Dear Prof. Fu (ACP Editor):

We are submitting a revised version of our manuscript (acp-2022-400), entitled "**Long-term Declines in Atmospheric Nitrogen and Sulfur Deposition Reduce Critical Loads Exceedances at Multiple Canadian Rural Sites, 2000-2018**", for further consideration in Atmospheric Chemistry and Physics. We have addressed all the comments provided by the four reviewers. Please see the enclosed response to reviewers for details. The submission also includes a manuscript and supplement file with tracked changes.

Thank you for taking care of the review process for this paper.

Sincerely,

Irene Cheng, Leiming Zhang, Zhuanshi He, Hazel Cathcart, Daniel Houle, Amanda Cole, Jian Feng, Jason O'Brien, Anne Marie Macdonald, Julian Aherne and Jeffrey Brook

Response to Reviewers' Comments
(acp-2022-400)

Reviewer #1

We appreciate the reviewer's comments on the overall impression of the manuscript and suggestions for further improvements. Our point-by-point responses to each comment are provided below.

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 NH3, 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.

Response: Reduced nitrogen is becoming increasingly important in both Canada and the U.S. and its deposition not only contributes to acidification of ecosystems but also eutrophication and loss of biodiversity. In the revised paper, we added the following text in the Introduction (1st paragraph). "In addition to acidification effects, increase N deposition accelerates algal growth in aquatic ecosystems and can negatively affect vegetation by decreasing plant species diversity, altering their physiology, increasing their vulnerability to parasites and pathogens, and reducing its ability to adapt to extreme weather (Bobbink et al., 2011; de Vries et al., 2015)."

Discussions were also included in section 3.5 (2nd last paragraph), which reads "It is important to note that only CL of acidity were evaluated in this study. N deposition contributes to eutrophication in N-limited aquatic and terrestrial ecosystems (Smith et al., 1999), as well as changes to plant communities (Field et al., 2014) including losses of species diversity (Bobbink and Hicks, 2014; Payne et al., 2017). The form of N deposited also has ecosystem health implications. Increased ammonium in the soil results in

lower base cation uptake, which can reduce tree growth (Gloser and Gloser, 2001; Bobbink et al., 2011; de Vries et al., 2015). Critical loads of N for eutrophication and biodiversity loss have not been widely established for Canadian ecosystems, but an area of future work is to assess eutrophication effects due to N deposition surrounding the monitoring stations (e.g. Forsius et al., 2021)."

Bobbink, R., Braun, S., Nordin, A., Power, S., Schutz, K., Strengbom, J., Weijters, M., and Tomassen, H.: Review and revision of empirical critical loads and dose-response relationships, Editors: Bobbink, R. and Hettelingh, J. –P, Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010, 2011.

Bobbink, R., and Hicks, W. K.: Factors Affecting Nitrogen Deposition Impacts on Biodiversity: An Overview, In M. A. Sutton, K. E. Mason, L. J. Sheppard, H. Sverdrup, R. Haeuber, and W. K. Hicks (Eds.), Nitrogen Deposition, Critical Loads and Biodiversity (pp. 127–138), Springer Netherlands, https://doi.org/10.1007/978-94-007-7939-6_14, 2014.

de Vries, W., Hettlingh, J.-P., and Posch, M.: Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems, *Environ. Pollut.*, 25, Springer Dordrecht, https://doi.org/10.1007/978-94-017-9508-1, 2015.

Field, C. D., Dise, N. B., Payne, R. J., Britton, A. J., Emmett, B. A., Helliwell, R. C., Hughes, S., Jones, L., Lees, S., Leake, J. R., Leith, I. D., Phoenix, G. K., Power, S. A., Sheppard, L. J., Southon, G. E., Stevens, C. J., and Caporn, S. J. M.: The Role of Nitrogen Deposition in Widespread Plant Community Change Across Semi-natural Habitats. Ecosystems, 17(5), 864–877, https://doi.org/10.1007/s10021-014-9765-5, 2014.

Forsius, M., Posch, M., Holmberg, M., Vuorenmaa, J., Kleemola, S., Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., deWit, H.A., Dirnböck, T., Frey, J., Grandin, U., Hakola, H., Kobler, J., Krám, P., Lindroos, A.-J., Löfgren, S., Pecka, T., Rönnback, P., Skotak, K., Szpikowski, J., Ukonmaanaho, L., Valinia, S., Váňa, M.: Assessing critical load exceedances and ecosystem impacts of anthropogenic nitrogen and sulphur deposition at unmanaged forested catchments in Europe, Sci. Total. Environ., 753, https://doi.org/10.1016/j.scitotenv.2020.141791, 2021.

Gloser, V., and Gloser, J.: Nitrogen and base cation uptake in seedlings of Acer pseudoplatanus and Calamagrostis villosa exposed to an acidified environment. In O. Gašparíková, M. Čiamporová, I. Mistrík, & F. Baluška (Eds.), Recent Advances of Plant Root Structure and Function (pp. 103–109), Springer Netherlands, https://doi.org/10.1007/978-94-017-2858-4_12, 2001.

Payne, R. J., Dise, N. B., Field, C. D., Dore, A. J., Caporn, S. J., and Stevens, C. J.: Nitrogen deposition and plant biodiversity: Past, present, and future, Front. Ecol. Environ., 15(8), 431–436. https://doi.org/10.1002/fee.1528, 2017.

Smith, V. H., Tilman, G. D., and Nekola, J. C.: Eutrophication: Impacts of excess nutrient inputs on freshwater, marine, and terrestrial ecosystems, Environ. Pollut., 100(1), 179–196, https://doi.org/10.1016/S0269-7491(99)00091-3, 1999.

Specific Comments (addressing individual scientific questions/issues)

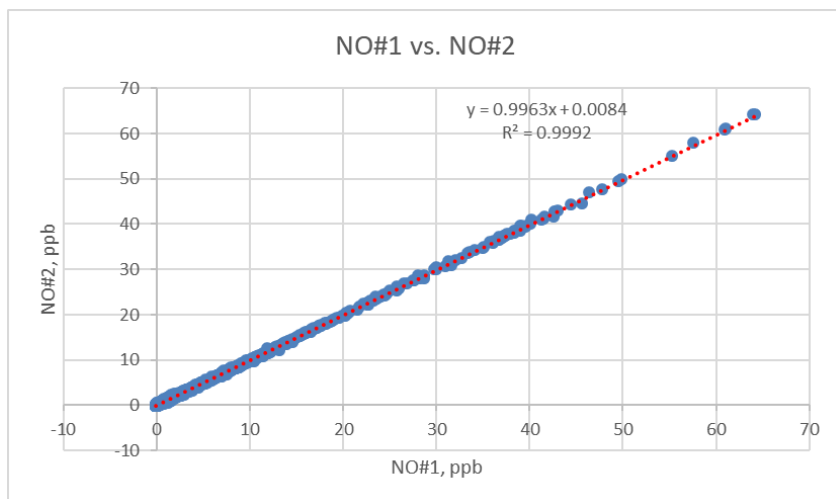
In the introduction include a short description on why NH_3 is difficult to include in dry deposition estimates but note that it is potentially a significant contributor to the overall nitrogen budget. Ammonia is not as important for looking at acidification, but it is worth mentioning as it's an area of uncertainty in the estimates of dry deposition.

Response: We agree with the reviewer that ammonia is an important component in terms of the total dry deposition budget. The main reason why NH_3 is not included in CAPMoN dry deposition estimates is because NH_3 has not been routinely measured by this network. We also agree with this reviewer's technical comment below that there are large model uncertainties associated with the bidirectional exchange of NH3 and its high deposition velocity, which make it difficult to include NH3 in dry deposition estimates. We have added a brief explanation in the Introduction (3rd paragraph), which reads: "Most estimates of dry N deposition include pNO_3^-, HNO_3, and pNH_4^+ given that their ambient air concentrations are routinely monitored or modeled. It is more difficult to include NH_3 in dry deposition estimates because of few ambient measurements and large model uncertainties associated with NH_3 bidirectional exchange and its high deposition velocity."

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.

Response: There were a total of three instruments (three detectors) that were deployed to measure NO, NO_2, NO_y, and NH3 (the latter measured by difference between total ambient nitrogen (Nt) and NO_y).

Instrument #1: NO and NO_y (unfiltered)
Instrument #2: NO and NO_c (NO_c=NO+converted NO_2)
Instrument #3: NO_y (particle-filtered) and Nt (NO_y particle-filtered) + reduced nitrogen (particle-filtered)

As all instruments are essentially monitoring NO in each channel, each instrument was calibrated every 25 h with the same calibration system, the same NO working gas and audited with the same NIST NO cylinder. Analog signals from each instrument were obtained on a common datalogger to minimize any potential effects posed by drifting clocks. We can evaluate if there are any biases by comparing the NO channel ambient measurements (5 min averages) on Instruments #1 and #2 as the sample paths are essentially the same. The plot below indicates an extremely good correlation of the two NO measurements with a negligible intercept (offset). Unfortunately, instrument #3 didn't have any common measurements with the other two instruments, but based on historical measurements at other sites, similar results have been observed for the same comparison.

