**Atmospheric
Chemistry
and Physics
Discussions**

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

W. M. Landing et al.

wlanding@fsu.edu

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Response to Referee Comments on: Interactive comment on "Atmospheric wet deposition of mercury and other trace elements in Pensacola, Florida" by W. M. Landing et al. Received and published: 20 January 2010, Review of ACPD 9, 27649-27674

Anonymous Referee #1, Major Comments, Factor analysis: I agree that this set data is a good candidate for factor analysis. Nonetheless, I do have a number of concerns about how the analysis was performed and how the results are presented and interpreted.

1.The appropriateness of this technique depends on the data being fairly normal. Based on figures, it looks like most of the deposition data are log-normal, which must

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be converted to a normal distribution (i.e. take log). Did you do this, or did SPSS do this automatically? Either way, this needs to be explain and/or corrected. This is important because factor analysis works to explain normally distributed variance, and if your data are log-normal, it will be disproportionally biased toward the element(s) with even slightly greater variance. You state that using deposition rather than concentration helps make them more normal, but considering they are log-normal, what do you mean? You should convert them to log, and test them for normality (e.g. Lilliefors test or Jarque–Bera test) and report. If they are close to normal you can leave them. It they are not normal, you should try to remove some outliers (e.g. Grubs test), reanalyze, and compare the results.

Response: The data were in fact log-transformed prior to factor analysis, and this is now explained in the paper. The log-transformed data had been visually inspected for symmetry, but we have now tested for normality using the Kolmogorov-Smirnov Statistic with Lilliefors' Significance. Only Cd (p=0.006) was found to be significantly different from a normal distribution; all other P values were greater than 0.2. The p values will be reported in Table 1 (which will be altered in response to Comment 4, to show only 3 factors). Statistics may suggest that certain data are outliers, but the data have already been pre-screened to remove anomalies due to known sampling, sample handling, and analytical errors, so without a physical justification we would prefer not to remove them from the data set.

2.Why was Varimax rotation used for the factor analysis? Varimax rotation tends to maximize the variance of a few of the individual features within a component (e.g. Na, Mg, Sr in Component 3) rather than the entire component. How do you know, a priori, that this more physically meaningful than unrotated? It may be correct, but you really should have a clear physical reason that is evident using unrotated components. Please justify why you've done this. Or, perform an unrotated analysis and compare to the rotated analysis. Also note that using rotated factors can usually be better supported if you have several factors with eigenvalues well above 1, but the values are

similar (i.e. not clearly distinct, see 3 below).

Response: Varimax rotation was chosen precisely because it maximizes the variance of the squared factor loadings. This is a commonly employed procedure because it tends to produce a simple structure (in the sense of Thurstone, 1947) in the retained factors such that each chemical species is associated with one or a few factors, thus making the results easier to interpret. Since Varimax is an orthogonal method, the factors remain uncorrelated. Unrotated solutions have been generated and inspected but leave the variable loadings spread over a large number of factors, making interpretation difficult.

3.Significance of the factors. Simply having an eigenvalue greater than 1 does not mean a factor is unique or statistically significant or physically meaningful. A set of purely random data will often produce at least one factor with eigenvalue >1. I don't know of a way to evaluate the significance of Varimax rotated factors, but it is straightforward with unrotated factors. See North et al. (Mon. Wea. Rev. 110:699 -706, 1982). You should use this to get an idea of how many factors are significant, likely only 2 or 3. Then, even if you want to use rotated factors, you shouldn't exceed this number of significant factors.

Response: Since we are employing factor analysis as an exploratory, rather than a confirmatory technique, the choice of how many factors to retain is less critical. Our final selection is based on a number of considerations: eigenvalues, scree plots, broken stick modeling, percentage of variance and interpretability. Because there is no universally accepted method of making this decision and our primary interest is in characterizing potential sources, interpretability is considered most important. We recognize that the number of factors retained will influence the outcome of the Varimax rotation. The results shown are those deemed most representative and informative from several analyses retaining and rotating different numbers of factors, as determined by application of all the methods listed. Our methods will be more clearly stated and we will avoid the use of the term "significant" where it might be construed to have a specific

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statistical meaning.

4.Table 1 and 2. Considering Components 4-6 are likely not unique (statistically significant, see above), showing the analysis with 3 and 6 components is unnecessary and not helpful. You should show only the analysis with 3 components. If the unrotated and rotated components look very similar, then just show the one that makes more physical sense and mention that both analysis are similar. If they are not similar, then you need to rethink if they really are physically meaningful and will have to better defend which one you use.

