Atmos. Chem. Phys. Discuss., referee comment RC2
https://doi.org/10.5194/acp-2022-400-RC2, 2022

Comment on acp-2022-400

Gregory Wetherbee (Referee)

Referee comment on "Long-term declines in atmospheric nitrogen and sulfur deposition reduce critical loads exceedances at multiple Canadian rural sites, 2000–2018" by Irene Cheng et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-400-RC2, 2022

Re: Long-term Atmospheric Deposition of Nitrogen and Sulfur and Assessment of Critical Loads Exceedances at Canadian Rural Locations, Atmospheric Chemistry and Physics,

MS No.: acp-2022-400

Dear Editorial Support Team:

Thank you for the opportunity to review the subject manuscript. In addition to this correspondence, I would like to send my suggested edits and technical comments in two .pdf files. Please let me know if this would be acceptable as I'm not concerned about my anonymity.

The authors have prepared an important and valuable paper that documents the spatial and temporal variability of nitrogen (N) and sulfur (S) deposition across Canada for the 2000 – 2018 period. They relate their results to estimated critical loads of acidity for

aquatic and terrestrial ecosystems. The paper is well written and has excellent graphics. The supplementary materials are also very useful and informative. In fact, the supplement is actually a true extension of the paper, no doubt due to limitations for publishing the many figures and tables in the article. Therefore, preparation of the tables and figures deserves as much attention to detail as in the manuscript.

Most of my comments are editorial in nature. I delve into technical issues only where the authors need to provide a clear or more detailed explanation of their findings. One general correction that I suggest globally is to provide figure captions and table titles that are more descriptive such that the figures and table could stand on their own if copied and pasted into other media, especially emails. Each caption and title should state that the data came from CAPMoN sites for the duration 2000 – 2018.

My technical comments are as follows.

Manuscript comments.

- This is an important paper, and it deserves a catchy title. The paper is actually extremely heavy on spatial and temporal characterization of dry and wet deposition and very light on critical loads, but the title made me think that I was going to read a lot more about critical loads. A title that better describes the content and makes the reader want to read the paper is needed. I suggest something like:

"Long-term Declines in Atmospheric Nitrogen and Sulfur Deposition Supress Critical Loads Exceedances Across Canada, 2000 - 2018."

