Atmos. Chem. Phys. Discuss., 9, C9936–C9941, 2010
www.atmos-chem-phys-discuss.net/9/C9936/2010/

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.

Anonymous Referee #1

Received and published: 20 January 2010

Review of ACPD 9, 27649-27674

Journal: ACP Title: Atmospheric deposition of mercury and other trace elements in Pensacola, Florida Author(s): W. M. Landing et al. MS No.: acp-2009-574 Special Issue: Atmospheric mercury (ICMGP2009)

Overall Comment

This manuscript presents wet deposition data for three sites near Pensacola, Florida from 2005-7. The analysis included a suite of elements, including important trace metals, but the manuscript focuses on mercury. The authors use factor analysis along with ordinary least squares correlations to interpret the sources of the data. Finally, the authors infer the fraction of annual mercury deposition due to local-regional sources.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

The topic of mercury deposition is important and relevant to ACP. The finding is not unexpected. But, the technique used to infer the contribution of local sources solely from observational data is sufficiently novel (to the best of the reviewer's knowledge) to warrant publication, after the authors address a number of concerns.

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 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.

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.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

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).

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.

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.

Also, reporting Rˆ2 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 (Rˆ2) explained.

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.

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. 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

Full Screen / Esc
Printer-friendly Version
Interactive Discussion
Discussion Paper

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.

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 airmasses 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)?

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’.

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

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.

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

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

27657, 12: Remove the detailed interpretation of Components 4-6. See general com-

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

ments 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.

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.

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

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

---

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