

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 #1

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The paper by Tang et al. presents 33 months long measurements of GEM, SO₂, NO_x, O₃, CO, and PM_{2.5} at an island not far from Shanghai. The authors find a pronounced GEM downward trend and analyse it in terms of regional sources, clusters of backward trajectories and principle component analysis. Seasonal variation is also discussed. The authors arrive at the conclusion that the downward trend is due to substantial reduction of the regional mercury sources. This is an important finding which deserves to be published. I think, however, that several substantial changes need to be made to the manuscript before it can be accepted for publication.

The suggestions are:

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Experimental: The authors mention that mercury species were measured but only GEM data are presented and their trend calculated. What was the contribution of GOM and PBM? Could they contribute to the trend? This is important to discuss because the regional atmospheric Hg emissions in Section 2.5 are probably not only those of GEM but of total mercury. What was the seasonal variation of GOM and PBM? Could it provide some additional evidence for the reasons of GEM seasonal variation?

Section 3.1: Averages and their standard deviations should always be stated with the number of measurements because only then statistical tests for significance of differences can be made. In line 194 the authors claim that GEM concentrations in 2014 were significantly higher than... - at which significance level? Line 199: the annual decrease rate should be given with its uncertainty and number of months.

Figure 3: It is not clear how the points in Figure 3 were calculated? Were the data detrended before the averaging? In view of the strong downward trend they should be. What is the standard deviation or standard error of the monthly means – please show them as vertical bars in Fig. 3. Are the differences between the months statistically significant? This is a precondition for the discussion of the seasonal variation.

Figure 4 and its capture: This figure needs substantial revision to illustrate the point the authors make and to make it understandable for the readers. Negative emissions are deposition fluxes and should be named as such. Thus “natural emissions” in spring, autumn and winter are in fact deposition fluxes. Net fluxes are needed to illustrate the point made by the authors but they are not shown. The capture should also state that it is about the emissions and depositions in the YRD region? This will rise a problem: trajectory analysis in section 3.4 shows large influence of transport from the NW provinces of China outside of the YRD region. How does this transport influence the seasonal variation?

Line 243-245: “The annual emissions from both natural source and anthropogenic source ... was 0.75 and 10.3 t, respectively” – the reader may think that natural emis-

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sions make less than 10% of the anthropogenic ones and cannot thus be responsible for the seasonal variation. One has to look in Fig. 4 to find out that the “natural emissions” are a sum of natural emissions in one season and “natural” deposition fluxes in three seasons. That provokes a question: how is anthropogenically emitted mercury removed from the atmosphere if there are only “natural” deposition fluxes? Please use the correct terminology and separate the natural and anthropogenic emissions from the deposition fluxes of both.

The results of PCA analysis and Table 2: The authors attribute the factor 2 to “exchange of PBL with free troposphere” but do not explain why. Last row in the table 2 called “variance explain” lists for 2016 exchange of PBL with the free troposphere 75.735 which together with 50.625 for “combustion” makes more than 100. As such the units of “variance explain” cannot be percent. What are the numbers in this row and does it make sense to present them with three valid numbers after decimal point?

Table 3: The numbers are probably annual emissions but the capture does not say it. The year of the emissions is not given. I wonder about the “other SO₂ sources” which are substantially larger than all coal, oil, and biomass burning taken together. If it is not an error, what are the “other SO₂ sources”?

Chapter 3.4 misses a major point: Table 3 of SO₂, NO_x, PM_{2.5}, and GEM emissions is only for one undefined year and only for the YRD region. Table 4 and Figure 6 show a dominant influence of transport from NW of China which is mostly outside of the YRD region. To illustrate convincingly the major conclusion of the paper one would need a table with the inventories for NW and SW (perhaps separately) and for 2014 and 2016.

Table 1: The paper is about regional trend and I wonder why it is necessary to discuss global background trends in such detail. Also because the reasons of downward trend of mercury at many background stations of the world at the time of increasing global emissions is still not well understood (compare Horowitz et al., EST 48, 10242-10250, 2014, with Soerensen et al., GRL, 39, L21810, doi:10.1029/2012GL053736, 2012).

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Figure S1: Here the least square fit of 4 points provides R^2 of 0.487 for January and 0.613 for February for which the authors claim $p < 0.01$ in both cases. This is surely incorrect because 4 points result statistically in only 2 degrees of freedom. Please explain.

Figure S2: The downward annual rate should be given with its standard error.

Editorial remarks:

Line 51: “Both GOM and PBM are more soluble..” than what? PBM is not necessarily more soluble than GEM but it is scavenged by wet deposition. Low solubility of GEM need to be mentioned before this statement.

Lines 71/72: ...have been estimated to decrease..

Line 86: ...is located...

Lines 102/103: ... the error between gold trap A and gold trap B was limited to...? Probably the difference instead of error was limited. What happens if the difference is more than the limit?

Lines 171/172: “uncertainties” would be better than “errors”

Line 199: Please state the decrease rate with its standard error.

Lines 207-209: A reference to Martin et al (2017) is not correct because the paper does not contain annual averages and the authors of this paper do not mention a gap in the measurements between 2004 and 2007. The correct reference would be: annual average GEM concentration decreased from 1.29 ng m⁻³ in 1996 to 1.19 ng m⁻³ in 2004 (Slemr et al., GRL 35, L11807, doi:10.1029/2008GL033741, 2008) and were increasing from 0.93 ng m⁻³ in 2007 (Slemr et al., ACP 15, 3125-3133, 2015) until 2016 (Martin et al, 2017).

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