We summarized this intercomparison result in section 2.1 (3rd paragraph) to explain that there were no biases between different NO detectors as follows. "NO, NO$_2$ and NO$_y$ were measured at 1-min intervals using Thermo Electron Model 42C trace level gas analyzers. The analyzers measured NO based on the chemiluminescent reaction with ozone. NO$_2$ was converted to NO using a Droplet Measurements BLC photolytic converter, while NO$_y$ (NO, NO$_2$, HNO$_3$, HO$_2$, pNO$_3^-$, N$_2$O$_5$, PAN, organic nitrates, and others) were converted to NO using a molybdenum converter heated to 325°C. An intercomparison of the NO measurement channels from different instruments indicated a high degree of correlation (slope=0.996, r2=0.9992) and minimal offset (intercept=0.008)."

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.

Response: The time period has been added in several places. Section 3.1 now reads, "Over the 2000-2018 period, the highest concentrations of atmospheric S and N were observed at LON and EGB…". Section 3.2.3 now reads, "The rate of decrease in annual dry N deposition ranged from -0.007 (BON) to -0.11 (LON) kg N ha^{-1} yr^{-1} during the 2000-2018 period."

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

Response: There was an error in the text. The sentences have been updated as follows. "Over the entire period (2000-2018), HNO$_3$ contributions to dry deposition of N were greater than those of pNO$_3^-$ and pNH$_4^+$ at all sites (Table 1). This is because the dry deposition velocity of gaseous HNO$_3$ is greater than that of particle N species (Table S2). The highest mean HNO$_3$ dry deposition was observed in southeastern Canada (0.6-0.8 kg N ha^{-1} yr^{-1}), but in the greater southeastern region (less affected by urban regional transport) dry deposition was lower than at the west coast and prairie sites." (paragraph after Table 2)

Around line 433, suggest including a statement about the potential increases in NH3 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 NOx emissions? What about changes in the energy sector?

Response: We added a sentence in the 2nd last paragraph before Table 4, which reads "Potential increases in NH_3 dry deposition could result in the weaker dry N deposition trends; however, this needs to be tracked through expanded ambient NH_3 monitoring across Canada." The major reasons for the NOx emissions decrease include the implementation of vehicular emissions regulations in both Canada (ECCC, 2022) and the U.S. (EPA, 2021) and reductions in the vehicle kilometers travelled (VKT) for the Light-Duty Gasoline Vehicles and Trucks categories in Canada (ECCC, 2022). In the U.S., vehicle miles travelled actually increased for the 2000-2018 period (EPA, 2022). While NOx emissions trends in eastern Canada were largely driven by those of the transportation and mobile equipment sector, declines in emissions from electric utilities also contributed to NOx emissions decrease in the eastern U.S. (Fig. S5a).

ECCC, 2022. Canada's Air Pollutant Emissions Inventory Report 2022: chapter 2.3. https://www.canada.ca/en/environment-climate-change/services/air-pollution/publications/emissions-inventory-report-2022/chapter-2-3.html

EPA, 2021. Reducing Emissions of Hazardous Air Pollutants. https://www.epa.gov/haps/reducing-emissions-hazardous-air-pollutants

EPA, 2022. Air Quality - National Summary. https://www.epa.gov/air-trends/air-quality-national-summary

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.

Response: This is indeed a good suggestion for filling the data gaps for species that are not routinely measured. We have produced NH_3 dry deposition across North America using satellite generated concentration data, but only for one season in 2013. Once the long-term satellite data are made available, we could revise the deposition budget for subsequent ecosystem impact studies. Since the present study focuses on long-term trends, we prefer to use only the available data from CAPMoN for generating the trends. While satellite measurements can be used to estimate dry deposition, there can be significant variability in the satellite-derived surface concentrations depending on the algorithms used for converting column data to surface concentrations (Liu et al., 2020 and references therein). Improved algorithms and further validation of satellite-derived concentrations with in-situ data will increase the usefulness of satellite measurements in the future. Satellite-derived N measurements have mainly focused on NH_3 and NO_2; other N species cannot be reliably estimated from satellite measurements currently which is also necessary for a complete N dry deposition budget (Liu et al., 2020). Another gap is that most of the acidic deposition at non-urban sites is in the form of wet deposition, which isn't measurable by satellites and requires additional wet deposition schemes. We have added a statement in section 3.3.1 (after Figure 6) as follows. "Satellite generated surface-level air

concentration data of some important nitrogen species (e.g., NH_3 and NO_2), once available for multiple years, could be potentially useful in filling the data gaps described in this study (Kharol et al., 2018)."

Kharol, S. K., Shephard, M. W., McLinden, C. A., Zhang, L., Sioris, C. E., O'Brien, J. M., Vet, R., Cady-Pereira, K. E., Hare, E., Siemons, J. and Krotkov, N. A., 2018. Dry deposition of reactive nitrogen from satellite observations of ammonia and nitrogen dioxide over North America. *Geophys. Res. Lett.*, *45*(2), 1157-1166.

Liu, L., Zhang, X., Xu, W., Liu, X., Lu, X., Wei, J., Li, Y., Yang, Y., Wang, Z., and Wong, A. Y. H., 2020. Reviewing global estimates of surface reactive nitrogen concentration and deposition using satellite retrievals, Atmos. Chem. Phys., 20, 8641–8658, https://doi.org/10.5194/acp-20-8641-2020.

In section 3.4, can you speculate why there are spatial differences causing differences in the response of wet NO3 deposition due to NOx emission reductions?

Response: The different responses in the west coast vs. eastern Canada could be due to differences in the relative frequency of air masses arriving from Canada and the U.S. and NOx emissions levels and trends. Regarding the latter, NOx emissions in western Canada were not decreasing; the levels were also greater than those of eastern Canada. In the U.S., NOx emission levels in the western region were also much lower than in the eastern region (Fig. S5a and b).

We added a sentence in section 3.4 (1st paragraph), which reads "Thus, both domestic and transboundary emissions reductions were pivotal in decreasing total oxidized N deposition in eastern Canada, whereas domestic emissions reductions were more effective in decreasing the deposition at the west coast site. The spatial differences in the responses could be due to differences in relative frequencies air masses from Canada and the U.S. and NO_x emissions levels and trends."

In section 3.5 is 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?)

Response: The approach used does not fix a threshold or a percentage of lakes that will not be affected. The critical load approach is based on an Acid Neutralizing Capacity limits value (corrected for dissolved organic carbon), which is a threshold that is protecting the biota. This was briefly explained in the methods section 2.4 (1st paragraph). The sentence reads, "Briefly, in the SSWC model, lake CL is quantified by the pre-acidification base cation flux exiting the lakes minus the Acid Neutralizing Capacity (ANC$_{limit}$), the threshold above which harmful effects on fish population are not observed (Jeffries et al., 2010; Aherne and Jeffries, 2015)."

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 its not expected that results be presented but it should be mentioned as an important assessment for understanding ecological impacts around the monitoring locations.

Response: We agree this research is needed. In section 3.5 (2nd last paragraph) where we discussed critical loads of acidity, we added the following text. "It is important to note that only CL of acidity were evaluated in this study. N deposition contributes to eutrophication in N-limited aquatic and terrestrial ecosystems (Smith et al., 1999), as well as changes to plant communities (Field et al., 2014) including losses of species diversity (Bobbink and Hicks, 2014; Payne et al., 2017). The form of N deposited also has ecosystem health implications. Increased ammonium in the soil results in lower base cation uptake, which can reduce tree growth (Gloser and Gloser, 2001; Bobbink et al., 2011; de Vries et al., 2015). Critical loads of N for eutrophication and biodiversity loss have not been widely established for Canadian ecosystems, but an area of future work is to assess eutrophication effects due to N deposition surrounding the monitoring stations (e.g. Forsius et al., 2021)."

Bobbink, R., Braun, S., Nordin, A., Power, S., Schutz, K., Strengbom, J., Weijters, M., and Tomassen, H.: Review and revision of empirical critical loads and dose-response relationships, Editors: Bobbink, R. and Hettelingh, J. –P., Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010, 2011.

Bobbink, R., and Hicks, W. K.: Factors Affecting Nitrogen Deposition Impacts on Biodiversity: An Overview, In M. A. Sutton, K. E. Mason, L. J. Sheppard, H. Sverdrup, R. Haeuber, and W. K. Hicks (Eds.), Nitrogen Deposition, Critical Loads and Biodiversity (pp. 127–138), Springer Netherlands, https://doi.org/10.1007/978-94-007-7939-6_14, 2014.

de Vries, W., Hettlingh, J.-P., and Posch, M.: Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems, *Environ. Pollut.*, 25, Springer Dordrecht, https://doi.org/10.1007/978-94-017-9508-1, 2015.

Field, C. D., Dise, N. B., Payne, R. J., Britton, A. J., Emmett, B. A., Helliwell, R. C., Hughes, S., Jones, L., Lees, S., Leake, J. R., Leith, I. D., Phoenix, G. K., Power, S. A., Sheppard, L. J., Southon, G. E., Stevens, C. J., and Caporn, S. J. M.: The Role of Nitrogen Deposition in Widespread Plant Community Change Across Semi-natural Habitats. Ecosystems, 17(5), 864–877, https://doi.org/10.1007/s10021-014-9765-5, 2014.

