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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Recent decrease trend of atmospheric mercury concentrations in East China: the influence of anthropogenic emissions" *by* Yi Tang et al.

Anonymous Referee #2

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General comments

This paper presents a multi-year record of GEM concentrations at Chongming Island, East China and reports a decreasing trend with a rate of -0.52 ng/m3/yr. The authors attribute this decreasing trend to air pollution control policies targeting SO2, NOx, and particulate matter. This paper could make a valuable addition to the literature. However, while I agree with the conclusions (e.g., decreasing anthropogenic emissions, co-benefit from pollution control policies targeting other compounds), I am not really convinced by the level of scientific evidence presented here. The paper could be suitable for publication in ACP after the authors address the following issues.

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Discussion paper

Major comments

I really think you should perform a trend decomposition of the signal (signal = seasonal + trend + random, example here: https://anomaly.io/seasonal-trend-decomposition-in-r/). There is a very strong seasonal cycle and you conclude that "the seasonal GEM cycle was dominated by the natural emissions". However, how can you explain that the seasonality is way more pronounced in 2014? To me, presenting emissions inventories is not convincing enough; how you can you be sure that the decreasing trend is not driven by a change in seasonality? While SO2, NO2, and PM concentrations were monitored, data are not presented nor discussed. Do you also observe a decreasing trend? That would be the best way to support that "air pollution control policies targeting SO2, NO2, and PM reductions had significant co-benefits on atmospheric Hg". Finally, I wonder why GOM and PBM data are not reported and discussed. Do you also observe a decreasing trend? You may have encountered issues with the speciation unit. If so, was the experimental setup identical in 2014 and 2016, or did you analyze GEM when the speciation unit was working vs. TGM when it wasn't? A discussion on analytical uncertainties would be much welcomed.

Line by line comments

Line 26: "GEM concentrations showed a significant decrease with a rate of -0.60 ng/m3/yr". According to Table 1, the rate is -0.52 ng/m3/yr.

Line 33: "It was find" should be "It was found".

Lines 47-48: "In the atmosphere, Hg mainly presents as GEM, accounting for over 95% or the total". Can you please add a reference? Is that also true at your site?

Line 61-62: "(. . .) there is no official national monitoring network of atmospheric Hg". Out of curiosity, what is the current status of the Asian-Pacific Mercury Monitoring Network (http://nadp.sws.uiuc.edu/newIssues/asia/)? Do you think that Chinese sites will be included?

Lines 64-67: "Atmospheric Hg emissions in China accounted for 27% of the global total in 2010 (UNEP, 2013), which led to high air Hg concentrations in China. Therefore, atmospheric Hg observations in China are critical to understand the Hg cycling at both regional and global scale". Please define "high" air Hg concentrations. Additionally, in order to emphasize the fact that observations in China are critical to understand the Hg cycling on a global scale, you could perhaps add a sentence about 1) future projections (e.g., Chen et al., 2018; Pacyna et al., 2016), and 2) long-range transport of Chinese emissions to other regions (e.g., Chen et al., 2018; Corbitt et al., 2011; Sung et al., 2018).

Lines 93-94: "we used Tekran 2537X/1130/1135 instruments to monitor speciated Hg in the atmosphere". I wonder why GOM and PBM concentrations are not reported in the manuscript. If concentrations were recorded, it would be interesting to discuss the results. Do you also see a decreasing trend from 2014 to 2016? From 1978 to 2014, the fractions of GEM and PBM decreased, while the GOM emission share gradually increased (Wu et al., 2016). What about the speciation of emissions since 2014? Can you observe a trend in GOM/PBM concentrations? Alternately, did you have issues with the speciation unit? It is quite common and I would appreciate an open discussion about that and associated analytical uncertainties. What kind of issues did you encounter? Are you confident that you collected and analyzed GEM (vs. TGM) during the entire experiment? Was the instrumental setup exactly the same during the entire experiment? If not, how can you compare GEM concentrations without discussing analytical uncertainties? See major comment.

Lines 103-104: "The impactor plates and quartz filter were changed in every two weeks. The quartz filter was changed once a month". Did you change the quartz filter every two weeks or once a month?

Line 106: "During the sampling campaigns, PM2.5, O3, NOx, CO and SO2 were monitored". Why aren't you discussing the data, especially SO2, NOx, PM2.5 while your main conclusion is that Hg decreasing trend in due to air pollution control policies tar-

geting SO2, NOx, and PM. I agree that you present emissions inventories, but I would really appreciate to see a real interpretation and discussion of these data. Do you also observe a decreasing trend? See major comment.