Response: We agree that there are only three factors that make obvious physical sense: a mineral dust factor, a sea-salt factor, and a "pollution" factor that includes Hg, As, Se, Sb, and non-sea salt sulfate. We will eliminate the table showing 6 factors and only use one table showing three factors, to be called Table 1, and edit the text appropriately to defend our interpretation of these three factors.

Also, reporting RËȨ2 values hides the sign of the correlation within each factor, which is very important for interpretation and crucial for the reader. Please change these tables to R values with signs. The reader can readily square them if they want the fraction of variance (RËȨ2) explained.

Response: We agree, and have changed the R^2 values to R values. The biggest impact of this is that chloride shows negative R values for the correlations with the "crustal" elements such as Al, Fe, Si, Th, etc.

5. If component 2 (or factor 2) is truly representative of coal combustion (as you assert and I would agree) then you should be able to use this (component 2) to estimate annual Hg deposition due to coal combustion, which be very interesting to compare with you other method using the minimum Hg:TE ratio (discussed more below). I don't know if SPSS allows you do this, but in matlab it very simple to recreate a time series of any of the components. In case the author is not familiar with this technique, as long as you are keeping track of the units and convert them back, you can obtain the values

of the original data (daily deposition in the respective units) due to only that specific component, as a time series. This can then be readily divided in to the appropriate years and an annual average calculated.

Response: We agree with this comment and have done the analysis as suggested. We find that Factor 2 (the "coal combustion pollution factor") accounts for 36% of the annual Hg deposition, as compared to the range of 22-33% obtained using the Hg:TE ratio technique (shown in the original Table 3). We think this is actually very good agreement, since the two estimates are obtained by very different analyses of the data. Adding this result to the text will add some length to the paper, but we agree that it is important to add.

Estimation of local-regional contribution to local Hg deposition This method for estimating the local-regional contribution is crude and is not based on a clear theoretical principle. Or, perhaps is just to be explained in more detail. The description should be revised and the author needs to address at least these additional issues: Why use the minimum Hg:TE ratio? This needs to be explained directly. After thinking about the equation shown, I can see this could be supported by the following: At the minimum ratio, it is assumed that 100% of the Hg deposited is due only to local sources. Therefore, the TE are a proxy (or scaling factor) for Hg deposition. Then, the scaling factor during those minimum ratios are assumed to be representative of the entire year, and thus the annual deposition can be inferred. Two problems: 1) You assume that either 100% of the Hg deposited in these minimum ratio events is due to local sources, or that it includes some fraction of global background, in which case the ratio is an overestimate. Neither of these is mentioned. Further, it almost certainly can't be 100% local Hg unless you're observing total deposition in these events which is vastly greater (say 100x) than from global background (i.e. your maximum Hg:TE ratios, or values from models). If this is the case, it should be clearly stated. 2) You assume that the ratio of Hg:TE during the minimum events is representative of the ratio of (local Hg):TE for the entire year, but you don't state it. This is a very critical assumption, and I think it is only

crudely correct.

Response: Two reviewers asked that we explain this calculation better, and we will do so, but Reviewer 1 has in fact interpreted it correctly up to this point. There are additional assumptions involved, for example that while Hg in rainfall can come from multiple sources (local, regional, and global) our calculation assumes that 100% of the As, Se, Sb, Sn, and nss-SO4 come from local plus regional sources (where "regional" means within a 1000km radius) . We will very carefully explain the assumptions we make in this calculation, and include a discussion of how the conclusions would change if the assumptions are changed.

Basically, this ignores the critical step of the elemental Hg getting into the rain, which requires oxidation! The amount of Hg that will be taken up by the rain depends on a number of factors, not the least of which is the amount of oxidized Hg (RGM) in the atmosphere. The local-regional oxidation of Hg (both locally emitted and from the global background or long-range transport) to RGM will be seasonal and highly variable, as will subsidence and entrainment of upper tropospheric RGM.

Response: We are not ignoring that GEM must be oxidized to RGM to get into rain. We expect that local and regional emissions of RGM could impact our sites, while local and regional emissions of GEM will probably not (due to the slow GEM oxidation rate). We further expect that the global contribution to Hg in our rain samples is due primarily to washout of RGM that has been formed in the "background". While our experiment was not designed to enable us to investigate the atmospheric processes producing RGM, we are in fact studying the meteorological conditions surrounding each rain event in an effort to gain more insight into the various RGM sources to our study region. This will be the subject of future papers.

