

## ***Interactive comment on “Bimodal variation in mercury wet deposition to the coastal zone of the southern Baltic” by P. Siudek et al.***

### **Anonymous Referee #3**

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This paper addresses an area of research concerning the variation of mercury wet deposition in Poland. The authors present a wealth of long-term measurement, however the data interpretation is not logical and I think their data need to be discussed a lot further. The major concerns on this article are, first, that the QA/QC procedure is not included. The Hg concentration in rain was generally very high in this study, but yet I cannot find any paragraph describing QA/QC results including field blank, relative percent difference (using duplicate samples), and precision and recovery (using SRM). Since mercury is a trace element in environment the stringent QA/QC procedure is required. Readers may think that the high Hg(T) concentration in rain in this study was possibly caused by contamination if the paper does not include the detailed QA/QC procedure. Second, the authors used the term “labile Hg” which probably indicate ‘Hg(II) form in this article. However I cannot understand what this means, how

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this was measured, and even why this form was useful from this article. Third, the authors repeated that wet precipitation over the coastal zone of the southern Baltic purifies the atmosphere effectively. I think that the authors are confused about the different chemical-physical characteristics of various mercury species. Most of Hg in atmosphere is Hg<sub>0</sub> which is very insoluble, but the most of Hg deposited via precipitation is considered to be gaseous divalent Hg and particulate Hg, not Hg<sub>0</sub>. In ambient air Hg<sub>0</sub> is predominant (generally more than 90% of total Hg), therefore precipitation cannot “purify” the atmosphere with respect to Hg.

Specific comments: 1. The introduction is way too long and not focused on wet deposition. 2. Page 22780 Line 5: What are the “both forms of Hg”? 3. Page 22780 Line 10-14: The dissolved fraction was separated by filtration only during from spring to late autumn. How are the authors convinced that there was no contamination from filtration process? Contamination by filtration could cause the seasonal variability of Hg conc. in precipitation. Also did the authors use “dissolved Hg” concentration as Hg(II) in the result? In that case, THg in this article is sum of “dissolved Hg” and “particulate Hg”? What does Hg(II) mean in this article? Terms are very confusing. . . 4. Method section: QA/QC procedure should be described. 5. Page 22781 Line 20: How did you measure “the concentration of reactive mercury forms”? What does the reactive mercury form mean? 6. 22781-22782: The average THg concentration in rain is very high in this study, which is even higher than the measurement in China. Clarify the reason (such as the site description. . .). 7. 22783, Line 5-9: The authors explained that the elevated THg concentration (in precipitation) in summer was derived by the reemission of Hg from natural surfaces. However, re-emitted form is almost entirely Hg<sub>0</sub>, which is very insoluble. On the other hand, the Hg forms that are easily deposited are gaseous divalent Hg and particulate Hg. Therefore reemission is hardly the reason of an increase in THg in rain. 8. 22783, Line 20-26: I cannot understand the logic here. The authors said that it was found that a 1000-fold lower volume contains a 14-fold higher amount of mercury, which proves. . . . effectively. What does this sentence mean? Most of TGM in ambient air is Hg<sub>0</sub>, and most of THg in rain is Hg(II). Hg<sub>0</sub> should have very little contri-

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tribution to THg concentration in rain. 9. Figure 1: Much more details should be included, such as adjacent countries. . . 10. Figure 2: THg concentration is volume-weighted? It should be volume-weighted, because concentration in rain is generally dependent on precipitation depth. Both fluxes and concentrations need to show 95% confidence interval or one standard deviation. Also precipitation depth should be inserted because it is a major factor influencing deposition amount. 11. Figure 3: not useful. The authors explained enough the difference between heating season and non-heating season in the body of the text. And the figure itself does not show the important finding. 12. 22784, Line 20-25, and Figure 4: This figure (this result) does not necessarily indicate that pH in rain can control the forms of Hg. I suggest the authors to create this figure using THg as well, and see whether the result would be same or not. If same, pH is not a factor on determining the forms of Hg. I suspect that SO<sub>x</sub> and NO<sub>x</sub>, the major pollutant emitting from fossil-fuel combustion caused the low pH during heating seasons, and THg in rain increased as well. Therefore low pH and high Hg might happen to be observed together, but one does not affect to another. Also since the both pH and Hg concentration in rain are likely to be affected by precipitation depth, the effect of precipitation depth should be excluded in interpretation. Maybe one can use volume-weighted concentration and volume-weighted pH. 13. 22787 Line 14-17. What does the sentence “The lower they were flowing, the higher were the concentration of Hg in wet precipitation” mean? How did you identify the height at which the air masses were flowing? 14. 22787 Line 20: There is no Table 3 in this article. 15. Fig. 5: What does the legend (>40, <24, <4) mean?

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