Lines 173-175: "Besides, this method required similar meteorological conditions of the periods participated in comparison so as to reduce the interference from meteorology". I am not sure I understand this sentence. Do you mean that you used similar meteorological data in 2014 and 2016 to compute the back-trajectories? Or are you referring to the fact that meteorological conditions were pretty much similar in 2014 and 2016 (lines 266-274)?

Lines 188: "For small emission sectors (. . .)". Which ones?

Lines 193-194: "The average concentrations of GEM in 2014 and 2016 were (. . .)". What about the mean concentration in 2015? Additionally, are the average annual concentrations actually referring to March-December? If so, please add something like "The average concentrations of GEM in 2014 (Mar-Dec) and 2016 (Mar-Dec) were (. . .)".

Lines 194-195: How does it compare to concentrations reported in Sprovieri et al. (2016)?

Lines 199-200: "During this period, monthly GEM concentrations showed a significant decrease with a rate of -0.60 ng/m3/yr". Table 1 refers to TGM concentrations, not GEM. Additionally, as mentioned earlier, the rate is -0.52 ng/m3/yr in Table 1. Please, try to be consistent throughout the manuscript.

Lines 201-216: To me, "GEM" and "TGM" are not interchangeable (see previous comment). While the difference between TGM and GEM is usually smaller than 1% (Soerensen et al., 2010), it might not be the case everywhere. What is the fraction of GOM at your site? I would appreciate a discussion on analytical uncertainties and instrumental setups. The sentence "at the Cape Point of South Africa, GEM concentrations

decreased from 1.35 ng/m3 in 1996 to 0.9 ng/m3 in 2008" is not entirely true. A downward trend has been observed from 1996 to 2005, while an upward one is observed since 2007 (Martin et al., 2017; Slemr et al., 2015). Additionally, the instrumental setup changed: a manual amalgamation technique was used from 1995 to 2004 while a Tekran instrument has been used since 2007 (Martin et al., 2017). It might also be the case at other stations in Table 1. How does it influence the various trends reported in Table 1?

Lines 212-214: "The decreasing trend observed in our study was accordant with the unpublished data in Mt. Changbai during 2014-2015 cited in the review of Fu et al. (2015). But much sharper decrease of Hg concentrations was observed in our study". Aren't the data at Mt. Changbai you are referring to in Sprovieri et al. (2016)? What is the trend at that site? Why isn't included in Table 1?

Line 224: Are you referring to Figure 2?

Lines 225-227: Is that based on the $\sim$3 years of data?

Section 3.2: I find this section quite confusing and difficult to follow.

Line 234: "The higher Hg concentrations in cold seasons in Mt. Ailao and Mt. Waliguan (. . .)". You say above that concentrations are lower in the cold season at these sites. This is confusing.

Line 250-251: "Therefore, we supposed that the seasonal cycle of GEM concentrations was dominated by natural emissions". How can you explain that the seasonal cycle is more pronounced in 2014 than in 2016? See major comment.

Lines 275-276: "This decline may be contributed by the downward trend of GEM concentrations in north hemisphere". Please, elaborate on this idea. I don't really understand what you mean here.

Section 3.4: See major comment. Please perform a trend decomposition of the signal. I don't know which software you use, but here is an example using R:

https://anomaly.io/seasonal-trend-decomposition-in-r/.

Lines 315-325: Do you get the same results if you perform this analysis on SO2, NOx, and PM concentrations?

Line 318: 34% should be 35% according to Table 4. Additionally, how can you explain this result? Is there a decline in anthropogenic emissions and a GEM decreasing trend in this region (China Sea, Japan, South Korea) as well? Cluster EAST explains 35% of the decline, i.e., 0.35 x 0.52 = 0.182 ng/m3/yr. Is that consistent with trends reported in this region (e.g., Kim et al., 2016)?

Lines 321-323: "We also noted that the largest decline of Hg concentrations was observed in the cluster SW, which indicated more effective air pollution control in the regions where the air mass of the cluster SW passed". What about the seasonality of GEM concentrations in the various clusters (NW, SW, EAST)? Could a difference in seasonality explain the observed Hg decline?

Figure 3: Could you please add the standard deviations? Is that the average over several years?

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