Forsius, M., Posch, M., Holmberg, M., Vuorenmaa, J., Kleemola, S., Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., deWit, H.A., Dirnböck, T., Frey, J., Grandin, U., Hakola, H., Kobler, J., Krám, P., Lindroos, A.-J., Löfgren, S., Pecka, T., Rönnback, P., Skotak, K., Szpikowski, J., Ukonmaanaho, L., Valinia, S., Váňa, M.: Assessing critical load exceedances and ecosystem impacts of anthropogenic nitrogen and sulphur deposition at unmanaged forested catchments in Europe, Sci. Total. Environ., 753, https://doi.org/10.1016/j.scitotenv.2020.141791, 2021.

Gloser, V., and Gloser, J.: Nitrogen and base cation uptake in seedlings of Acer pseudoplatanus and Calamagrostis villosa exposed to an acidified environment. In O. Gašparíková, M. Čiamporová, I. Mistrík, & F. Baluška (Eds.), Recent Advances of Plant Root Structure and Function (pp. 103–109), Springer Netherlands, https://doi.org/10.1007/978-94-017-2858-4_12, 2001.

Payne, R. J., Dise, N. B., Field, C. D., Dore, A. J., Caporn, S. J., and Stevens, C. J.: Nitrogen deposition and plant biodiversity: Past, present, and future, Front. Ecol. Environ., 15(8), 431–436. https://doi.org/10.1002/fee.1528, 2017.

Smith, V. H., Tilman, G. D., and Nekola, J. C.: Eutrophication: Impacts of excess nutrient inputs on freshwater, marine, and terrestrial ecosystems, Environ. Pollut., 100(1), 179–196, https://doi.org/10.1016/S0269-7491(99)00091-3, 1999.

Technical Corrections

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

Response: Revised according to your suggestion. (abstract)

Line 66: don't include "and/or NH_3". This should be a separate sentence since NH3 measurements were not included in this paper. The sentence should include why it is more difficult to include NH3 in dry deposition estimates (e.g., few ambient measurements, high deposition velocity, bi-directional flux).

Response: The sentences in the Introduction (3rd paragraph) have been revised. They now read, "Most estimates of dry N deposition include pNO_3^-, HNO_3, and pNH_4^+ given that their ambient air concentrations are routinely monitored or modeled. It is more difficult to include NH_3 in dry deposition estimates because of limited ambient measurements and larger model uncertainties associated with NH3 bidirectional exchange and its high deposition velocity."

Line 73: "both to agricultural and wildfire emissions of NH_3. Emissions reductions in NOx (ref)have also been shown to contribute to increases in free NH_3 (Yu et al. 2018 Atmospheres https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2018JD028412)."

Response: Revised according to your suggestion; the reference has been added. (Introduction)

Yu, F., Nair, A. A., and Luo, G., 2018. Long-term trend of gaseous ammonia over the United States: Modeling and comparison with observations. J. Geophys. Res. Atmos., 123, 8315–8325. https://doi.org/10.1029/2018JD028412.

Line 83: suggest added Simkin et al 2016 from PNAS

Response: Reference has been added. (Introduction)

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

Response: Revised according to your suggestion. (paragraph before section 2)

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)."

Response: Revised according to your suggestion. (paragraph before section 3.2)

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_3 being unaccounted for."

Response: Revised according to your suggestion. (paragraph before Table 3)

Line 326: "including NH$_3$ 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

Response: We have revised the sentence and included the Walker et al. (2019) reference. We agree that if additional parameters that are needed as model input for bi-directional flux models (such as the one described in Zhang et al. (2010)) can be measured, NH$_3$ dry deposition can be quantified using a bi-directional approach. Alternatively, using conservative V$_d$ from a big-leaf model (such as the one of Zhang et al. (2003) used in this study) is also acceptable.

The sentence now reads, "Nevertheless, given NH$_3$ was by far the largest contributor to N dry deposition for this one-year period at EGB, including NH$_3$ in routine monitoring and supplemental measurements (e.g. relating to air-surface exchange processes) should be a priority, in order to accurately assess the long-term levels and effects of N deposition and constrain bidirectional flux models (Zhang et al., 2010; Walker et al., 2019a)." (paragraph before Table 3)

Walker, J. T., Beachley, G., Amos, H. M., Baron, J. S., Bash, J., Baumgardner, R., Bell, M. D., Benedict, K. B., Chen, X., Clow, D. W., Cole, A., Coughlin, J. G., Cruz, K., Daly, R. W., Decina, S. M., Elliott, E. M., Fenn, M. E., Ganzeveld, L., Gebhart, K., Isil, S. S., Kerschner, B. M., Larson, R. S., Lavery, T., Lear, G. G., Macy, T., Mast, M. A., Mishoe, K., Morris, K. H., Padgett, P. E., Pouyat, R. V., Puchalski, M., Pye, H., Rea, A. W., Rhodes, M. F., Rogers, C. M., Saylor, R., Scheffe, R., Schichtel, B. A., Schwede, D. B., Sexstone, G.A., Sive, B.C., Sosa, R., Templer, P.H., Thompson, T., Tong, D., Wetherbee, G. A., Whitlow, T. H., Wu, Z., Yu, Z., and Zhang, L., 2019. Toward the improvement of total nitrogen deposition budgets in the United States. Sci. Total Environ., 691, 1328–1352.

Zhang L., Wright L.P., and Asman W.A.H., 2010. Bi-directional air-surface exchange of atmospheric ammonia - A review of measurements and a development of a big-leaf model for applications in regional-scale air-quality models. J. Geophys. Res. Atmos., 115, https://doi.org/10.1029/2009JD013589.

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

Response: Revised according to your suggestion. (2nd paragraph of section 4)

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

Response: We added a sentence in the conclusion, which reads, "The impacts of wildfires, changes in the atmospheric chemistry, extreme precipitation events, and other climate-related environmental stressors on N and S deposition trends require further study." (3rd paragraph of section 4)

Line 740: include organic N in the list

Response: The sentences in section 4 (3rd paragraph) have been revised as follows. "As oxidized N in air and in deposition continue to decrease, reduced N will comprise a larger proportion of the total N

deposition at a greater number of sites. Thus, a greater focus is needed to track ammonia emissions and reduced N deposition trends. It should be noted that organic N is not included in the total N deposition budget, which should be considered in future monitoring and analysis studies. This can be done by measuring dissolved organic N in precipitation and total NO_y in air, with the former for estimating organic N wet deposition and the latter for approximating organic N dry deposition (Walker et al., 2020)."

Walker, J. T., Beachley, G., Zhang, L., Benedict, K. B., Sive, B. C., and Schwede, D. B., 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.

Reviewer #2 (Gregory Wetherbee)

We appreciate the reviewer's comments on the overall impression of the manuscript and suggestions for further improvements. Our point-by-point responses to each comment are provided below. We also addressed additional comments that were in the reviewer's annotated PDF version (acp-2022-400-RC2-supplement.pdf).

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.

Response: We added more descriptions to the figure and table captions and noted in every caption that the data apply to CAPMoN sites for the 2000-2018 period.

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."

Response: Thank you for coming up with a better title, which we think would be more attractive to readers. Since the study assessed critical loads exceedances at only a subset of CAPMoN sites, we used "at Multiple Canadian Rural Sites" instead of "Across Canada". The updated title is "Long-term Declines

in Atmospheric Nitrogen and Sulfur Deposition Reduce Critical Loads Exceedances at Multiple Canadian Rural Sites, 2000 - 2018."

Lines 73: missing a space.

Response: Space added.

Line 75: continued to dominate? Over what period?   How about "...continues to exceed reduced N deposition or exhibit..."

Response: Revised the sentence to, "oxidized N deposition continues to exceed reduced N or exhibit increasing trends in some regions" (2nd last paragraph of Introduction)

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?

Response: Most of the references cited were published from 2015 onwards; only the study by D. Fowler et al. (2005) was published earlier. Although the recent studies on both dry and wet deposition trends are sufficient, the findings are limited to the U.S. and China. The study by Fowler et al. was conducted for U.K. sites, and we believe it is important to acknowledge that similar studies have been conducted elsewhere.

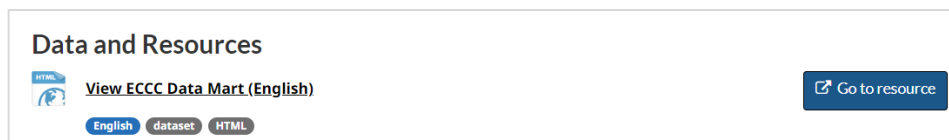
Figure 1: Missing label for New Brunswick.

Response: Labels have been added for all provinces and territories in Figure 1 (NB – New Brunswick, PE – Prince Edward Island, YT – Yukon Territory, NT – Northwest Territories and NU - Nunavut).

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 .

Response: We verified the links cited in the references and the datasets are accessible. However, on that page you need to click on the 'Go to resource' link to transfer to the page where the datasets (csv files) are housed (see screenshot below).

