

A review on the manuscript ACP-2019-567 entitled "Air quality in the eastern United States and Eastern Canada for 1990-2015: 25 years of change in response to emission reductions of SO₂ and NO_x in the region".

The paper is relevant for understanding the impact of emission reduction on air quality across eastern-US and Canada, between 1990 and 2015, providing a temporal and spatial analysis of observational data, and what are the possible chemical and physical mechanisms responsible for the that evolution.

General:

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.

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.

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.

Detailed:

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.

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.

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.

Chemistry Warm/cold

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.

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.

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.

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.

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

P9/L4 Did the authors mean "spatially uniform"?

P9/L16 Please add references to corroborate your assumption.

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.

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.

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 SO_4^{2-}

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

Section 3.3

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

Table 3 There is a possible highlighting mistake for concentrations above $1.0 \mu\text{g m}^{-3}$, e.g. NO_3^-

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

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.

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

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

Figure 5 Please explain why NO_3 shows a different pattern than the remaining species.

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.

Section 5

P23/L16 Please revise section title.

P23/L19 Why did the authors chose RSO_2 instead of 1-RSO_4 ?

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

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.

P30/L23 again inconsistency between periods