Long-range transport The author describes deposition as either coming from local-regional sources or from long-range transport. This omits contribution from global background, which is a much larger contributor than long-range transport. (Selin et

al., 2008) Long-range transport generally refers to specific air masses or pockets of air which are distinguishable from the global background and can be linked to a source. Perhaps the author means "global background" when they say "long-range transport" (e.g. 27659, 11, 16)?

Response: The Reviewer is correct that we mean something like "global background" when we use the phrase "long-range transport". However, at what point does an Hg emission source change from being "local" to "regional" to "long-range" to "global"? Given the long atmospheric residence time of GEM, one could imagine that the rainfall Hg at our sites includes at least some Hg derived from GEM oxidation to RGM from every point source in the northern hemisphere, while the impact from any single source would be a complicated function of its emission rates of GEM and RGM, the distance to our sites, the meteorology in between, and the rate of net GEM oxidation between emission and deposition (which should both vary seasonally). The influences from RGM sources are expected to "decay" away relatively quickly with distance, but a very large RGM source far away could still have an impact at our sites. One could interpret "global background" to mean "from outside the continental US", but a large source in Cuba could be considered "long-range" or perhaps even "regional" relative to Pensacola. So, when we use the phrase "long-range transport" we mean something farther away than what one might call "regional". If we define "local" to mean within a 200km radius, then we would be concerned mostly with RGM sources since GEM emissions would not oxidize fast enough to matter much. If we define "regional" to mean within a 1000km radius, then we would perhaps want to consider large and small sources of RGM and large sources of GEM. Beyond 1000km, we would perhaps be considering very large RGM sources, moderate-to-large GEM sources, and whatever blows in from other countries. So, the "global background" in Pensacola should include some "long-range transport" from distant sources in the US, Canada, and Mexico. We have clarified the text with this set of definitions in mind.

Specific Comments Page, line 27650, 23: 'half-life' is incorrect. If you are reporting

values of 1/2 to 2 years, then you mean 'lifetime'.

Response: Yes, lifetime is what we meant.

27651, 19, 23, 24: need references for these three Hg emission rates

Response: These are from the 2005 EPA National Emissions Inventory; the proper citation has been added.

27652, 8 to end of paragraph: This sentence is awkward and the point isn't clear. What is meant by 'significant fraction?' Do you mean that a majority of mercury deposition is actually from long-range transport but that local sources do contribute 22-33% of mercury dep? See above, also, I think you mean a majority of deposition is from the background. Generally, using 'significant' without further explanation is indicative of ambiguity and subjectivity.

Response: We will modify the text in this section to reflect our conclusion that the majority of the rainfall Hg in Pensacola comes from distant sources (global plus long-range transport) and that on the order of 20-30% can be attributed to sources that we interpret to be "local plus regional" coal combustion. We agree about the use of the word "significant", and will revise the text with this in mind.

27652, 25: please clarify which 2005 inventory, presumably EPA?

Response: Yes, this is from the 2005 EPA National Emissions Inventory, and we will add the proper citation.

27653, 25 and remainder of paragraph on 27564: This paragraph belongs in the Methods section.

Response: We agree, and will move this text to the Methods section.

27657, 12: Remove the detailed interpretation of Components 4-6. See general comments regarding Factor Analysis. You have no way to establish that these factors are unique (much less statistically significant or physically meaningful) and the fraction of

variance they explain is very small. This discussion is purely speculation and should be condensed or removed.

Response: We agree with this comment, and have proposed to restrict the discussion to the three most significant factors. We will revise this section to reflect this.

27659, 5+: ". . .it becomes de-coupled from other tracers. . ." This is incorrect. Emissions such as gaseous elemental mercury remain coupled (correlated) with all other emitted tracers that have similar or longer atmospheric lifetimes. As the author is probably aware, multiple studies have used the enhancement ratio of observed Hg:CO to the emitted ratio of Hg:CO (i.e. they are still coupled 7+ days after emission) to identify long-range transport of coal combustion and biomass burning (e.g. Jaffe et al., Atmos. Environ. 39 (2005) 3029–3038; Slemr et al., Atmos. Environ. 40 (2006) 6966–6974). The author is probably referring to short-lived or soluble trace species and so should correct and clarify this section.

Response: We agree with this comment and will revise the text as suggested. The original intention was to contrast the behavior of GEM (long lifetime) with that of the other tracers of coal combustion (As, Se, Sb, Sn, and nss-SO4) (shorter lifetimes).