- Lines 73: missing a space.
- Line 75: continued to dominate? Over what period? How about "...continues to exceed reduced N deposition or exhibit..."
- Line 76: I did not take the time to read the cited articles for my review. Although these are well-respected studies, if you want to look at long-term trends, are not the more recent articles actually more important? Do you really need to cite work back to 2005? Are not the more recent studies sufficient?
- Figure 1: Missing label for New Brunswick.
- Line 122: I could not access the data on this site: https://data-donnees.ec.gc.ca/data/air/monitor/monitoring-of-atmospheric-precipitation-chemistry/?lang=en .
- Line 130: examination of
- Line 190: Please specify whether this is base cation atmospheric flux or the surface-water loading into the catchment or both. If this is done using surface-water concentrations, then I'm confused about where the data came from for these 31 lakes.
- Line 194: This sounds like you have lake water quality data for these lakes as well as stream discharge. What is the source of those data? Did I miss it? The sources should be footnoted for figures and tables containing the data.
- Line 227: See my comment on Figure S3 in the Supplement. This ranking doesn't do much for me, but Figure S3 does, and it could convey more information if the circles were color-coded by region as in S4.
- Line 241: Is ECCC, 2004 cited in your references list?
- Line 280: Table 1. I recommend that this information be displayed as a bar chart in the Supplement if you are limited to a certain number of figures.
- Line 338: Reference to Fig. 2 is incorrect. I think you mean Fig. S2 because Fig. 2 has no years, just sites.
- Line 341: Fig. 2 does not show dates, nor does it differentiate between pSO4 and SO2. Is this a correct figure reference?
- Line 375: For the novice dry deposition scientist, it is not obvious that when you say pSO4 in the text and refer to Fig. 3 where SO4 is shown in the graph that, since dry deposition is being plotted, the graph is actually SO2(g) and SO4(s). Same goes for the N species where HNO3(g) and NO3(g or s) and NH3(g or s)? I assume that this is correct since you talk about pNO3 and pNH4 in the text. By the way, I hope that readers are not confused into thinking that pNO3 = log10[NO3]. Bottom line is that confusion about phases of species should be eliminated.
- Line 380: Again, I would make captions more descriptive so that figures can stand alone when extracted from context of the article. Specify that these are CAPMoN site IDs. Might explain species represented in the legend with respect to phase as well.
- Line 401 and Fig. 4, Fig. 5: Those green labels are tiny! Please make those larger. Good caption that specifies "CAPMoN sites"!
- Line 436: How can you "confirm future trends"? I don't think that is what you really mean. How about "…required to enhance dry N deposition monitoring."
- Line 444: Table 4 title should read: …) at CAPMoN sites, based on....
- Line 481: Table 5 title should read: …) at CAPMoN sites across Canada.
- Line 486: Figure 6 caption: …N at CAPMoN sites during 2000-2018.
- Line 513: Figure 7 caption: …sulfur at CAPMoN sites during 2000 - 2018.
- Line 517: Awkward start of sentence. "Mean cold and warm season fluxes of total N were 1.4 to 9.3 ..."
- Line 526: I suggest revising these sentences: "Seasonal differences in total S flux were large at only a few sites. For example, the cold season flux at SAT ..."
- Line 530: Same sites, right? Suggest linking these 2 sentences: "...(Fig. 8), whereas wet ..."
- Line 534: Figure 8 caption: "...seasons at CAPMoN sites during 2000-2018."
- Line 554: The pNH_4^+ was not "relatively less important." I suggest: contributed a

small portion of total N deposition because pNH_4^+.. (See line 556 as this is what you really mean).

- Line 565: I suggest: "...driven by a decline in oxidized N species."
- Line 569: Table 6. I published Theil-Sen slopes with the same number of significant figures before as well. However, I wonder if 2 sig. figs might be one too many. If you round to 1 sig. fig, then you have a lot of these trends turn out to be equal for pre-2010 and post-2010. How much of a change in concentration or precipitation depth would constitute a change in trend on the order of 0.05 kg/ha-yr? Could changes in the pre- and post-2010 slopes be attributed to shifts in chemical analysis methods or biases, age or calibration of precipitation gages, etc.? I think that we (me included) can make conclusions about trends that might be due to changes in data quality that have nothing to do with the environmental signal. I'm not saying that you're wrong, but you might want to be careful to note that subtle shifts like this could be due to changing data-collection methods, bias, etc. Perhaps a non-parametric test (Wilcoxon Signed-Ranks?) between pre- and post-2010 concentrations and precipitation depths would help confirm subtle differences in the trend slopes for these two periods.
- Line 573: "...) at CAPMoN sites based..."
- Line 611: Figure 9 caption: "...of nitrogen at CAPMoN sites, 2000 - 2018."
- Line 616: Figure 10 caption: "... of sulfur at CAPMoN sites, 2000 - 2018."
- Line 636: You are not explaining that ratios >100 indicate removal of transboundary emissions in addition to domestic emissions, whereas ratios <100 indicate relative removal of only domestic emissions. At least this is what I think you are implying here. Seems to me like these ratios could also be influenced by the ways in which you delineate your source regions for the back trajectory analyses whereby ratios<100 could indicate that your source region is simply broader, whether the region is transboundary or not.
- Line 656: What do you mean by this? This seems like an odd interpretation. Do you mean that wet deposition removes pollutants from higher in the troposphere and higher up for some intense thunderstorms, thereby including contaminants from long-range transport, whereas dry deposition is more regionalized (i.e. "local")? This seems like what you're trying to say, which would make sense, but the brevity affects the clarity.
- Line 659: "...(CL) for oxidant-produced acidity for lakes near the the five stations: ALG, ELA, LED, BAB, and KEJ, ranged ..." Please be clear that your critical loads are for acidity produced by the oxidized N and S species, and that you are not evaluating critical loads of N, which could affect eutrophication or terrestrial plant communities. However, addressing N critical loads would be a nice addition to the paper, but obviously more work.
- Line 660: Specify that these are CLs for acidity. The units should be eq H^+ ha^{-1} yr^{-1}, correct?
- Line 661: Specify CL for acidity. I think you mean aquatic H^+ CL or aquatic acidity CL, but not CL of N deposition to the surface water, correct?
- Line 663: "...high CLs for these lakes (Table S9)."
- Line 666: It is interesting that you show this shift in deposition around 2010, and right around this time, the lakes seemed to respond to reductions in acidic input. However, you don't seem to draw that connection in the paper. Why not? Maybe the connection just needs to be more obvious?
- Line 684: Be consistent and use the CL abbreviation. "Terrestrial acidity CLs were estimated at 14 stations ..." Also, please see Table S10 comment in the supplement. I recommend that you explain these "min" and "max" terms a little better and provide a bit of context here where you are using the terms as well. Finally, "…(208 eq H^+ ha^{-1} yr^{-1})", right?
- Line 687: I'm confused by this. Are you evaluating S+N acidity or S for acidity and N for nutrient loading? Why treat these independently if you are looking at total acidity?
- Line 719: "Total S depositions for the respective periods were…"
- Line 731: I'm not sure that this is really a conclusion of this paper. It seems like there is a lot of discussion of items in this section that really are not conclusions of the