Environment and Climate Change Canada (ECCC), 2021b. Major ions. Government of Canada Open Data Portal. Available at: https://open.canada.ca/data/en/dataset/9974e51f-2616-42bf-8b40-ed12de91a304

Environment and Climate Change Canada (ECCC), 2021c. Major ions and acidifying gases. Government of Canada Open Data Portal. Available at: https://open.canada.ca/data/en/dataset/10ec2a54-9b6d-4dd7-9b05-5c30b9fa4920

Line 130: examination of

Response: The sentence was revised as follows. "Measurements at the EGB site in 2010 were analyzed in this study to estimate total N dry deposition after including the missing N species and to examine the relative contributions of various N species to N dry deposition, following the approach described in Zhang et al. (2009)." (paragraph before section 2.2)

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.

Response: It is the base cation fluxes exiting the lakes. It is now specified in section 2.4 (1st paragraph). Lake chemistry data were presented in Table S9. We have added the data sources in the table footnotes.

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.

Response: As mentioned in the previous response, the sources of the water quality data are now mentioned in the footnotes under Table S9. Runoff was calculated with the BioSim model for Quebec and Maritimes lakes, while direct discharge measurements were available for Ontario lakes (Experimental Lake Area and Turkey lakes watershed).

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.

Response: The rank order of the sites in the text has been deleted from section 3.1 (1st paragraph). As mentioned in your comment regarding Figure S3, the sites are arranged in order from west to east by longitude to be consistent with the discussion of spatial patterns in the text. The circles in Figure S3 are now color-coded by region as in S4.

Line 241: Is ECCC, 2004 cited in your references list?

Response: Yes ECCC, 2004 was cited in the reference list. We updated this reference by adding the URL to the report and the year at the end following the ACP reference format.

Environment and Climate Change Canada (ECCC) Meteorological Services of Canada: 2004 Canadian Acid Deposition Science Assessment, ISBN 0-662-38754-6, www.publications.gc.ca/pub?id=9.688243&sl=0, 2004.

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.

Response: Although it is nice to show the results in a figure, we prefer to present the dry deposition fluxes in a table in case readers would like to extract the data for other purposes.

Line 338: Reference to Fig. 2 is incorrect. I think you mean Fig. S2 because Fig. 2 has no years, just sites.

Response: The sentence is referring to the results in Fig. 2. Figure 2 is comparing the annual mean dry deposition fluxes and the relative percentages of the species for the 5-year period in 2000-2004 versus in 2014-2018. For each site, the first bar represents the 2000-2004 values and the second bar represents the 2014-2018 values.

We included more description in the Figure 2 caption as follows. "Figure 2: Dry N and S deposition fluxes (kg N or S ha^{-1} yr^{-1}) and the percentage of N or S species in dry deposition for the 5-year period in 2000-2004 and in 2014-2018 at CAPMoN sites. For each site, the first bar represents the 2000-2004 values and the second bar represents the 2014-2018 values. Gaseous species: HNO_3 and SO_2; particulate (p) species: pNO_3, pNH_4 and pSO_4."

Line 341: Fig. 2 does not show dates, nor does it differentiate between pSO4 and SO2. Is this a correct figure reference?

Response: The sentence is referring to the results in Fig. 2. The bottom-right graph in Fig. 2 shows the relative percentages of SO_2 (light blue) and pSO_4^{2-} (dark blue) dry deposition. For each site, the first bar represents the relative percentages in 2000-2004 and the second bar represents the relative percentages in 2014-2018.

Figure 2 has been revised. The SO4 in the legend has been changed to pSO4. NO3 and NH4 in the legend of the upper-right graph have been changed to pNO3 and pNH4. More description was added to the Figure 2 caption as detailed in the previous comment.

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.

Response: Figure 3 has been revised. The SO4 in the legend has been changed to pSO4. NO3 and NH4 in the legend of the upper-right graph have been changed to pNO3 and pNH4. The caption was also updated with more description. "Figure 3: Mean dry deposition fluxes of N and S species (kg N or S ha^{-1} yr^{-1}) during the cold (Nov-Apr) and warm (May-Oct) seasons during 2000-2018 at CAPMoN sites. C: cold season; W: warm season. Gaseous species: HNO_3 and SO_2; particulate (p) species: pNO_3, pNH_4 and pSO_4.

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.

Response: Please see response to previous comment.

Line 401 and Fig. 4, Fig. 5: Those green labels are tiny! Please make those larger. Good caption that specifies "CAPMoN sites"!

Response: The font size for the green labels (rate of change in deposition fluxes) has been increased in Figures 4 and 5.

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."

Response: The sentence was revised as follows. "Potential increases in NH_3 dry deposition could result in weaker dry N deposition trends; however, this needs to be tracked through expanded ambient NH_3 monitoring across Canada." (2nd last paragraph before Table 4)

Line 444: Table 4 title should read: …) at CAPMoN sites, based on….

Response: Revised according to your suggestion.

Line 481: Table 5 title should read: …) at CAPMoN sites across Canada.

Response: The Table 5 title was updated as follows. "Table 5: Regional breakdown of mean total (dry+wet) N and S deposition fluxes (kg N or S $ha^{-1} yr^{-1}$) across Canada. Regional values are based on deposition fluxes from representative CAPMoN sites."

Line 486: Figure 6 caption: …N at CAPMoN sites during 2000-2018.

Response: Revised according to your suggestion.

Line 513: Figure 7 caption: …sulfur at CAPMoN sites during 2000 - 2018.

Response: Revised according to your suggestion.

Line 517: Awkward start of sentence. "Mean cold and warm season fluxes of total N were 1.4 to 9.3 …"

Response: Yes it was an awkward beginning. Revised according to your suggestion. (1st paragraph of section 3.3.3)

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 …"

Response: Revised according to your suggestion. (2nd paragraph of section 3.3.3)

Line 530: Same sites, right? Suggest linking these 2 sentences: "…(Fig. 8), whereas wet …"

Response: The sentence in line 530 applies to most of the sites, whereas the previous sentence applies to west coast and prairie sites and two southeastern sites. We kept them as two separate sentences and revised the second sentence as follows. "At these sites, dry S and wet SO_4^{2-} deposition were nearly equivalent during the cold season (Fig. 8). At most of the sites, wet SO_4^{2-} deposition was greater while

dry S deposition was smaller in the warm season than in the cold season." (2nd paragraph of section 3.3.3)

Line 534: Figure 8 caption: "...seasons at CAPMoN sites during 2000-2018."

Response: Revised according to your suggestion.

Line 553 (from acp-2022-400-RC2-supplement.pdf): So, [pNH4+] didn't decrease because of declining pSO42- and pNO3-, but rather [pNH4+] did not decrease because SO2 and NOx concentrations declined, thus producing less particulate ammonium sulfate and ammonium nitrate, right?

Line 554: The pNH4+ was not "relatively less important." I suggest: contributed a small portion of total N deposition because pNH4+.. (See line 556 as this is what you really mean).

Response: The sentences were revised as follows to improve the clarity of the explanations. "Second, although ambient concentrations of pNH_4^+ decreased as a result of declines in ambient SO_2 and NOx concentrations, dry deposition of pNH_4^+ contributed a small portion of the total N deposition because pNH_4^+ was predominately in $PM_{2.5}$ which has a smaller V_d compared with coarse or ultrafine particulates (d < 0.1 μm) (Zhang and He, 2014). However, with less pNH_4^+ formed, ambient NH_3 increased over the U.S. and Canada (Butler et al., 2016; Yao and Zhang, 2016; Feng et al., 2021)." (2nd paragraph of section 3.3.4)

Line 565: I suggest: "...driven by a decline in oxidized N species."

Response: Revised according to your suggestion. (paragraph before Table 6)

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.

Response: For the total S deposition trends, the Sen's slopes pre-2010 and post-2010 were not very different and the interpretation may depend on the number of significant figures used. Two significant figures is very reasonable, but one is likely not sufficient given the deposition flux units are in kg ha^{-1} yr^{-1}. As suggested, the Wilcoxon Rank Sum test (or Mann-Whitney U test) was performed on the pre- and post-2010 annual precipitation-weighted (p-w) mean sulfate concentrations and annual precipitation amounts to assess whether the differences were statistically significant (p<0.05) between the two periods. Below are the p-values for the test, which shows the p-w mean concentrations were

significantly different between the two periods, whereas the annual precipitation amounts were not except at SAT. The differences in the sulfate concentrations in precipitation were statistically significant, which reflects changes in precursor emissions rather than changes in precipitation amounts. There were no changes to the CAPMoN data collection methods that could affect the wet concentrations during the study period. There were some changes to the CAPMoN lab analysis, but the impacts to the measured concentrations were negligible (J. Feng, ECCC, personal communication).

p-values from Wilcoxon Rank Sum test comparing the 2000-2009 and 2010-2018 periods

Site	SO4 p-w mean	precip gauge
ALG	0.00108	0.22246
EGB	0.00108	0.1556
SPR	0.00308	0.74896
LED	0.00244	0.4777
SAT	0.00094	0.0198
LON	0.00028	0.71138
CHA	0.0007	0.20408
FRE	0.00244	0.95216
CPS	0.00076	0.40654

Note that for total N and total oxidized N deposition, there were clear differences in the trends before 2010 (significant decrease) and after 2010 (no trend, indicated by "ns"). For total reduced N deposition, no trends were found before and after 2010.

