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Comment on acp-2022-400

Anonymous Referee #3

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-RC3, 2022

General

This paper is well written and generally clear, with strong support for the key takeaways. It provides a comprehensive overview of almost two decades of changes in observed or observationally derived deposition estimates in Canada, and uses this data to assess the impact on sensitive ecosystems with a critical load comparison. It's likely that this paper will serve as a key reference for future deposition research, and I believe that there are some places where the manuscript would benefit from the authors' experienced perspective on their vision for future directions in deposition research. Including this is optional, so that I have marked specific locations for this in the minor technical comments section. Although I list below several suggested revisions, most are optional/very minor and relate to re-formatting the figures or sentence re-wording for clarity. The manuscript is overall in very good shape and in my opinion would be ready for publication following a few regarding the language around the reduced and dry deposition analyses.

Major, conceptual comments

In general, I take issue with the representation of "total N deposition" and in particular "total reduced N deposition," when the authors estimate that neglecting dry ammonia deposition results in a 32% underestimate in N deposition overall. With respect that this analysis is well-founded on the best available measurements, I find the comparisons of wet and dry (section 3.3.2) and reduced versus oxidized deposition unconvincing because the reduced component only includes a minority contributor, pNH_4^+. At present the reduced versus oxidized N discussion undermines one of the paper's key conclusions, that more observations of NH_3 and organic N deposition are needed. While the authors include careful discussion on the limitations of the approach given the available measurements, it would improve the clarity of this manuscript to refer to reduced N deposition more specifically. For example, the comparison of oxidized N with reduced N is really a comparison between the majority component of oxidized N and pNH_4^+.

Major, technical comments

Line 201: Including the limitations of this approach related to soil N saturation and the role of N deposition in acidity is helpful. However, this signal is offset by an under-estimate in total N deposition used in this calculation, and I would suggest mentioning that.

Table 1: As this table is described as for the period 2000-2018, should the Atlantic sites include only KEJ? I believe that the others could bring down the average significantly because they were only operational in the latter part of the period.

Lines 452 – 453: I'm not convinced that the N fluxes in Canada were lower than in the USA because of the underestimate in total N used in this study, which could bring the Canada-based fluxes to a closer level to those in the USA (9.5 * 130% = 12.4). Indeed, the following paragraph cites Zhang et al. 2009 to express that the site-based fluxes can be as high as 11.6 kg/ha-y when including NH_3, NO_2 and organic N. Consider restricting this clause to only S or clarify the estimate for Canada-based N fluxes when accounting for the species included in the US flux estimates.

Lines 478-479: Is the clause "however, oxidized N deposition continued to be greater than reduced N deposition in the west coast, prairie, and Atlantic regions" correct? It seems to contradict lines 457-458.

Figures 6, 7: Include in the legend or elsewhere that oxidized N includes only HNO_3 and pNO_3^-, while reduced includes only pNH_4^+.

Line 548-549: Clarify this is specific to pNH_4^+.

Line 566: Again, please clarify that "reduced N" is specific to pNH_4^+.

Table 6: I do appreciate the note in the caption of this table, but I think it would be clearer to clarify that "total reduced N" is pNH_4^+ (and possibly also that oxidized N is HNO_3 and pNO_3^-) in the table heading.

Minor, technical comments

Line 99: Consider rephrasing “meaning that the influence of local pollution sources is minimal” to "local pollution sources do not have an outsized influence"

Line 128: The way that this is written, “total reactive N” NO_y seems to exclude NO_x because NO_x is listed separately, but the below text indicates the NO_y measurement includes NO_x. Reactive N generally also includes reduced forms. Consider re-naming NO_y as “total oxidized N.”

Following paragraph lines 255-267: This is one place where I believe that offering some insight or context into anticipated directions or implications for your research would be helpful. For example, if trends continue (NO_x/SO_2 decrease while NH_3 increases out west), do you think that western ecosystems will be threatened? Would it be worth a more tailored study of NH_3 impacts in this region?