27659, 18: "TE" is trace elements, I presume. This should be clearly indicated or defined somewhere around here before you use only the abbreviation.

Response: We agree, and will define TE to include trace elements and other tracers, like nss-SO4.

All Figures, particularly #4: the fonts for the axes, labels, and legends are too small to easily read.

Response: We agree, and will enlarge the fonts in all figures.

Anonymous Referee #2 Throughout the manuscript the authors use the term "significant" or "significantly" without providing the actual statistics to support their claim of significantly higher or significantly lower. If there are statistics, then provide them in paren-

thesis following the statement. If there are no statistics, then choose another word to describe a large or noteworthy difference that does not indicate statistical significance. The authors use anthropomorphic words and this should be corrected. For example "this study looks at", when I read the work "look" it implies having eyeballs/eyesight, which uses human terminology to describe an inanimate scientific study.

Response; We agree with the comments above, and have added the statistical significance information where appropriate, and have edited the text to eliminate anthropomorphic phrases.

Specific comments are provided below. The abstract changes from past to present tense Why split the abstract into 2 paragraphs (one is 4 sentences long and the other is 3 sentences long)? Since the authors do not discuss the nutrients and major ions in this paper, they could omit that sentence and just have a one paragraph abstract.

Response: We have fixed the tense change, and merged the Abstract into a single paragraph. The nutrient and major ion data are indeed included in the suite of chemical species we use in the factor analysis (see Table 1).

Section 27652, Lines 7-12 are not clearly written. Lines 7-8 states " Thus, a major source of mercury to rainfall in the Pensacola area appears to be via local and regional combustion of coal." Then lines 8-12 state " Our data demonstrate that coal combustion in the SE US pollutes the regional airshed with volatile elements such as mercury, arsenic, antimony, selenium, tine , and non sea salt sulfate, but that a significant fraction of the rainfall mercury appears to be the result of long range transport". How can all three (local, regional, long range) be the dominant source? What is the major source; local and regional emission of Hg, Hg from long range transport (perhaps 5-6 states away), or Hg from the global hemispheric pool (which is Hg from everywhere) that is "scrubbed" out of the atmosphere during the almost daily summer convective thunderstorm activity (which also accounts for the high amount of precipitation during the summer) in Florida and then the air shed resuplied with Hg from the global hemi-

spheric pool via wind patterns that move air in a strong flow from the SE to the SW across FL (trade winds). Also note the use of the term "significant" without statistics.

Response: We agree with these comments, and have edited this section to rank the importance of "local plus regional" vs. "long-range plus global" sources of Hg to our rain samples. We have added the statistical significance where appropriate.

Section 27653 Lines 1-5. Emissions are quantified for the Crist plant, International paper boiler, medical waste incinerator, and a landfill in the area. Lines 4-5 state that "A significant source of atmospheric mercury and trace elements is wild fires, which occur frequently in the region.". The authors should provide a reference for this statement or provide the quantity of Hg and trace metals emitted during the wild fires. They do acknowledge further in the paper (section 27657) that there is a Cd, P, Zn component, that they cannot explain, and that they are analyzing aerosol samples from controlled burns, to try to verify this component, and that is good, but they don't have an actual reference in section 27653 to support their assertion that wildfires contribute a significant amount of Hg and other trace metals in this region. They might want to re-write the sentence to say something like, "although the emissions have not been quantified, burning in the area might release large amounts of Hg and other trace metals. Other studies by ????? et al., have demonstrated : : :..". There are some publications on Hg release during biomass burning of rainforests and large scale forest fires.

Response: We agree with these comments. We have not yet acquired the aerosol samples from controlled burns, as we had hoped, so we cannot address the importance of wild fires to Hg deposition in our study region. Also, in response to comments from Reviewer 1, we have limited our discussion of significant factors to three components. Because of these two issues, we have edited the text to remove discussion of the "Cd, P, and Zn" component and our speculation on what process it might represent. We propose to re-address this issue in the future, if aerosol samples from local and regional wildfires support the hypothesis.

Section 27654 Lines 5-10 discuss bottles and changing collars/funnels about every 6 months. This seems to be a slight lapse in QA/QC. This changing should occur on a routine, fixed schedule, not just "about every 6 months. It is understandable to change when you have bird droppings or other debris, but there should be a fixed change out schedule. Although it is difficult to "blank a rain sampler" were collar/bottle blanks checked and if so, what were the blanks?

