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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of various methods to measure particulate bound mercury and associated artifacts" *by* S. Wang et al.

S. Wang et al.

youngji@kangwon.ac.kr

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Response to the general comment of Reviewer 1.

The main purpose of this study was to evaluate artifacts associated with various particulate bound mercury (PBM) measurement techniques. The measurement techniques used in this study have already been accepted to be appropriate methods to measure PBM concentration in many previous investigations even though artifacts associated with these measurements have not been evaluated. Although we agree it would have been better to have enough replicate samples to determine the overall precision of the methods used, we do not think that it is absolutely necessary in this study in which potential artifacts associated with sampling in remote locations are examined. Our objective was to determine if artifacts were important. Future work will better quan-

tify the absolute magnitude of the artifacts and explore potential corrective measures. In addition to collocated samples, this study included stringent QA/QC procedures including SRM recoveries, detection limits, field blanks and lab blanks. Although for thermo-desorption experiments, we analyzed only one filter for each day; we obtained statistically significant results.

The reviewer stated that the sample numbers of 18 and 8 are not enough for each sampling site. However, these two sampling locations do not have significantly different characteristics, as both locations are not considered to be significantly impacted by local sources. Therefore, it is reasonable to state that 26 samples were collected with different sampling durations. In order to verify the effect of sampling duration on artifacts, different sampling durations were used at CU (two days) and at HF (seven days) to provide a systematic comparison of sampling times. We found that the negative artifacts were significant from the comparison of the dataset between the MOUDI and TekSpec, between OFF and TekSpec, and between the MOUDI and OFF. Therefore, negative artifacts were further evaluated during shorter time durations of 4 hrs using exposure to zero air in lab experiments, and there was still a significant decrease in Hg mass. These findings are important and valuable to other researchers in this field. In summary the conclusions contained in this paper were derived from a comprehensive analysis of data obtained from field and lab investigations and indicates that a sampling duration even as short as four hrs may cause negative artifacts on PBM measurement (needless to say greater artifacts may be associated with two and seven day sampling times). We are convinced that 26 samples for each measurement device are enough to indicate the significance of negative artifacts in remote locations.

Response to the technical comments:

The figures have misspelled words. The line for equations need to be written as y=mx + b. - The manuscript will be revised as suggested.

Pg8587 line 17 particle size cut for Tekran needs to be introduced. - It will be revised.

Pg 8587 sentence that starts line 25 needs a reference. - The manuscript will be revised as suggested.

Pg 8588 there was some particle size fraction work done in the southeast that should be referenced by Engle et al. I think it is in Applied Geochemistry. - This reference will be added.

Pg 8592 –The information on temperature partitioning associated with particles is not new and there are papers by J. Schauer's group and one by Amos et al ACP that should be referenced. - We are aware of the papers published by Schauer's group and Amos et al.(2012), and already referenced two papers by Schauer's group in this article (Rutter and Schauer, Atmos. Environ., 2007; Rutter and Schauer, Environ. Sci. Technol., 2007). Amos et al. (2012) will be referenced in the revised article.

English throughout the paper should be checked i.e. Line 25 pg 8593 "is" should be "are." - English will be checked throughout the paper once again.

Line 8594- I am not really sure what they are talking about in the last line of section 3.2.2. Is the TekSpec Frit in the instrument with the elutriator in line? - Yes, the frit is the impaction plate of the elutriator before the denuder in the Tekran 1130 that removes particles > 2.5 μm from the sampled stream. This clarification will be added to the manuscript. One of the limitations of automatic Tekran Speciation System (TekSpec) is that PBM>2.5 μm is not measured. In this study, it was determined that the Tekran frit can be used to measure PBM>2.5 μm as there was no statistical difference in concentrations of PBM>2.5 μm between the MOUDI and TekSpec frit. We think that this finding is one of the most valuable outcomes in this paper, and will be extensively applied in future research.

The comparison with the Feng paper should consider the difference in loading and the matrix of loading. - Feng et al. (2004) indicated the appearance temperature for release of different Hg species from different matrices including rock, soil, fly ash, and airborne particulate matter. In this study, PBM speciation was identified based on the

study of Feng et al.(2004) for airborne particulate matter.

Figure 1 does not seem acceptable for ACP. - The quality of this figure will be improved.

Figure 2 define high and low. Is the shift observed statistically significant? - There was a statistically significant difference between high and low atmospheric temperatures for fine fraction of PBM ($\leq$1 μm) (p-value= 0.001, Mann-Whitney test) and coarse fraction of PBM (> 1 μm) (p-value= 0.037, Mann-Whitney test). Therefore, the shift observed is statistically significant, and we will add the results of Mann-Whitney test to the revised paper. In addition, the fine fraction of PBM (PBM in PM1/total PBM) significantly increased as the atmospheric temperature decreased (p-value= 0.01, spearman rank order correlation).

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/13/C1678/2013/acpd-13-C1678-2013-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 13, 8585, 2013.