Line 573: "...) at CAPMoN sites based..."

Response: Revised according to your suggestion. (Table 6 caption)

Line 611: Figure 9 caption: "...of nitrogen at CAPMoN sites, 2000 - 2018."

Response: Revised according to your suggestion.

Line 616: Figure 10 caption: "... of sulfur at CAPMoN sites, 2000 - 2018."

Response: Revised according to your suggestion.

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.

Response: As stated in the beginning of section 3.4, the ratio is the percentage change in deposition of N or S to the percentage change in precursor emissions. The closer the ratio is to 1 (or the percentage is

to 100%), the greater the efficacy of precursor emissions reductions on decreasing N or S deposition. Based on previous studies, a percentage between 80% and 120% suggests good efficacy, whereas a percentage below 80% suggests precursor emissions reductions were ineffective at decreasing N or S deposition. We have added these explanations in section 3.4 (1st paragraph) to clarify the interpretation of the response ratios.

The ratio takes into account the air mass patterns and where the emission sources are likely to be. As described in section 2.3, CAPMoN sites in eastern Canada are strongly influenced by eastern Canada and the eastern U.S. SO_2 and NO_x emissions sources based on previous source attribution analyses using back trajectory modeling (ECCC, 2004; Zhang et al., 2008). Thus, we calculated the ratio as the percentage change in deposition of N and S to the percentage change in precursor emissions for eastern Canada or the eastern U.S. region. For the west coast site (SAT), sources of SO_2 and NO_x emissions are located on both sides of the Canada-U.S. border (ECCC, 2004), but most likely from the western region. Thus, the ratio is calculated as the percentage change in deposition of N and S to the percentage change in precursor emissions for western Canada or the western U.S. region.

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.

Response: The explanation at the end of section 3.4 has been clarified as follows. "Thus, changes in emissions likely result in a more immediate change in dry deposition of S and oxidized N, since dry deposition is more localized. Changes in wet deposition are less regionally coupled with emissions because contaminants are removed from higher in the troposphere and higher up for some intense thunderstorms, thereby including contaminants from long-range transport."

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.

Response: We are referring to critical loads of acidity only. This is now stated in several places in section 3.5. For example, "Aquatic critical loads (CL) of acidity for the lakes around the five stations studied…"; "Five of the 14 stations (ALG, CHA, CPS, LED, SPR) received total N+S deposition in exceedance of their terrestrial CL of acidity at some point in time across the study period…"; "Terrestrial ecosystems in exceedance of their CL of acidity are at risk for decreased soil base saturation, mobilization of root-toxic aluminum ions…"

A brief discussion on addressing critical loads of eutrophication in future work was also included as follows. "It is important to note that only CL of acidity were evaluated in this study. N deposition contributes to eutrophication in N-limited aquatic and terrestrial ecosystems (Smith et al., 1999), as well

as changes to plant communities (Field et al., 2014) including losses of species diversity (Bobbink and Hicks, 2014; Payne et al., 2017). The form of N deposited also has ecosystem health implications. Increased ammonium in the soil results in lower base cation uptake, which can reduce tree growth (Gloser and Gloser, 2001; Bobbink et al., 2011; de Vries et al., 2015). Critical loads of N for eutrophication and biodiversity loss have not been widely established for Canadian ecosystems, but an area of future work is to assess eutrophication effects due to N deposition surrounding the monitoring stations (e.g. Forsius et al., 2021)." (2nd last paragraph before section 4)

Bobbink, R., Braun, S., Nordin, A., Power, S., Schutz, K., Strengbom, J., Weijters, M., and Tomassen, H.: Review and revision of empirical critical loads and dose-response relationships, Editors: Bobbink, R. and Hettelingh, J. –P, Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010, 2011.

Bobbink, R., and Hicks, W. K.: Factors Affecting Nitrogen Deposition Impacts on Biodiversity: An Overview, In M. A. Sutton, K. E. Mason, L. J. Sheppard, H. Sverdrup, R. Haeuber, and W. K. Hicks (Eds.), Nitrogen Deposition, Critical Loads and Biodiversity (pp. 127–138), Springer Netherlands, https://doi.org/10.1007/978-94-007-7939-6_14, 2014.

de Vries, W., Hettlingh, J.-P., and Posch, M.: Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems, *Environ. Pollut.*, 25, Springer Dordrecht, https://doi.org/10.1007/978-94-017-9508-1, 2015.

Field, C. D., Dise, N. B., Payne, R. J., Britton, A. J., Emmett, B. A., Helliwell, R. C., Hughes, S., Jones, L., Lees, S., Leake, J. R., Leith, I. D., Phoenix, G. K., Power, S. A., Sheppard, L. J., Southon, G. E., Stevens, C. J., and Caporn, S. J. M.: The Role of Nitrogen Deposition in Widespread Plant Community Change Across Semi-natural Habitats. Ecosystems, 17(5), 864–877, https://doi.org/10.1007/s10021-014-9765-5, 2014.

Forsius, M., Posch, M., Holmberg, M., Vuorenmaa, J., Kleemola, S., Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., deWit, H.A., Dirnböck, T., Frey, J., Grandin, U., Hakola, H., Kobler, J., Krám, P., Lindroos, A.-J., Löfgren, S., Pecka, T., Rönnback, P., Skotak, K., Szpikowski, J., Ukonmaanaho, L., Valinia, S., Váňa, M.: Assessing critical load exceedances and ecosystem impacts of anthropogenic nitrogen and sulphur deposition at unmanaged forested catchments in Europe, Sci. Total. Environ., 753, https://doi.org/10.1016/j.scitotenv.2020.141791, 2021.

Gloser, V., and Gloser, J.: Nitrogen and base cation uptake in seedlings of Acer pseudoplatanus and Calamagrostis villosa exposed to an acidified environment. In O. Gašparíková, M. Čiamporová, I. Mistrík, & F. Baluška (Eds.), Recent Advances of Plant Root Structure and Function (pp. 103–109), Springer Netherlands, https://doi.org/10.1007/978-94-017-2858-4_12, 2001.

Payne, R. J., Dise, N. B., Field, C. D., Dore, A. J., Caporn, S. J., and Stevens, C. J.: Nitrogen deposition and plant biodiversity: Past, present, and future, Front. Ecol. Environ., 15(8), 431–436. https://doi.org/10.1002/fee.1528, 2017.

Smith, V. H., Tilman, G. D., and Nekola, J. C.: Eutrophication: Impacts of excess nutrient inputs on freshwater, marine, and terrestrial ecosystems, Environ. Pollut., 100(1), 179–196, https://doi.org/10.1016/S0269-7491(99)00091-3, 1999.

Line 660: Specify that these are CLs for acidity. The units should be eq H+ ha-1 yr-1, correct?

Response: Critical loads (CL) of acidity is now stated in the first sentence and in several places in section 3.5. The units of eq ha^{-1} yr^{-1} is also correct. There is no need to include H+, since we have now clarified in the text these are CL of acidity.

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?

Response: Critical loads (CL) of acidity is now stated in the previous sentence and in several places in section 3.5. There is no need to include H+, since we have now clarified in the text these are CL of acidity.

Line 663: "...high CLs for these lakes (Table S9)."

Response: Revised according to your suggestion. (1st paragraph of section 3.5)

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?

Response: The shift in deposition around 2010 is actually a slowdown in the decreasing trend in N deposition (Table 6). Total N deposition (excluding NH_3 and NO_2 deposition) trends after 2010 were not statistically significant at most of the sites; however, the small deposition fluxes after 2010 likely contributed to non-exceedance of critical loads (CL) in Fig. 11. The main reason for not drawing the connection between the deposition and exceedance results is the lack of long-term data on NH_3 and NO_2 dry deposition, which can affect the exceedance if those data were included. As stated in the 2nd last paragraph of section 3.5, the inclusion of NO_2, NH_3, PAN and unknown NO_y dry deposition reported in our previous study (Zhang et al., 2009) resulted in CL exceedance at CHA in 2004 and near exceedance at LED in 2003 (Table S11).

Figure 11 caption (from acp-2022-400-RC2-supplement.pdf): Suggestion: Annual acidic loading from total sulfur and nitrogen deposition to aquatic and terrestrial environs near CAPMoN sites and comparison to critical loads (CL) and indentification of exceedances, 2000 - 2018. The grey area...

Maybe add an explanation / legend to tell the reader that the different lines are for CAPMoN Stations? If one copied and pasted this figure into an email, would the recpient know what ELA and KEJ are?

Response: Please note that the y-axis refers to the magnitude of the exceedance and not the total S and N deposition. The exceedance shown in the plots is a function of the CL. We have revised the caption for Figure 11 and included the CAPMoN site names that are referred to in the plots.

Figure 11: Critical load (CL) exceedances from total sulfur and nitrogen deposition near selected CAPMoN stations at Algoma (ALG), Chalk River (CHA), Chapais (CPS), Experimental Lakes Area (ELA), Lac Edouard (LED), Kejimkujik National Park (KEJ), and Sprucedale (SPR) for lakes (left) and soils (right). Stations with deposition fluxes below CL in every year from 2000-2018 are not shown. The grey area represents non-exceedance.

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?

