

Atmos. Chem. Phys. Discuss., referee comment RC1  
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## **Comment on acp-2022-400**

Anonymous Referee #1

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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-RC1>, 2022

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### **General Comments (*evaluating overall quality of the preprint*)**

The article is a well written analysis of regional trends in air quality, atmospheric deposition, and critical loads across the Canadian landscape. The authors clearly describe the environmental changes that have occurred as a result of emission reduction programs in Canada as well the United States. There continues to be a need for more measurements, including the number of monitoring sites as well as the pollutants measured. As the authors note, they may be missing up to 32% of the total nitrogen budget by not measuring NH<sub>3</sub>, HONO, PAN, organic nitrogen, and other unmeasured species. The paper provides an important reference for the continued need for ground-based monitoring to improve and constrain the atmospheric deposition models.

The authors only looked at acidification of aquatic and terrestrial (soil) ecosystems, while reduced nitrogen is becoming increasingly important in both Canada and the US. I would suggest that the authors, at a minimum, include language on the idea that eutrophication may also be contributing to regions experiencing algal blooms, loss of biodiversity, vegetation damage, etc. from increases in nitrogen deposition.

Overall, the paper as written provides a substantial contribution to the scientific literature by providing a complete summary of the latest trends from the CAPMON network. The data collected over the past 15 years have been validated and presented using accepted criteria, the results are clear with complementary figures and tables.

### **Specific Comments (*addressing individual scientific questions/issues*)**

In the introduction include a short description on why NH<sub>3</sub> is difficult to include in dry deposition estimates but note that it is potentially a significant contributor to the overall nitrogen budget. Ammonia is not as important for looking at acidification, but it is worth mentioning as it's an area of uncertainty in the estimates of dry deposition.

In the paragraph starting with line 128 it's unclear how many detectors were used with the continuous analyzers. If multiple detectors were used, how were biases addressed? Some clarification is needed.

In the Results section it is important to be clear which time periods are being discussed. For example, it's unclear on line 221 if the highest concentrations were measured over the 15-year time period (the whole period) or the beginning/end of the period. Make sure you consistently describe the time period for which results are being shared.

The values in Table 1 don't match the results described on page 11. Please check the values in both places.

Around line 433, suggest including a statement about the potential increases in NH<sub>3</sub> dry deposition also increasing resulting in the weaker dry N deposition trends. Also on line 433, were there shifts in the vehicle miles traveled which would have caused a trend in NO<sub>x</sub> emissions? What about changes in the energy sector?

Around line 471 – can you say anything about the role that satellites may play in helping to address the gaps in measurements to evaluate spatial and temporal trends in deposition.

In section 3.4, can you speculate why there are spatial differences causing difference in the response of wet NO<sub>3</sub> deposition due to NO<sub>x</sub> emission reductions?

In section 3.5 it was not clear what the threshold was for a level of protection/level at which a species would not be harmed. (was it set at a deposition level where it was expected that 50% of the lakes were unaffected?)

In the Discussion suggest including plans to assess eutrophication impacts due to nitrogen deposition in each of the regions. This is an area for future work and it's not expected that results be presented but it should be mentioned as an important assessment for understanding ecological impacts around the monitoring locations.

## Technical Corrections

**Line 20:** "included due to lack of *monitoring* data."

**Line 66:** don't include "and/or NH<sub>3</sub>". This should be a separate sentence since NH<sub>3</sub> measurements were not included in this paper. The sentence should include why it is more difficult to include NH<sub>3</sub> in dry deposition estimates (e.g., few ambient measurements, high deposition velocity, bi-directional flux).

Line 73: "both to agricultural and wildfire emissions of NH<sub>3</sub>. Emissions reductions in NO<sub>x</sub> (*ref*) have also been shown to contribute to increases in free NH<sub>3</sub> (Yu et al. 2018 Atmospheres <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2018JD028412>)."

Line 83: suggest added Simkin et al 2016 from PNAS

Line 90: "total deposition (dry + wet) deposition *as they relate to changes in anthropogenic emissions;*"

Line 266: "Ongoing monitoring is required to assess the *air quality and ecological* impacts of these industrial activities on ambient N and S in *the Northern Great Plains region (US) and Prairie (Canada).*"

Line 324: "in agricultural *and unmanaged areas*, and its dry deposition estimates at EGB presented here should be treated as upper-end values *due to emission of NH<sub>3</sub> being unaccounted for.*"

Line 326: "including NH<sub>3</sub> concentrations and supplemental measurements that can be used to constrain bi-directional flux models (Walker et al. 2020)" <https://www.sciencedirect.com/science/article/pii/S0048969719331717?via%3Dihub>

Line 731: "the ecosystem *critical loads* are not exceeded, they do not indicate when ecosystems will *fully recover from pre-2012 acidification.*"

Line 739: suggesting adding a sentence about impacts from wildfires, extreme precipitation events, other climate related environmental stressors. May see new trends in deposition.

Line 740: include organic N in the list