Line 295: It took me a while to understand that this paragraph is comparing the gas versus aerosol phase components of dry deposition, so it may be worth mentioning that specifically in the opening sentence, potentially: “the relative role of gas versus aerosol phase dry deposition of routinely-measured species…”

Lines 299-300: Based on Figure 2, I think that it is probably true that the Atlantic sites had lower fluxes through the whole period, but maybe it would be worth clarifying whether this is still true if you include only KEJ.

Figure 2: Consider clarifying which species are in gas or aerosol phase in the legend to improve agreement with the text.

Figures 4, 5, 9, 10: Consider including only the sites that you discuss in-text and moving the remaining figures to supplement. It is difficult to read the text in these figures in this format.

Line 441: Do you have any thoughts on why the rate of decline in annual dry S deposition would have accelerated at SAT and ELA after 2010?

Table 4: Nice, very clear table.

Figure 6: Is there a reason the sites are sorted in this way? Would it be possible to change to either alphabetical or sorted so that the % shifts in a consistent direction? I am concerned that passive observers may currently view this as a temporal trend.

Lines 530-531: Consider expanding on the impact of the seasonal patterns observed here—for example, does this have implications for terrestrial acidification?

Line 541: Consider noting that BON, MIN and BAB were not operational early in the period of study, which might influence the lack of significance in their annual trends.

Line 557: I'm not sure I understand what "is possible" – that dry deposition increased? Please consider clarifying. An alternative interpretation is that this references the ambient NH_3 concentration increases, which the cited studies demonstrate did increase (in other words, "possible" is not an appropriate descriptor).

Line 599: Is this finding robust when considering the impact of dry NH_3 deposition? Similarly, for lines 607-608, I think that it would be worth mentioning that this is also a conservative estimate of the contribution of dry deposition, in addition to reduced, for the same reason.

Figures 9, 10: Consider adopting the same color scheme, or some variant, as in Figure 2. It's a little confusing that some of these colors were used to denote the warm season in the more recent figures.

Sentence on lines 623-625: Is there a citation for the back trajectory analyses?

Paragraph lines 641 – 648: Would it be possible to speculate what the cited analyses imply for the reduced N deposition response in Canada? Do you have thoughts on what are the process-based drivers of the distinction in response between the USA and the UK (maybe based on precipitation, or aerosol formation chemistry, given differences in NO_x trends in either region)?

Also: Do your findings on the relatively high efficiency of dry versus wet N or S deposition suggest that an increase in dry N associated with NH_3 would cause a >100% increase in deposition per unit?

Line 702: The discussion on uncertainty in the calculation of CL in this section is very nice. Consider including also a discussion on the uncertainty associated with the atmospheric measurements as well, possibly related to this paper:

Walker, et al. (2019), Aspects of uncertainty in total reactive nitrogen deposition estimates for North American critical load applications. Science of The Total Environment, 690: 1005-1018, ISSN 0048-9697, https://doi.org/10.1016/j.scitotenv.2019.06.337.

Line 728: This doesn't change the overall point of this sentence, but from my read, Figure 11 seems to suggest that SBR had an exceedance in 2014.

Sentence starting line 738: could you give examples of where those sites should be based on your research?

Sentence starting line 741: consider expanding to include organic N as well as emphasizing the need for dry deposition constraints specifically. Also, please consider including more citations as this has been discussed recently, possibly:

Sutton et al. (2013). Towards a climate-dependent paradigm of ammonia emission and deposition. Phil. Trans. R. Soc. B3682013016620130166. http://doi.org/10.1098/rstb.2013.0166.

Walker et a. (2020). A review of measurements of air-surface exchange of reactive nitrogen in natural ecosystems across North America, Science of The Total Environment, 698(133975), ISSN 0048-9697, https://doi.org/10.1016/j.scitotenv.2019.133975.

Minor, syntactical comments

Sentence on lines 141 – 142: Not sure why this statement about NO_y follows a description of the NH_3 instrument. Consider moving it to start ~ line 134 (following the description of the NO_y instrument).

Line 225: "their pSO_4^{2-} and pNH_4^+ concentrations" – clarify whether "their" refers to west coast and prairie or southeastern.

Table 3: Move to precede last paragraph, so that it follows first mention of this table.

Figure 6: Move up to where it is first introduced.