

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 #1

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Review for ACP-2019-418 Decoding long-term trends in the wet deposition of sulfate, nitrate and ammonium after reducing the perturbation from climate anomalies. Xiao-hong Yao and Leiming Zhang

General comments

The objective of the study is to understand the effect of emission reduction on long term trends of wet deposition of inorganic ions. In that purpose, the effects of climate anomalies must be isolated to better highlight emission reduction effects. A two decade dataset of wet deposition of SO42-, NO3- and NH4+ is studied with a new approach at rural Canadian sites. A new method is applied to extract trends and inflection points,

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by pre processing the data sets and applying further common statistical tools such as M-K and L-R methods.

The presentation of the new pre processing method based on slopes for monthly wet deposition fluxes during 2 year periods should be clarified and better explained to avoid losing the reader. Indeed, there are several explanations that would need to be better justified to highlight the true added value of this new method.

The summary stipulated that more robust results are found with this new method, but this seems to be only true because some points are excluded from the correlation analysis. The robustness of the method needs further justification. This sentence on robustness in the abstract has to be removed, unless it is really justified.

This analysis is based on the assumption that removing the maximum wet deposition flux corresponds to removing climate anomalies, based on a pre processing of the data: this is exactly the point that has to be better justified, because all the analysis of the results relies on that statement.

When reaching the conclusion, the reader understands that the role of climate anomalies is also very important. The displayed purpose of the paper should be to highlight both the roles of climate anomalies and emission trends, rather than only focusing on emissions. The link with climate anomalies is also an interesting way in understanding the wet deposition flux trends.

The statistical approach lacks from being scientifically justified in terms of geophysical variable influence. I recommend major revision for this study, especially concerning the climate anomalies justification. Indeed, if this part is not well justified, the rest of the study cannot follow.

Generally, a table with a summary of different phases of trends for each site and each ion would help to better capture the results.

Specific comments

Introduction

Wet deposition fluxes of sulfate, nitrate and ammonium are affected by emissions of precursors, atmospheric processes, and climate anomalies. A definition of complex atmospheric processes and climate anomalies that are specifically linked to this study would be useful. Moreover, some more details are expected to explain these three processes, references to literature are not sufficient.

Methodology

This paragraph should be separated into different sub-paragraphs, with 2.1 statistical methods (line 112), 2.2 Data sets (line127), 2.3 Filtering climate anomalies (or something like that, line 152).

Line 125 and below: it is not clear why you use annual wet deposition fluxes as input data, whereas a modified dataset is based on monthly wet deposition fluxes. A figure would be useful to understand how this new dataset is built.

Line 145, what is the scientific explanation of excluding the maximum deposition flux when it deviates from the general regression? You only give a statistical explanation, which does not help in understanding the underlying geophysical causes.

Line 154: do you mean twelve two year periods of data?

Line 159: again, you mention the exclusion of maximum values of wet deposition fluxes, which leads to an increase of the R2 values: this is obvious statistically, but the scientific justification of this exclusion must be clarified. The explanation line 167 that maximum values are believed to be caused by climate anomalies is just a hypothesis and not sufficient to prove that you can exclude this maximum. Furthermore, this paragraph about filtering and excluding values is a bit strange in the methodological section, as it presents results already.

Line 190: Comparisons between this new approach (Approach C) and traditional approaches (A and B) are given in supporting information. Why a 12 month period is

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used in approaches A and B, whereas a 24 month period is used in approach C?

Line 195: what do you mean by "a small portion of climate anomalies that are unable to be removed by the new approach"? This is not precise enough.

Results and discussion

Line 208: as mentioned in the general comment, all the analysis of results here relies on the assumption that removing maximum wet deposition fluxes are associated to climate anomalies, which has to be better justified in the methodological section.

Line 210: please specify Fig 2a for SO42-, 2d for NO3- (and so on).

Line 212: where can we check that NH4+ exhibits a stable trend from M-K analysis, in Fig 2?

From line 215 and below, are you still commenting Fig 2? Please specify to facilitate the reading.

Line 241: the sharp increase in NO3- wet deposition flux in 1999 is supposed to be due to a "probable large perturbation from climate anomalies": this is not sufficiently justified. A scientifically argument needs to be provided.

Line 252: "Note that... here" should be declared in the method section, not in the results section. Moreover, R2 are written in the figures, and the text stipulates that R values will be used: this is not consistent.

Line 268: again, perturbations from climate anomalies unable to be removed by the new approach needs to be specified: what can they be exactly? What do they represent in terms of geophysical variables?

Line 282: please detail "many other factors" for describing NH4+ trends.

Line 293: after comparing m-values and annual deposition fluxes in the paragraph (lines 284-293), what is the interpretation of the statements? What do you want to

highlight here?

Line 301: paragraph 3.2 should be separated into different sub paragraphs (based on ions for example).

Line 388: again, justify which climate anomalies you are talking about to remove m-values

Line 413: what is the reason of unrealistic emission inventory? It could be useful to recall here which emission inventory is used here.

Conclusions

Line 456: this statement about the importance of climate anomalies vs emission trends is really interesting but unfortunately it is not specified earlier as an objective of the study: rather than removing climate anomalies, the purpose of the study could be to highlight the roles of both emission trends and climate anomalies, depending on the periods. The conclusion ends with results consideration that should be in the results section. The conclusion has to be more general and give some general clues for the interpretation of results that were presented. In the present state, it seems that the conclusion is not terminated.

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