TGM or was it averaged to the nearest hour?

Lines 183-185 –How does this method account for the full distribution of concentrations rather than concentrations exceeding a threshold? Based on Eq. 2, CBPF analyzes the subset of concentrations above a threshold as well. Another thing is how does this method account for sources with different dispersion characteristics? The equation is based on horizontal wind speeds, which is advection rather than dispersion.

Lines 193-195 – This explanation is not clear. Can you give some examples of mercury sources with different wind speed dependencies?

Lines 201-215 – this part needs to be rewritten by improving on the wording

Line 220 –Please justify the use of 24 hr trajectories. TGM is mainly GEM which has a longer residence time and capable of long range transport. This means a longer trajectory duration would be more suitable.

Line 229 – should be “meteorological data”

Lines 293-294 – This point was not discussed in later sections

Lines 299-302 – It should be stated more clearly that combustion was not a source of TGM because of a lack of correlation between TGM and the other combustion pollutant markers.

Lines 315 – Fig. S4 shows the CPF and CBPF plots in each season. Should this be discussed in section 5.5? It’s not clear how these plots relate to the correlation results.

Lines 326-333 – It’s surprising that long-range transport from China did not impact this site considering that it affected elevated Hg events in Seoul, Japan, and North America in previous studies. I would think long-range transport impacts a larger region including this sampling location. The TGM/CO slopes during elevated Hg events in Seoul were attributed to both long-range transport and local source impacts (Choi et al., 2009). Is it possible that the one-week sampling period in each season did not capture the long-range transport events? More data is needed to confirm these results. There are also uncertainties from the potential mixing between long-range transported airflows

and local air making it difficult to distinguish between distant and local source impacts.

Lines 345-346 – what are the time periods for daytime and nighttime concentrations? This sentence states daytime TGM were higher during daytime than nighttime. But in the previous sentence, the minimum TGM concentration occurs in the afternoon.

Lines 353-355 – You discussed about the land-sea/lake breeze effect on TGM diel patterns from another study in the previous sentence. Does this atmospheric process affect this particular site since it is near the ocean and lower TGM were also observed during daytime?

Lines 355-356 – The negative correlation between TGM and temperature is very small (as mentioned in the abstract, r = -0.08) despite a significant p-value. It's more accurate to state there is little relationship between TGM and air temperature.

Lines 357-360 – Similar to the above comment, the correlation between TGM and O3 is too small (r = -0.18) to suggest that it is indicative of GEM oxidation. It's more correct to state there is little relationship between TGM and O3. If GEM oxidation occurred, GOM concentrations would increase. There are some uncertainties on the net effect on TGM (GEM+GOM).

Lines 369-378 – The results here are inconsistent. If the small negative correlation between TGM and temperature indicates an increase in mixing height which leads to a decrease in TGM, how can it explain surface emissions in the morning which should increase with temperature? Is there a positive correlation between TGM and temperature in the morning?

Section 5.4 – This section is lacking discussion on seasonal differences in the diurnal variation. The fall diurnal pattern appears very different from those in other seasons in Fig. 3. Can you discuss why the TGM were much higher overnight in the fall but daytime concentrations were similar to those in other seasons? Why was there a large drop in TGM from 4:00 to 5:00 in the fall?

Lines 381-383 – These conclusions are not well-supported by the correlation analyses because the correlation coefficients were very small.

Line 390 – What are the potential Hg sources from the northeast direction?

Lines 391-393 – Please clarify this sentence. Are the higher wind speeds associated with stack emissions and lower wind speeds associated with surface emissions? There are several issues or uncertainties with this point. (1) Wind speeds were only measured at one height. How can you tell that the lower wind speeds are from lower elevation and vice-versa? What is the height of the wind measurements? (2) As mentioned in the diurnal variation section, boundary layer mixing occurs during the day. Is it possible to distinguish between ground emissions and stack emissions? (3) It seems only the west directions had both high and low wind speeds, while the east directions had only lower wind speeds (Fig. S1). Thus, CBPF doesn't seem useful when there is a lack of wind speed variation. It appears that it is by coincidence that both ground level and stack emissions were identified in the west direction because there happened to be a wind speed variation from this direction. Based on these points, the CBPF results don't seem to reveal more about TGM sources than CPF. More discussion is needed on the relationship between specific sources and wind speeds. Instead of wind speed, what other variables would be useful for source identification using CBPF?

Lines 394-395 – Are there industrial sources south of the sampling site? High probability areas are also identified in this direction in the TPSCF plot in Fig. 4.

Fig. 4 – the source areas seem confined to the industrial complex near the sampling site because of the short trajectory duration (24 hrs). Use of longer trajectories would help expand the source region and identify potential regional transport to the site. In addition to this uncertainty, other PSCF uncertainties should be discussed.

Section 5.5 – This section is lacking discussion on the CPF and CBPF results in different seasons. The seasonal plots are shown in Fig. S4, but they are not discussed in this section.

Lines 421-423 – This conclusion was not discussed in the results section. The wind direction frequency plots in different seasons are shown in Fig. S1 but the results were not discussed in the paper.

Lines 427-430 – p-value was significant but the correlation coefficient (magnitude of the relationship) is too small. I also don't understand the logic in these results. There should be a positive TGM and temperature correlation if daytime TGM concentrations were related to surface emissions.

Line 434 – JP-PSCF was not used anywhere else in the paper. Did you mean TPSCF?

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