

Reply to the comments and suggestions of reviewer #1

The study presented is very rich in data and analysis. However, some of the information could be summarized making the manuscript more readable, especially in the results section. Every sub-section of the results section has a summary after the description of the results, the summary is good but the text prior to it is overwhelming and unnecessary when having adequate figures/tables supporting the text. The figures and tables should be more carefully thought through, as it takes a lot of time to grasp the information.

Re: Following the suggestions from the reviewer and Tom Butler (Reviewer #2), we have substantially revised the text to make it more concise and readable. The graphs and tables have been rearranged, eg. Fig. 2 has been moved to Supplemental materials, and a more concise Fig. 2 (as suggested by the reviewer) has been added.

The reader might have issues to grasp what was done every single step of the way to obtain the results and the analysis, as the methodology is not really clear. This might be a consequence of not having a dedicated section for the methodology. Additionally, it might be challenging the reading when interchanging time periods and not giving a clear context, especially when trends and changes in concentrations are calculated based on different time period, and not really explained why.

Re: Following the suggestions of the reviewer in the detailed part of the review, we have substantially revised the text to have a clearer context in presenting the results, and explained, for example, why we calculated 10 and 25 years of changes. We also added a dedicated subsection “Statistical analysis and method” to explain how the analysis was carried out.

More information is needed to understand the levels of concentration behind the clusters, this might be due to emissions, meteorology, terrain, etc. It is important to know if one cluster can be more populated than others in terms of sources, and if that is still the case throughout the study period.

Re: The fundamental justifications for clustering of 4 regions are: (1) difference in emissions of SO₂, NO_x and NH₃; (2) the latitudinal gradient of the region, which affects the temperature and the solar radiation; (3) the prevailing atmospheric circulation due to mid-latitude Rossby waves. Regions 1 and 4 can be separated from Regions 2 and 3 because of the low emissions of SO₂, NO_x as well as NH₃, and also their low/high latitudes; Regions 2 and 3 can be separated from each other due to the large difference in NH₃ emissions. We used ambient concentration of SO₂ during the cold season to indicate the difference in SO₂ emissions; used NO₃ and RNO₃ during the cold season to indicate the difference in NH₃ emissions.

To support the justification of the clustering, we have added a table in Supplemental materials to show the correlation coefficients of the ambient annual concentration of each site vs. the regional averaged value for each species. For most site and species (except for NO₃), the correlation coefficients are larger than 0.95. Also as shown in the time series (Fig. S2), the annual concentrations of sites within each region are highly correlated, which is a demonstration that the clustering of sites is successful.

Responses to detailed comments:

Section 1

P2/L20: the reference to WHO fact sheets should be substituted by papers available describing the impact of air pollution on human health.

Re: We have substituted the web link by the journal paper references.

P3/L4-P4/L25: the authors appropriately describe how emissions have been evolving throughout the study period and the legislation to enforce such changes, but there is no single reference to corroborate the numbers. Please add the references grounding your narrative. Additionally, only the US situation was described, there is similar discourse for Canada. Please revise.

Re: References have been added, and the discussion for Canadian emissions has been added.

P5/L1-4: the authors describe trend analysis studies for other regions than US, and this might be misleading, even unnecessary. Please consider to remove.

Re: The mentioned part has been removed, and the text has been revised.

Section 2.1

P6/L25 Please add a reference describing the CASTNET network

The authors describe the temporal resolution of the measurements available for each network, but was never mentioned which resolution is actually taken for the analysis; this is mentioned in the abstract only.

Re: The references for CASTNET have been added. How annual means were derived was added in the new "Statistical analysis and method".

Section 2.2

P7/L14: When referring to 'mean concentration', is it temporal mean and if so for which time period? Please describe this step.

Re: We have specified it was "3-years mean concentration of NO₃ and SO₂ of each site".

*P7/L24-25 Here the authors refer to clustering of data between 1989 and 1991. Please describe how was the clustering obtained and why these 3 years and not any other period.
Title for section 3 should only refer to results.*

Re: The method of how to cluster the sub-regions was now clearly specified in the text; We also added an extra table (Table S.3) in Supplemental materials, showing the correlation coefficients of the annual

concentration from each site vs. the averaged value of the cluster for each species. The text has been accordingly revised (P9 L18-24).

We have specified that we used the mean concentrations at the beginning of the period because SO₂ was highest and it clearly separated regions 1 and 4 from regions 2 and 3.