Response: The CL abbreviation is used throughout section 3.5 after it is defined at the beginning. The unit of eq ha^{-1} yr^{-1} is correct. There is no need to include H+, since we have now clarified in the text these are CL of acidity. We have included interpretations of the CLminN, CLmaxN and CLmaxS in section 3.5 (2nd paragraph) as follows.

"Five of the 14 stations (ALG, CHA, CPS, LED, SPR) received total N+S deposition in exceedance of their terrestrial CL of acidity at some point in time across the study period (Fig. 11). The SPR site represented the highest initial terrestrial CL exceedance (beginning in 2003) and steepest decline into non-exceedance of the five exceeded sites; ALG followed a similar trend. The CPS site was only briefly in exceedance during a period of high deposition in 2000 and 2002. Wet deposition was the main driver of the majority of the acidic deposition, and wet SO_4^{2-} alone resulted in terrestrial CL exceedance at these five stations. Note that there is no unique combination of S and N deposition to achieve or maintain non-exceedance; in some cases, changes to S or N deposition individually could result in a change of exceedance status, while in others, it may be easier to achieve change through varying reductions in both S and N. At four stations that did not experience exceedance over the time period, sensitivity to S deposition alone (that is, very low $CL_{max}S$) was estimated at BAB (208 eq ha^{-1} yr^{-1}) and $CL_{max}S$ values below 500 eq ha^{-1} yr^{-1} were estimated for MIN, CPS, and LED (Table S10). Poor weathering rates characterized the CL at these four sites, which have shallow soils (i.e. 30 cm or less) underlain by acidic parent material. The three southern stations (LON, EGB, FRE) had the highest $CL_{max}S$ (that is, they are tolerant to higher levels of S deposition, up to 2,936 eq ha^{-1} yr^{-1} at LON; Table S10); these stations are situated in areas with deeper soils (up to 50 cm) and alkaline parent material. The BON site showed high estimated tolerance to N deposition ($CL_{max}N$) at 2,589 eq ha^{-1} yr^{-1}, which is likely driven by poorly drained soils (and therefore a high denitrification fraction); other sites ranged between 398 and 1,719 eq ha^{-1} yr^{-1} (Table S10), with BAB being very sensitive to acidifying N (that is, $CL_{max}N$-$CL_{min}N$ is the smallest among the sites). Terrestrial ecosystems in exceedance of their CL of acidity are at risk for decreased soil base saturation, mobilization of root-toxic aluminum ions, and reduced plant growth. While soils may be slower to recover from acidification, the trend into non exceedance by the terrestrial sites is encouraging and consistent with indications that forest soils in North America are gradually recovering (Lawrence et al., 2015; Hazlett et al., 2020)."

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?

Response: Please see response to previous comment and explanations added to the text in section 3.5 (2nd paragraph). In section 2.4, we added a reference to a figure in the Supplement showing the CL function for terrestrial ecosystems with an explanation on how exceedance is calculated.

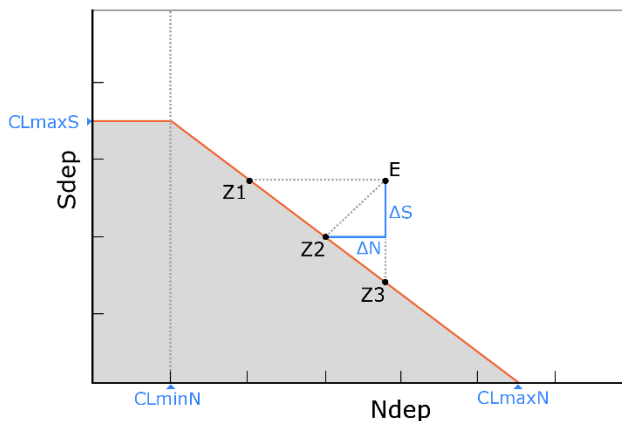



Figure S9: The critical load function for sulfur and nitrogen is defined by the maximum critical load of sulfur when nitrogen deposition (Ndep) is zero (CLmaxS), the maximum critical load of nitrogen when sulfur deposition (Sdep) is zero (CLmaxN), and a minimum critical load of nitrogen (CLminN) above which nitrogen deposition is acidifying. Sites where Sdep and Ndep are below the function line (the grey area) are protected from damage from acid deposition (e.g. not in exceedance). Exceedance (point E) is defined as the shortest path to non-exceedance, as illustrated by the points Z1, Z2, and Z3, but there is no unique path to non-exceedance. For more details see Posch et al. (2015).

Posch, M., de Vries, W., Sverdrup, H. (2015). Mass Balance Models to Derive Critical Loads of Nitrogen and Acidity for Terrestrial and Aquatic Ecosystems. In: de Vries, W., Hettelingh, JP., Posch, M. (eds) Critical Loads and Dynamic Risk Assessments. Environmental Pollution, vol 25. Springer, Dordrecht. https://doi.org/10.1007/978-94-017-9508-1_6

Line 719: "Total S depositions for the respective periods were…"

Response: Revised according to your suggestion. (1st paragraph of section 4)

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?

Response: According to the ACP website, there are no specific criteria on what should be discussed in the Conclusions. In this paper, we summarized the major results, uncertainties and limitations of the study and future work to address the uncertainties and limitations. We have revised the subheading to Conclusions and Future Work if this is permitted by ACP.

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

Response: Figure 11 provides some encouraging results with respect to ecosystem recovery from historical acidification. However, there are some caveats that we thought should be further emphasized in the conclusion. These include: (1) the limited number sites where critical loads exceedance was evaluated, (2) deposition sites were not selected based on their representation of acid-sensitive soils or lakes, (3) the lack of deposition monitoring in western Canada and prairie provinces, (4) the increasing importance of reduced N deposition, etc.

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.

Response: We revised the figure captions in the Supplement so that it is clear that we are referring to CAPMoN sites and the data cover the period from 2000 to 2018.

Section S1.2: Citation for Zhang et al., (2008) is not in the references list.

Response: Reference added to the Supplement.

Zhang, L., Vet, R., Wiebe, A., Mihele, C., Sukloff, B., Chan, E., Moran, M. D., and Iqbal, S., 2008. Characterization of the size-segregated water-soluble inorganic ions at eight Canadian rural sites. *Atmos. Chem. Phys.*, *8*(23), 7133-7151.

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."

Response: Revised according to your suggestion.

Tables S9 and S10 – What are the data sources for the tables? Please include footnotes to reference data sources.

Response: Sources of data for Table S9 are now provided in the table footnotes. Sources of data for Table S10 are discussed in Supplement Section S2 Soil critical loads (CL) calculations and references to the data sources are provided in the reference list.

Table S11: "...) near CAPMoN sites after including..." Also, what are the source(s) of the lake and soil chemistry data? Please footnote those.

Response: Revised according to your suggestion. The sources of lake and soil chemistry data are provided in the caption of Table S11.

Figure S2: One cannot read those tiny green labels.

Response: The font size for green labels have been increased.

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?

Response: Revised according to your suggestion.

Figure S4: Caption: "Annual trends in atmospheric S and N species concentrations (mg/m3) at CAPMoN sites grouped by region."

Response: Revised according to your suggestion.

Figure S5a: This caption is great! This is what you need on every figure!

Response: Thank you!

Figure S6: Caption: "…(kg N or S/ha/yr) measured at CAPMoN sites during 2000-2018…"

Response: Revised according to your suggestion.

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)."

Response: Revised according to your suggestion.

Figure S8: Change caption in the same way as described above for S7.

Response: Revised according to your suggestion.

Supplementary Information References: Add Zhang, 2008 as mentioned above.

Response: Reference added.

Reviewer #3

We appreciate the reviewer's comments on the overall impression of the manuscript and suggestions for further improvements. Our point-by-point responses to each comment are provided below.

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^+.

Response: Reduced N deposition is only referring to NH_4^+ deposition in the paper. We have made that clear in the revised paper. For example, "Trends in reduced N (NH_4^+ deposition) were not statistically significant…" We also listed the N species in the legend of Figures 6 and 7 and in Table 6 in order to clarify which species are included in oxidized and reduced N deposition.

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.

Response: A sentence was added in section 2.4 (1st paragraph) to mention the underestimate in total N deposition as follows. "This assumption is a worst-case scenario and may result in higher CL exceedance. However, the higher exceedance may be offset by an underestimate of total N deposition due to missing NH_3 dry deposition."

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.

Response: The Atlantic region of Canada comprises the provinces of Nova Scotia (KEJ site), New Brunswick, Prince Edward Island and Newfoundland and Labrador (BAB site). The MIN site is not part of Atlantic Canada, but as shown in Figure 1, this site is located much closer to the Atlantic region than it is to other regions we have defined. Thus, KEJ, BAB and MIN are classified as Atlantic sites based on their geographical location. The more recent data at BAB and MIN might have reduced the mean dry deposition flux in the Atlantic region; however, we cannot confirm how much it is reduced (and whether it is significant) without the earlier data.

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.

Response: The comparison between Canadian and U.S. values is now restricted to total S deposition fluxes. The sentence in section 3.3.1 (1st paragraph) reads as follows. "Total S deposition flux in Canada was lower than that in the continental U.S. over a similar timeframe (2002-2017), namely 1-11 kg S ha^{-1} yr^{-1} (Benish et al., 2022)."