study. Are you able to use the subtitle: Discussion and Conclusions, or maybe just Discussion?

- Line 738: Well, Fig. 11 gives the impression that all is well and deposition is < CL for all areas now. Am I missing something?

Supplementary Information comments

Globally, I suggest that each figure in this Supplement should have a caption that is more descriptive of data sources and locations so that the figures will stand on their own if extracted from the document and displayed somewhere else. For example, in Figure S4, the graphs could be excerpted from the supplement, and they would not stand on their own because people generally do not know that EGB and LON are CAPMoN sites, nor do they know where they are located.

- Section S1.2: Citation for Zhang et al., (2008) is not in the references list.
- Globally include specifications that data were obtained at CAPMoN sites during 2000 – 2018. Also, for Tables S6, S7, and S8 captions should read "...deposition at CAPMoN sites to % change in NOx emissions in back-trajectory determined source regions."
- Tables S9 and S10 – What are the data sources for the tables? Please include footnotes to reference data sources.
- Table S11: "...) near CAPMoN sites after including..." Also, what are the source(s) of the lake and soil chemistry data? Please footnote those.
- Figure S2: One cannot read those tiny green labels.
- Figure S3: Perhaps include in the captions that the sites are ordered longitudinally from west to east? Wouldn't this also convey more information if the circles were colored by the same regional colors in S4 along with the same legend as in S4?
- Figure S4: Caption: "Annual trends in atmospheric S and N species concentrations (mg/m^3) at CAPMoN sites grouped by region."
- Figure S5a: This caption is great! This is what you need on every figure!
- Figure S6: Caption: "…(kg N or S/ha/yr) measured at CAPMoN sites during 2000-2018…"
- Figure S7: Caption: "…for the cold (Nov-Apr) and warm (May-Oct) seasons during 2000 - 2018 at CAPMoN monitoring sites denoted by site IDs plotted longitudinally from left (west) to right (east)."
- Figure S8: Change caption in the same way as described above for S7.
- Supplementary Information References: Add Zhang, 2008 as mentioned above.

End of Comments

Please also note the supplement to this comment:
https://acp.copernicus.org/preprints/acp-2022-400/acp-2022-400-RC2-supplement.pdf