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Interactive comment on "Atmospheric wet deposition of mercury and other trace elements in Pensacola, Florida" by W. M. Landing et al.

Anonymous Referee #3

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This is an interesting study evaluating the provenance of atmospheric Hg in Florida by determining Hg and numerous trace elements in a large set of rain samples collected during several years. I believe this manuscript is well within the scope of Atmospheric Chemistry and Physics. The authors make a good job to point out a prevalent non-local origin of Hg by using factor analysis and an original approach for inferring the Hg provenance. In particular this last method deserves to be presented and discussed within the scientific community. Overall, this work is also a good example of how local/regional policy makers dealing with Hg emission can be advised by atmospheric scientists. Nevertheless, there are several points that the authors should check.

1) The list of ancillary trace elements (TE) determined is impressive but it appears that not all TE provide the same significant information. This is illustrated for instance in Fig

C10560

3D by the scatter of Ba and by the fact that, although Ba is a typical crustal element, this is weakly linked to the crustal component. The authors point out that Ba concentrations are near the detection limit (LOD) (however, this is strange as concentrations in the order of 1000 ng/L were determined, and LOD in the order of few ng/L can be routinely obtained for Ba by using an instrument like Element1: e.g. Atmos. Environ. 39 (2005), 6420-6429). I would recommend the authors to review the reliability of the concentrations of every single TE and to eliminate from the data set all of those variables (Ba, Zr, Hf, Ta? as suggested in section 27655 line 19) that have a median value too close to the LOD (3 x LOD could be taken as a critical level, for instance).

2) The quality assurance information reported by the authors for Hg and TE determinations are too much limited. They might want to provide information (even using Supplementary sections, if necessary) about: accuracy, precision, blanks levels (procedural and instrumental), external calibration curves, limit of detections and instrumental parameters used (e.g. isotopes determined, kind of resolution used etc.). The authors may also show in a Table the main statistics of the concentrations (mean, median, min, max etc.).

3) To make more robust their conclusions about the crustal or anthropogenic origin of Hg certain trace elements and, the authors may try to calculate the crustal enrichment factors by using the most appropriate TE as a crustal reference (AI?) (e.g. Atmos. Environ. 39 (2005), 6420-6429) or to compare TE/AI mass ratios in the rain samples with the expected mean terrestrial crustal ratios: see for instance Geochim. Cosmochim. Acta 59 (1995) 1217-1232, Tab. 2.2).

4) Factor analysis: it is important that the authors show the total variance explained by each factor. Probably they could focus on the description of the data set by using just 3 factors (as they do later) without mentioning factors 4-5-6 of the preliminary factor analysis (thus eventually eliminating Table 1 and keeping Table 2).

5) If the assumption of the local origin for As, Sb, Se etc. is correct (27659, 11-15),

seasonally, they may behave like Zn (Fig. 4b). The authors may try to plot As, Sb, Se etc in the same way to eventually make more robust their case.

6) The method to disentangle the local and the remote component of Hg (illustrated with the Equation 1) should be presented more clearly by stating and discussing the basic assumptions made.

7) If appropriate, the authors may also compare their Hg fluxes with the critical loads for this ecosystem.

Some specific points:

Section 27652 lines 2-13: This is more likely part of the conclusions and may be moved to the end of the text. The authors should also report more homogeneously the TE with their chemical symbol (e.g. "Hg" and not "mercury").

Section 27652 Study area. In this part the authors should describe the geography of the territory involved in this study and try to put it in an appropriate larger context. I feel that they could move most of the preliminary information about the local regional emissions in this region to the introduction. Likewise all the information characterizing the rain samplers should be moved to the method section. They should recall Fig. 1 within the text: here, as long range transport seems to be important for Hg and other TE, it would be probably appropriate to display Florida also at continental scale.

A graph indicating the precipitation and winds regimes during the sampling period may be useful to better appreciate the fluxes variations and the provenance of the trace elements.

Figure 2: I would suggest to use some arrows to indicate the parts described within the text (e.g. "splash guard" in Fig. 2a)

27654 line 18: The percentage of of HCl and HNO3 spiked and the grade of HCl used should be specified.

C10562

27655 lines 10-15: This part should be expanded and more quantitative details should be provided.

27657 lines 2-4: This is interesting and important: the authors may show a comparison of the distributions of few TE concentrations and their fluxes and put in evidence whether they are quasi-normal or not by using the appropriate statistical tests.

27658 line 3 and wherever necessary: the authors should specify the level of significance assigned.

27659 lines 5-10. The authors should discuss the chemical form of the local Hg as well.

Table 3: nss-SO4 is not a TE.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 27649, 2009.