Section 3.1

Please consider to add a table with the average values and STD for each region, this would improve readability for quick check and cross comparison and avoid a long text.

Re: It has been added in Supplemented materials as Table S.1 and S.2.

P8/L18 Consider to enumerate the species considered in this section.

Re: It has been revised accordingly (P10 L3-4).

P9/L4 Did the authors mean “spatially uniform”?

Re: We specifically mentioned “was spatially uniform”.

P9/L16 Please add references to corroborate your assumption.

Re: References have been added, and more corroboration has been added. (P10 L24 – P11 L2)

P9/L8-13 There is a degree of repetition in this paragraph, suggestion that it would fit best at the beginning of the section, before going into detail about each region.

Section 3.2

P10/L16 Discrepancy between the time period described in the title and the introduction of the work.

Re: It was corrected.

P10/L19 Why was the year 2000 chosen for normalization?

P10/L20 Typically one would cluster the time series on the basis of similarity, usually comparing magnitude or time variation, but as referred before, nothing was told before how were the stations clustered.

Re: The text explaining why year 2000 was chosen for normalization was added (P12 L10-13). In the Section 2.3, we have substantially improved the justification of clustering. Essentially it was based on similarity (correlation coefficients, Table S.3) and magnitude of time variation.

A suggestion for Figure 2 to be moved to the Supplements, and have a figure with average across the region and its variation for both cold and warm. Note that it's hard to read a single plot and across. For figure 3 would be best to put cold and warm seasons together, helping the reader to analyze the results. There is a typo in the legend for SO₄₂.

Generally, the section lacks which figures the reader should be looking at while reading the results section.

Re: Fig. 2 has been removed to the Supplemental materials, and a new Fig. 2 (as suggested) has been added. Fig. 3 has been revised according to the suggestions. We also specified in the revised text the results presented in this section are mainly based on Fig. S3 and Fig. 3

Section 3.3 Generally, the section lacks which figures the reader should be looking at while reading the results section.

Re: We specified the results presented in Section 3.3 are summarized from Tables 2 and 3.

Table 3 There is a possible highlighting mistake for concentrations above 1.0 ug m⁻³, e.g. NO₃.

Re: The mistake was corrected.

P17/L10 Please explain why these years were chosen and how the averaging was calculated.

Re: The explanation has been added, and text has been revised accordingly (P19 L6-L14)

P19/L25-26 The last sentence (“the difference...”) is importantly mentioned but the only discussion being written so far. To keep it consistent, it should be moved to the discussion.

Re: We have revised the text, and added two more references for the extra reduction of NO_x during O₃ season. Because of lack of emission data, it is difficult to have a more in-depth discussion.

P22/L18 How was the normalization of the annual means done? Is it related to the year 2000? Please describe.

Re: We explicitly mentioned that “The regressed trends are normalized to the regressed value of year 2000 as this is the turning point for the trend of NO₃-“.

Figure 4 does not describe which of the panels is depicted the pollutants.

Re: The mistake was corrected.

Figure 5 Please explain why NO₃ shows a different pattern that the remaining species.

Re: In the caption of Fig. 5, we explicitly mentioned that “The dot lines link the annual concentrations from 1990 to 2016 for species except NO₃- to show the temporal trends”. The reason that we didn’t use dot-line for NO₃ is that NO₃ during cold season in region 3 has no trend.

Section 4

P23/L2-3 here is explained why the year 2000 is chosen, please revise the structure so this information is prior to the results.

Re: It was revised accordingly.

Section 5

P23/L16 Please revise section title.

Re: The title has been revised to “4.1 RSO₄ and correlations of RSO₄ vs. SO₂”.

P23/L19 Why did the authors chose RSO₂ instead of 1-RSO₄?

Re: We don’t know why “Sickles and Shadwick (2015) chose RSO₂. For us, because we are concerned about the fraction of SO₂ being oxidized to SO₄, so RSO₄ is a more direct metrics than RSO₂.

Figure 8b has a typo in the legend, and displaying the R would be a good addition

Re: The typo has been corrected. R² has been added in the graphs.

Section 6

The paper is already long, it does not need a summary. PL11-L20 is a section on their own, as there is no real description of the emission in the Eat-US and Canada.

Re: We have corrected it accordingly.

P30/L23 again inconsistency between periods.

Re: The inconsistency has been corrected.

We sincerely thank the reviewer for his/her exceptionally detailed, very careful, thoughtful, and constructive comments and suggestions. We thank the reviewer for putting time and efforts and helping to improve the quality of the manuscript.