Response: We have edited this section to state that our experience in the Florida Atmospheric Mercury Study showed that there was no need to arbitrarily change funnels and collars when they were not obviously contaminated (Landing et al., 1998; already in our reference list). We have indeed checked funnel/collar blanks, and that information has been added.

Sections 27654 and 27655 Hg and ICPMS analysis. What were the recoveries? That is part of QA/QC.

Response: We state on 27654 that the whole-bottle acidification/UV oxidation method we use recovers 100% of the rainfall Hg, and we demonstrated this during the FAMS project (Landing et al., 1998). We have added text to explain that we check the for recovery for the other trace elements by re-treating a randomly selected subset (10%) of our Teflon receiving bottles (filling with ultrapure water, acidified to the same levels as for samples, and UV oxidized), and that these "bottle blanks" are not significantly different from reagent-only blanks, and are very low relative to the trace element concentrations in our rain samples. We could include a table comparing these "bottle blanks" to the minimum trace element concentrations in our rain samples if the reviewer and the editor feel it would be helpful.

Section 27660 This section discusses the % Hg from coal, as quantified by using the Hg/TE ratio, TE deposition, and Hg deposition. This is the most creative part of the paper. The authors might want to redefine it and instead of saying % Hg from coal, they might want to say "% Hg from local sources". There are several reasons for considering

this; (1) although the coal plant emits the most with respect to the local emissions inventory there are other sources (landfill, incinerator, automobiles and other vehicles, and possibly biomass burning) in the region, so it is not all from coal, (2) the Hg in the global hemispheric background pool is a mixture of natural and anthropogenic sources (of these coal burning from other countries is a major contributor). It is my impression that the authors are trying to quantify the amount Hg in rain that is derived from the local anthropogenic inventory relative to what might, come from the global pool, which contains natural emissions and anthropogenic elemental gaseous Hg emissions that have traveled hemispherically, become oxidized to Hg2+ and rained out in their region. A large part of the Hg in the that comes from other long range or global sources is most likely from coal, so the authors might want to say % Hg from local sources, or % Hg from local emissions, and define "local emissions" within a certain kilometer radius of your sample sites.

Response: We agree with these comments. Our calculation for the "% Hg from coal" is indeed meant to refer to Hg from "local plus regional" Hg emission sources, the vast majority of which are in fact coal-fired power plants. The calculation is based on the assumption that 100% of the As, Se, Sb, Sn, and nss-SO4 come from "local plus regional" coal-fired power plants since such pollutants will have relatively short atmospheric residence times so that the contribution from "long-range plus global" sources of those tracers should be small. The influences from reactive gaseous mercury (RGM) sources are expected to "decay" away relatively quickly with distance, but a very large RGM source far away could still have an impact at our sites. Given the long atmospheric residence time of gaseous elemental mercury (GEM), one could imagine that the rainfall Hg at our sites includes at least some Hg from every point source in the northern hemisphere, while the impact from any single source would be a complicated function of its emission rate, the distance to our sites, the meteorology in between, and the average rate of net GEM oxidation between emission and deposition (which should both vary seasonally). One could interpret "global background" to mean "from outside the continental US", but a large source in Cuba could be considered "long-range" or

perhaps even "regional" relative to Pensacola. So, when we use the phrase "long-range transport" we mean something farther away than what one might call "regional". If we define "local" to mean within a 200km radius, then we would be concerned mostly with RGM sources since GEM emissions would not oxidize fast enough to matter much. If we define "regional" to mean within a 1000km radius, then we would perhaps want to consider all sources of RGM and large sources of GEM. Beyond 1000km, we would be considering very large RGM sources, moderate-to-large GEM sources, and whatever blows in from other countries. So, the "global background" in Pensacola should include some "long-range transport" from distant sources in the US, Canada, and Mexico. We will clarify the text with this set of definitions in mind.

The text on figures 3-7 is to small. The graphs should be redone with larger font so one can see the axis, scale etc.

Response: We have re-drawn the figures and enlarged the fonts.

Anonymous Referee #3 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 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).

Response: Our interpretation of the obvious scatter in the Ba data in Fig. 3D is that our "total" method, which includes all aspects of sample collection, handling, processing,

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and ICPMS analysis has resulted in relatively "poor" agreement for Ba between some of the A/B sample pairs, while many of the samples across the entire concentration range do indeed fall on the 1:1 line. The longer list of elements in the text is simply to show which elements we analyze for. The subset of elements used for the factor analysis in Table 1 is indeed the set of trace elements for which we obtained "reliable" data. We will clarify this in the text.

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.).

