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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impacts of Large-Scale Circulation on Urban Ambient Concentrations of Gaseous Elemental Mercury in New York, USA" *by* Huiting Mao et al.

Anonymous Referee #2

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This paper analyses seven years of GEM measurements in New York city. The authors observed large inter annual and seasonal variations in GEM concentrations and investigate the impact of mesoscale atmospheric transport patterns on Hg concentrations in NYC. The presented manuscript includes many interesting observations and conclusions but fails to present them in a consistent and concise way. I think that this paper would merit publication in ACP but only after significant improvement and a thorough correction.

P1 L45: 1 ng/m^3 = 112 ppqv (please add that ng/m^3 refer to standard conditions at 0°C and 1014hPa). I understand that it is a common convention to use mixing ratios

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for air pollutants in the US. Nonetheless, I urge the authors to use ng/m^3 at standard conditions throughout the paper. This has been a common convention for Hg related studies even in US journals.

P4 L75pp: You should also mention that primary emissions due to more coal combustion is larger during winter time.

P6 L123: Please briefly describe the mentioned emission scenarios.

As you do not consider atmospheric chemistry, did you emit 100% of the Hg as GEM?

P7 L149-150: You state that GEM mixing rations were often larger during the warm seasons. Often is a very vague expression and I think that Figure 2 does not fully support this claim. Please give a quantitative measure e.g. the ratio of summer winter average mixing ratios for each year. Just looking at figure 2 I would conclude that this is true for 2014 and 2009 only.

P9 L181: Figure 3 is difficult to read as the different percentile values are not clearly distinguishable in the plot.

P9 L 190pp: You assume that the Hg emitted in the Eastern US is perfectly mixed within the PBL and ignore transport, chemistry, deposition as well as the temporal variability of emissions by simply dividing by 365 to get an daily change. I can fully understand that you start by estimating the maximal possible impact of emission changes on regional Hg mixing ratios but your calculation seems implausible. Your estimate for the Eastern US for 2008-2011 is -200 ppqv Hg (by the way please add signs to indicate increase or decrease before the numbers). This value is higher than the northern hemispheric average Hg mixing ratio of 168 ppqv (1.5 ng/m^3). This obviously makes no sense. For NYC you estimate even larger values with increases in the range of 6 ppqv per day.

In 2014 you observed mixing ratios 90 ppqv higher than in the previous year and state that: '90 ppqv throughout the seasonal averaged diurnal cycle, a factor of 15 larger than the effect of local emissions'. I can only guess that this is based on the calculation

90 ppav / 6 ppqv d-1 = 15. Which leads me to assume that you made a mistake with the units here. If you give the emissions in P9 L190 as kg/a and assume an average residence time for the emissions inside your domain of 1 day the resulting change in Hg mixing ratio would be 6ppqv and not 6ppqv d-1. This would at least lead to plausible values.

However, I still object this oversimplified approach. Given the fact that already in P9 L197 you state that this estimate is inconsistent with observations rises the question why you include this flawed approach in the first place. Why don't you use the results from the HYSPLIT simulation here?

P11 L222pp: Please elaborate whether the local wind directions are representative of the regional transport patterns. As your measurement station is located inside a major city local wind direction are not necessarily consistent. Please add more information ion the measurement site (e.g. height of measurement point compared to surrounding buildings) in P5 Section 2.

P11 L229pp: It would be highly interesting if you could calculate wind direction adjusted inter-annual changes in Hg mixing ratios.

P12 Figure 5: It would be of great value if you could add SO2 observations to this figure as they are an indicator of regional Hg sources (mainly coal combustion in the US).

P14 L301p: Please check grammar.

P17 L371pp: It is very interesting that GEM trends in all four wind quadrants were similar. Was this a local effect in NYC or did other Hg stations in the Eastern US see similar increases? Moreover, did you see a correlated increase in other pollutants (reference to the following section)?

P17 L380: Please clarify what emission increases could explain the much higher Hg mixing ratios in summer 2014. Otherwise, this sentence seems to be just a filler.

P18: Please do not swap between r and r^2.

Hg sources in the US should be much more linked to SO2 than CO. Thus, I would suggest to include the analysis for Hg/SO2 in Figure 9. Your finding that SO2 is only weakly correlated to Hg at the Bronx site is very interesting and needs some additional clarification. E.g. what sources were driving the SO2 emission reduction in the area? The Hg/SO2 ratio from local sources is also strongly influenced by the type of coal used for combustion. Maybe if you use the Hg/SO2 emission ratios for each you could normalize the observed ratio?

P18 L394pp: Please note that CO/Hg ratio is mostly indicative of Hg from biomass burning. In urban areas vehicles are a major CO source and will dominate the CO signal. Thus, I would argue that the Hg/CO ratio is mostly a sign of wind speed and buildup of regional air pollution in NYC and obviously will behave differently than at an urban stations.

Section 5: It is commendable that you include a modeling exercise into the paper. However, it seems just attached as a supplement in the end. You need to better describe the model setup and also openly discuss the weaknesses and shortcomings (e.g. long range transport, chemistry). And then include the model results already in the discussion of the other sections.

P19 Section 5: Please describe in more detail your model setup. In the introduction you mention 2 emission scenarios which are not further described anywhere. Moreover, it is important to know what you used as boundary conditions to estimate the background concentrations of Hg.

You estimate that 75% of the anthropogenically induces GEM in NYC originates from local sources (and with a larger domain this could even be enhanced). It seems that you underestimate the long range transport of Hg as your results do not seem consistent with findings from similar mode lstudies (e.g. Cohen et al., 2016. Modeling the atmospheric transport and deposition of mercury to the Great Lakes. Environmental Research)

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P22 L 486: "... regional contributions to NYC ambient concentrations would be even more dominant." You need to clarify that this assumption neglects all long range transport of Hg.

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