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Atmospheric Chemistry and Physics Discussions

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Interactive comment

# *Interactive comment on* "Decoding long-term trends in the wet deposition of sulfate, nitrate and ammonium after reducing the perturbation from climate anomalies" *by* X. Yao and L. Zhang

**Anonymous Referee #2**

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General comments

This paper asserts that statistical trends analysis of the linkage between emissions changes and measured wet deposition is obscured by multiple factors including climate anomalies. The target analytes of wet deposition measurements (SO4-2, NO3-, NH4+) undergo complex atmospheric transformations from their emitted precursors and washout or entrainment in precipitation are dependent on the climate. The climate anomalies are not specifically identified, but evidence exists that they influence relationships between wet deposition and emission trends and are occurring more frequently. Thus, a need exists for a statistical analysis technique to reduce the impact

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of the climate anomalies and increase the time interval of comparisons as emission changes, especially those due to regulations, are phased in incrementally and are not linear. The authors propose a statistical method based on the development of an input dataset termed “climatology” (an average of 12 ranked 24-month wet deposition flux measurements) and trends analyses to produce regression slopes for each of the 12 individual 2-year periods considered and the averaged “climatology” dataset. The regressions used are based off the Mann-Kendall (M-K) method, linear regression (LR), and piecewise linear regression (PLR). The authors propose that the time-series of the derived regression slopes better reflects the trends in reported emissions of precursor gases, than the time-series of the annual wet deposition flux data itself.

The method is novel and the m-value time-series relates better to emissions time-series than wet deposition flux (Fwet) time-series at Site 1 for SO4-2 and to a lesser degree for NO3. The m-value time-series appears to reflect inflection points in the emissions time-series that are not as easily observable in the wet deposition flux time-series. However, the method does not improve the relationship of m-values of NH4+ to NH3 emissions at site 1. Furthermore, the method does not seem to show improved m-value correlation with emissions over the annual Fwet data any other location (at Sites 2, 3, and 4) or species. There is no direct comparison metrics to gauge the improvement of the m-values over the annual Fwet other than visual interpretation of plots. The m-value time-series will obviously be visually “cleaner” since a) the m-value has outliers removed and b) the m-value represents 24 datapoints and the annual Fwet represents 12 datapoints.

The largest problem with the study is that that technique is not demonstrated to be robust. The method hinges on the stability of the m-values, but they are very susceptible to the large-value outliers (e.g. example described in text for ‘90-‘91, causes a 0.2 change in m-value; shown in Fig 1). Moreover, for Site 1, the authors acknowledge that 8 of 12 (67%) of datasets needed to have an outlier removed, which from my interpretation greatly compromises the robustness of this technique and its applicability to different datasets. There appears to be a m-value error analysis conducted with three different approaches in the Supplemental section (Approaches A through C), but no summary or assessment of m-value stability or uncertainty is given. This needs to be developed much more.

Sources of uncertainty in the m-values are not discussed. A reader will likely ask why are large Fwet values so frequently (the 8 of 12 datasets mentioned above) in disagreement with the rest of the monthly values? This question is not answered. What causes the large flux (other than climate anomaly?) Is this a high or low rain event month? Is this rain after a stagnation event?

There is too much assigning uncertainty to vague “Climate anomalies” and “interannual climate variability”. These concepts are neither adequately defined nor is any impact that they might have on monthly wet deposition values identified. The section on “interannual climate variability” could be strengthened with local ambient concentrations which are possibly available. At the very least, some more detail and explanation describing the meaning of Fig 4 and how it was derived and its effect on sulfur could be provided.

The reader will also pause as to why so much network-validated data is omitted. Most of the rationale appears to be statistically based (i.e. ‘because it doesn’t fit the trend’; see the $\pm 3\sigma$ criteria presented on line 173) which is insufficient without some scientific support (see the discussion on uncertainty of m-values above). More worrisome is the omission of the m-values (i.e. omission of 24 network-validated datapoints) in 1999 on the basis that they don’t fit the expected emission trend and are “probably caused by a large perturbation in climate anomalies”, but no real evidence is presented.

Specific comments (Individual Science Q)

On page 8, the authors state (line 172) which in turn increase the relative contribution of the air pollutants’ emissions to the calculated value. I assume that the authors are presuming that a monthly change in emissions would not impact the Fwet as much as a large monthly change in precipitation depth or concentration in precipitations. This point should be stressed more in the discussion.

The text does not adequately describe Fig 2 (lines 208 to 213). What is shown and why? I assume the objective of this plot is 1) to show the improvement of the fitted trend of the top row (m-value time-series) to the middle row (Fwet time-series). A metric (correlations with emissions?) is needed to demonstrate the advantage of the m-value over the annual Fwet.

The secondary objective of Fig 2 is to show the incremental trends or "phases". The plots do not currently accomplish this as Phase 1, 2, or 3 are not shown. Also, the PLR segments for Phase 1, 2, and 3 identified in the text are not described. The overall fits shown (e.g. R2 = 0.81 in 2a and R2 = 0.62 in 2b) are not significant in the analysis, but are shown on the plots. The PLR segments should be shown for the emissions as well (or at least compared with the 2a PLR segments).

Considering both of these objectives, the strength in this technique appears to be that the PLR segments for the top-row more closely resemble the PLR segments for the bottom row and that the PLR segments for the middle row do not reflect this. Please reorganize the discussion and analysis to support this. For example, the lines from 283-293 describing the improvement of the m-values over the annual Fwet data should be elaborated on and moved up in the discussion.

I assume the phase year classifications proposed by the authors (Phase 1, 2 and 3) are derived from the emissions data patterns, but the logic behind the years of the phases is not specifically discussed (i.e. why 1988 to 1993 and not 1995?) Do the phases align with emissions regulation implementation?

The PLR segments are often derived from a set of points as low as N=5 (e.g. Phase 1 from 1988 to 1993). Comparisons should state that this is a low N for comparison. On lines 348 -354; the m-value time-series for Site 2 NO3- (Fig 3d) is interpreted to support the decadal shift hypothesis. However, strictly observing the data, without the





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hypothesis in mind, it is clear that the four m-values from 1990 and ’96-’00 are elevated, while the values from ’92 and ’94 are similar to values observed after the decadal shift has taken place. This is acknowledged in the text, but no support given other than it is attributable to climate anomalies.

TECHNICAL COMMENTS 1. Figures need descriptive captions and local explanations. 2. Labels on Fig 2 (title incorrect) 3. line 223: “in contrast”. Suggest removal, not really in contrast. 4. line 237: Vlaue should read value 5. For Figure 1, distinguish the outlier point removed for each plot (as done in Fig 2) also specify which fit (R2 and p-value applies to the modified fit (I believe it is *, but it is not labeled).

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