Response: We can create a new table that includes this information, if the editor agrees that it would be useful.

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 (Al?) (e.g. Atmos. Environ. 39 (2005), 6420-6429) or to compare TE/Al 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).

Response: We agree with this comment, and have modified our calculation to subtract the "crustal" or "mineral dust" fraction prior to using the "excess" Hg and trace element data to estimate the fraction resulting from local plus regional coal combustion. This correction for Hg and the volatile tracers is on the order of 1-2%. The impact these corrections have on our Hg:TE ratio calculations are very slight, since the corrected TE fluxes are in the denominator of the minimum ratios from the data set (making the

minimum ratios slightly larger), while the minimum ratios are then multiplied by the corrected annual fluxes (making the products slightly smaller).

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).

Response: We agree with this comment, which is similar to comments made by Reviewer 1. We have modified the paper to discuss only the three most significant factors we see in the data: mineral dust, sea-salt, and an anthropogenic component that includes Hg, As, Se, Sb, Sn, and nss-SO4. We have included the variance results in the revised Table 1.

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.

Response: Once again, we are trying to keep the paper reasonably short while trying to understand and quantify the sources of rainfall Hg. The seasonal deposition pattern varies for each element, and we only show a few as examples. Also, Zn is not a volatile elements like Hg, As, Se, Sb, Sn, and nss-SO4, and is not known to be strongly associated with emissions from coal-fired power plants. It is shown as an example of a "pollutant" element that is found as a primary aerosol in the atmosphere.

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.

Response: We agree, and have edited the text to discuss our assumptions more clearly.

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

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for this ecosystem.

Response: The critical loads for Hg in the freshwater and coastal marine environments of Florida are being studied now under a mandate from the US EPA. The results won't be available until 2012. Our goal in this brief paper is to show how we can use multi-element analysis to conduct a semi-quantitative source apportionment for rainfall Hg. Our Introduction does mention that atmospheric Hg gets into fish, and also mentions anthropogenic pollution of the atmosphere with Hg.

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").

Response: We have edited the text to use element symbols rather than element names. In this section of the Introduction, we are briefly summarizing our findings while the Discussion section describes how we reached these conclusions. This is a "style" issue, and as a reader, I prefer it when an author briefly describes his major findings as early in the paper as possible, to be followed by the in-depth discussion.

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.

Response: We are not sure how much geographical information we should include, if any. We would need more specific guidance on exactly what to include. The discussion of the local and regional emissions is in integral part of why the study area was chosen, so it seemed appropriate to put that text in that section. We have moved the description of the rain samplers to the methods section. Online maps are so readily available

from Google maps or Google Earth that we did not think it was necessary to add an additional map just to show where Florida sits.

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.

Response: We are in fact working on evaluating the meteorological conditions for each rain event to see if we can find factors that will help us understand why each rain sample has the chemistry it has. The results of this effort will be reported in the future. We could add more figures showing the precipitation patterns and wind regimes on any temporal scale one might want to consider (event based, weekly, monthly, seasonal, annual) however that would add length to the paper and, based on our meteorological evaluations to date, would not add anything meaningful to this paper.

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

Response: We agree, and have added arrows to Fig. 2 illustrating the components.

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

Response: The concentrations and grades of the acid used to acidify the samples are specified in the sentence immediately before this sentence. We prefer the use of molar concentration units, since this avoids the misunderstandings that can arise when acid concentrations are expressed as percentages.

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

Response: We are saying that we found no significant differences in the ICPMS analytical sensitivities (commonly measured in counts per second) for external standards or standard additions to actual samples. This should not be a surprise since the acid ma-

trix (0.048M HCl plus 0.45M HNO3) is nearly identical for all samples and standards, and since the rain samples are essentially very dilute solutions.

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.

Response: In response to Reviewer 1, we have already added statistical results regarding the normality of the data distributions for the log-transformed flux data that was used for factor analysis. We would have to add more figures and text if we make the suggested change, and would need guidance from the editor.

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

Response: We agree, and will specify the level of significance where appropriate.

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

Response: We agree, and will add text stating that the Hg emissions from Florida and the surrounding five states (Mississippi, Tennessee, Alabama, Georgia, and South Carolina) average 36-41% RGM (oxidized Hg) and <10% particulate Hg. The Hg emission speciation from the Crist Plant is mentioned in the Introduction.

Table 3: nss-SO4 is not a TE.

Response: We agree, and will refer to nss-SO4 as a "volatile" and a tracer of anthropogenic pollution rather than a trace element.

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

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