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.

Response: Prairie is incorrect in this sentence. It should read, "During 2014-2018, reduced N deposition surpassed oxidized N deposition in the southeastern region; however, oxidized N deposition continued to be greater than reduced N deposition at the west coast site and in the Atlantic region." (paragraph before Table 5)

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^+.

Response: Oxidized N and reduced N species are now listed in the legend of Figure 6. The caption in Figure 7 has also been updated as follows. "Figure 7: Percentage of wet and dry annual deposition of (a) nitrogen and (b) sulfur at CAPMoN sites during 2000-2018. %ddep TN includes dry deposition of oxidized

N (HNO_3, pNO_3^-) and reduced N (pNH_4^+). Mean percentage (circles); range or annual variability in the percentage (error bars); mean annual precipitation amount (diamonds)."

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

Response: The sentence now reads, "Trends in reduced N (NH_4^+ deposition) were not statistically significant at most sites except for the slight decreasing trends at ALG, CHA and CPS." (1st paragraph of section 3.3.4)

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

Response: The sentence now reads, "Trends in reduced N (NH_4^+) deposition were not statistically significant in either period. Decreasing trends in total deposition of S were statistically significant in the periods before and after 2010." (paragraph before Table 6)

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.

Response: The N species are now listed in the table heading for Total oxidized N and Total reduced N.

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"

Response: Revised according to your suggestion. (1st paragraph of section 2.1)

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."

Response: Revised according to your suggestion. (paragraph before section 2.2)

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?

Response: There was a study conducted in the Athabasca oil sands region that showed oil sands emissions contributed to critical loads exceedances in Alberta and Saskatchewan. We discussed this study and included some future research implications as follows. "SO_2 and NO_x emissions in North Dakota, U.S., which borders Saskatchewan to the south, have seen a dramatic increase between the early 2000s and 2014-2018 due to the Bakken oil and gas development (USEPA, 2021). Further, emissions from the Canadian Athabasca oil sands region have contributed to exceedances of terrestrial and aquatic critical loads, which suggest potential ecosystem damage (Makar et al., 2018). In response to these anthropogenic activities, additional CAPMoN and provincial monitoring sites have started

operating in this region in recent years. 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)." (paragraph before section 3.2)

Makar, P. A., Akingunola, A., Aherne, J., Cole, A. S., Aklilu, Y.-A., Zhang, J., Wong, I., Hayden, K., Li, S.-M., Kirk, J., Scott, K., Moran, M. D., Robichaud, A., Cathcart, H., Baratzedah, P., Pabla, B., Cheung, P., Zheng, Q., and Jeffries, D. S., 2018. Estimates of exceedances of critical loads for acidifying deposition in Alberta and Saskatchewan. *Atmos. Chem. Phys.*, 18, 9897–9927.

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…"

Response: Revised according to your suggestion. (paragraph after Table 2)

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.

Response: The sentence states, "Fluxes of HNO_3 in the Atlantic region and remote sites (0.05-0.2 kg N ha^{-1} yr^{-1}) were among the lowest in Canada." The flux ranges in Table 1 for the Atlantic region and remote sites support this statement. If we include only KEJ (0.2 kgN ha^{-1} yr^{-1}), this statement is also true.

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

Response: We clarified in the caption and legend of Figure 2 the species that are in the gas or aerosol phase. The revised caption reads, "Figure 2: Dry N and S deposition fluxes (kg N or S ha^{-1} yr^{-1}) and the percentage of N or S species in dry deposition for the 5-year period in 2000-2004 and in 2014-2018 at CAPMoN sites. For each site, the first bar represents the 2000-2004 values and the second bar represents the 2014-2018 values. Gaseous species: HNO_3 and SO_2; particulate (p) species: pNO_3, pNH_4 and pSO_4."

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.

Response: All 15 sites are discussed in the text. Rather than focusing on individual sites, we combined the results from several sites to discuss regional patterns in N and S deposition. We prefer to include all the sites in the main paper as there is a lot of information currently in the Supplement. The size of the green text in Figures 4 and 5 have been increased in the revised paper for greater legibility.

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?

Response: The decrease in annual dry S deposition at SAT was driven by the decrease in SO_2 dry deposition (Table 4), which in turn is due to a significant decrease in ambient SO_2. A recent study of air quality in Canadian port cities suggest that most of the reductions in ambient SO_2 in the port of Vancouver/Victoria (Canadian west coast) and other ports had been attributed to new regulations requiring the use of low-sulfur fuel in marine transportation in Canadian waters (Anastasopolos et al., 2021). Since SAT is a west coast site, the findings in Anastasopolos et al. likely apply to this site. The findings are also consistent with the significant drop in British Columbia's SO_2 emissions from 2015 onwards (Fig. S5b), which was mainly attributed to the transportation and mobile equipment sector. Similarly, the decrease in annual dry S deposition at ELA was driven by the decrease in SO_2 dry deposition. However, unlike SAT, the rate of decline in SO_2 dry deposition was much smaller at ELA (Table 4). A more focused study on this particular site is necessary to understand what is driving the slight decrease in ambient SO_2 after 2010.

We have added the following explanation. "Interestingly, the rate of decline in annual dry S deposition accelerated after 2010 at SAT and ELA (Table 4). The decline at SAT is likely associated with reductions in ambient SO_2 in the port of Vancouver and Victoria (Canadian west coast), which was driven by the implementation of new regulations requiring the use of low-sulfur fuel in marine transportation in Canadian waters (Anastasopolos et al., 2021)." (paragraph before Table 4)

Anastasopolos, A.T., Sofowote, U.M., Hopke, P.K., Rouleau, M., Shin, T., Dheri, A., Peng, H., Kulka, R., Gibson, M.D., Farah, P.M. and Sundar, N., 2021. Air quality in Canadian port cities after regulation of low-sulphur marine fuel in the North American Emissions Control Area. Science of The Total Environment, 791, https://doi.org/10.1016/j.scitotenv.2021.147949

Table 4: Nice, very clear table.

Response: Thank you!

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.

Response: For the figures containing bar graphs (e.g. Fig. 6), the sites are listed in order longitudinally from west (left) to east (right) in order to be consistent with the regional pattern discussions in the text. We added the description of the site order in the figure captions.

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

Response: The total N or total S deposition fluxes do not appear to differ substantially between the cold and warm seasons. The seasonal effect on terrestrial acidification is likely not as important as the year over year effect of annual deposition on acidification, which is presented in section 3.5.

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.

Response: The annual trends discussed in this section cover the entire 2000-2018 period. There were sufficient years of data at BON, MIN and BAB during this period. The lack of significance is a result of the flat trends rather than the lack of data. We clarified that the trends discussed here refer to the 2000-2018 period. The sentence now reads, "Annual trends were not significant at BON, MIN and BAB (p>0.05) during the 2000-2018 period." (1st paragraph of section 3.3.4)

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).

Response: The sentence has been restated as follows. "It is very likely that the increased dry deposition of NH_3 could compensate for the decreased dry deposition of pNH_4^+ considering the much higher (by 5 to 10 times) V_d of NH_3 than pNH_4^+; however, additional long-term measurements of ambient NH_3 would be required in order to verify this (e.g., via ongoing remote sensing and passive measurements)." (2nd paragraph of section 3.3.4)

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.

Response: The trend in the proportion of total deposition of N and S from wet deposition may change if NH_3 dry deposition were included. Based on data at the EGB site in 2002 (Zhang et al., 2009) and 2010 (this study), the proportion of N wet deposition decreased from 62% in 2002 to 53% in 2010 when NH_3 dry deposition was included. This translates to an annual decrease of ~1% in the percentage of N wet deposition. It is important to note that the EGB site is near agricultural areas, which is a significant source of ambient NH_3. Ambient NH_3 at other rural non-agricultural sites may be lower. However, we do not have long-term ambient NH_3 data at other sites to quantify the impact. We have added a caveat that "the trend in the percentage of wet N deposition may change if NH_3 dry deposition were included. An increasing trend in the percentage of dry N deposition is expected at sites with high ambient NH_3." (paragraph before Figure 9)

Regarding lines 607-608, we have clearly stated that "The rise in the percentage of reduced N is a conservative estimate since NH3 dry deposition was not included." (paragraph before Figure 9)

Zhang, L., Vet, R., O'Brien, J. M., Mihele, C., Liang, Z., and Wiebe, A., 2009. Dry deposition of individual nitrogen species at eight Canadian rural sites. *J. Geophys. Res. Atmos.*, 114, D02301, doi:10.1029/2008JD010640.

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.

Response: The species are not as clear on the bar graphs when using the same color scheme especially for the lighter tones. The use of different colors for different species makes it easier to see the differences.

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

Response: References to the back trajectory analyses for CAPMoN sites have been included.

Environment and Climate Change Canada (ECCC) (2004), Meteorological Services of Canada. 2004 Canadian Acid Deposition Science Assessment. ISBN 0-662-38754-6. Available at: www.publications.gc.ca/pub?id=9.688243&sl=0

Zhang, L., Vet, R., Wiebe, A., Mihele, C., Sukloff, B., Chan, E., Moran, M. D., and Iqbal, S., 2008. Characterization of the size-segregated water-soluble inorganic ions at eight Canadian rural sites. *Atmospheric Chemistry and Physics*, *8*(23), 7133-7151.

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?

Response: As discussed in the paper, "The lack of trend in wet NH_4^+ deposition at Canadian rural sites was consistent with NH_3 emissions in eastern Canada (a slight decreasing trend) and the eastern U.S (no trend) (Fig. S5a)." If we included NH_3 dry deposition, reduced N deposition would likely increase at agricultural sites. This is because we observed an increase in ambient NH_3 at the EGB site from 2002 to 2010, which means an increase in NH_3 dry deposition. Thus, the reduced N deposition response is likely more than 100% for agricultural sites. It is important to note that the NH_3 emissions in Fig. S5a and in the response ratios are mostly from anthropogenic sources. Climate driven effects, such as the increased volatilization of NH_3 due to increasing temperatures (Sutton et al., 2013), are not considered in the emissions.

The nitrate and sulfate deposition response to NO_x and SO_2 emissions reductions were close to ideal in the USA, UK and Canada as discussed in section 3.4. NO_x and SO_2 emissions have been decreasing in Canada and the U.S. as well as in the UK (1986-2001). There are definitely some process-based drivers that can have an effect on the nitrate, reduced N, and sulfate deposition response, such as variability in precipitation amounts, prevailing air mass patterns and aerosol chemistry. Long-range transport and precipitation variability contributes to some variability in the response (Tan et al., 2020). Similarly, the amount of time that air masses had travelled over polluted areas which arrived at the deposition monitoring site and prevailing atmospheric circulation patterns that promotes precipitation scavenging in certain regions. Changes in the relative amounts of ambient SO_2, NOx and NH_3 affect the gas-aerosol chemistry, which in turn affect dry and wet N and S deposition (Fowler et al., 2005). NH_3 dry deposition

flux is also highly sensitive to the bidirectional exchange of gaseous NH_3 (Tan et al., 2020). The discussion in section 3.4 (2nd and 3rd paragraphs) have been updated to take into account the above points.

Fowler, D., Smith, R. I., Muller, J. B. A., Hayman, G., and Vincent, K. J., 2005. Changes in the atmospheric deposition of acidifying compounds in the UK between 1986 and 2001. *Environmental Pollution*, *137*(1), 15-25.

Sutton, M. A., et al.., 2013. Towards a climate-dependent paradigm of ammonia emission and deposition, Philos. Trans. R. Soc. B. Biol. Sci., 368, 20130166, doi:10.1098/rstb.2013.0166.

Tan, J., Fu, J. S., and Seinfeld, J. H., 2020. Ammonia emission abatement does not fully control reduced forms of nitrogen deposition. *Proc. Natl.l Acad. Sci.*, *117*(18), 9771-9775.

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:

Response: We added a sentence in section 3.5 (3rd paragraph) related to the uncertainties in the atmospheric measurements and how this may affect the assessment of CL exceedance. The sentence reads, "This suggests that the dry deposition of other N compounds cannot be neglected at sites where the difference between the CL and total deposition from routinely-monitored N is small. Further, uncertainties in the air and precipitation monitoring data also contribute to uncertainties in dry and wet deposition fluxes, which may affect the assessment of CL exceedance especially at locations close to critical thresholds (Walker et al., 2019b).

Walker, J. T., Bell, M. D., Schwede, D., Cole, A., Beachley, G., Lear, G., and Wu, Z., 2019b. Aspects of uncertainty in total reactive nitrogen deposition estimates for North American critical load applications. *Science of the Total Environment*, *690*, 1005-1018.

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.

Response: Figure 11 shows exceedance of CL of acidity in 2014 at SPR, but it does not appear significant as stated in the sentence "no significant exceedances of aquatic and terrestrial critical loads were observed after 2012…"

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

Response: We meant to say new sites that have been added to CAPMoN, which was already stated in the previous sentence. The sentence has been deleted to avoid repetition (last paragraph of section 4).

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.

Response: We included the following in the last paragraph of section 4. "It should be noted that organic N is not included in the total N deposition budget, which should be considered in future monitoring and analysis studies. This can be done by measuring dissolved organic N in precipitation and total NO_y in air, with the former for estimating organic N wet deposition and the latter for approximating organic N dry deposition (Walker et al. 2020). In terms of deposition model improvements, one important area is constraining the air-surface exchange estimates for reactive N by incorporating measurements from process-level studies and integrating spatial and time specific meteorological data to improve NH_3 emission estimates (Sutton et al., 2013; Walker et al., 2020)."

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).

Response: Revised according to your suggestion. (paragraph before section 2.2)

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

Response: The sentence was restated as follows. "Concentrations of N and S at the west coast (SAT) and prairie sites (BRA) were similar to those of the greater southeastern region, except for the lower pSO_4^{2-} and pNH_4^+ concentrations at the west coast and prairie sites." (1st paragraph of section 3.1)

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

Response: Revised according to your suggestion.

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

Response: Revised according to your suggestion.

Reviewer #4

We appreciate the reviewer's comments on the overall impression of the manuscript and suggestions for further improvements. Our point-by-point responses to each comment are provided below.

This study presents an analysis of sulfur and nitrogen deposition monitored at 15 rural CAPMoN sites for the 2000-2018 time-period to characterize the spatiotemporal trends in atmospheric deposition across Canada and assess likely changes in ecosystem damage through inferences in changes in critical load exceedance during the period analyzed. The changes in atmospheric S and N deposition and the relative proportions of wet and dry deposition amounts reported by this analysis are along expected lines given the changes in NOx/SOx/NH3 emissions and those reported in several similar analyses (both measurement and model based) for atmospheric deposition across the Continental U.S. Nevertheless, the documentation of these changes across Canada and from the CAPMoN measurements is a useful contribution. Before the manuscript can be considered suitable for publication, I feel there are aspects of the analysis that would benefit from additional elaboration as well as several conclusions that need more substantiation. The following comments are offered, addressing which may help improve the usefulness of this manuscript.

From a total N deposition perspective, one of the shortcomings of this analysis relates to the limitations in the estimates of dry deposition amounts: (1) ambient measurements of select N species are combined with a deposition model to estimate their dry-deposition fluxes; (2) air sampling at majority of the locations only report HNO3, pNO3, and pNH4. The lack of measurements of NO2 (possibly) and NH3 (likely) represent a potentially large proportion of the dry deposited N and total atmospheric deposition budget. This largely renders the comparisons of wet vs dry N deposition amounts somewhat qualitative and confounds any quantitative interpretation of the relative contribution of oxidized vs reduced N to the total atmospheric N deposition amounts and their influence on subsequent CL exceedance and changes. To the authors credit, they do present the possible impact of these missing pieces through analyses of a more complete measurement set at the EGB site, which suggests non-negligible impacts in terms of overall budget. It is not readily apparent whether the relative contributions of the missing components inferred from the EGB site could be extrapolated to the other locations, but perhaps a more explicit recognition of the missing N-deposition components in the discussion of results at other locations may help caveat the results more appropriately. As examples:

Response: Regarding point (1), the inferential method has been typically used for long-term and/or large-scale dry deposition estimation due to the difficulties in directly measuring dry deposition fluxes. We have referenced many studies in the paper (e.g., Holland et al., 2005; Zhang et al., 2003, 2009; Flechard et al., 2011; Li et al., 2016; Schwede et al., 2011; Staelens et al., 2012; Zhang et al., 2018; Tan et al., 2020) to show that the inferential method is a common approach taken to estimate dry deposition fluxes. Regarding point (2), we acknowledged the importance of other N species, such as NH_3, NO_2 and unknown NOy species, that are not routinely measured by CAPMoN and discussed the need for expanded measurements throughout the paper. For example, we stated "dry deposition of NH_3 and some oxidized N species were not included due to a lack of data." "Gaseous NH_3, NO_2, PAN, PPN and other NO_y can also contribute to N dry deposition." "given NH_3 was by far the largest contributor to N

dry deposition for this one-year period at EGB, including NH_3 in routine monitoring should be a priority…" "Ambient NH_3 monitoring would be required to confirm the future trends in N dry deposition." "…additional long-term measurements of ambient NH_3 would be required in order to verify this (e.g., via ongoing remote sensing and passive measurements)." "This suggests that the dry deposition of other N compounds cannot be neglected at sites where the difference between the CL and total deposition from routinely-monitored N is small." "a greater focus is needed to track ammonia emissions and reduced N deposition trends." These examples are explicit recognitions of the missing N-deposition components.

Our response to specific comments below addresses potential impacts on the results had we included NH_3 dry deposition in the analysis. Note that the discussion is speculative because additional N measurements were only available at one site (EGB) for one year (2010). We also included findings from Zhang et al. (2009), where short-term additional N measurements were conducted at a greater number of CAPMoN sites during the 2002-2005 period. Note that earlier measurements of ambient NH_3 and NO_2 likely do not reflect measurements in the recent period because of significant NO_x and SO_2 emissions reductions and changes in NH_3 emissions over time.

L286: would dry deposition of N decrease by 50% if the estimates also included changes induced by NH3 emissions?

Response: Based on the additional N measurements at EGB in 2002 (Zhang et al., 2009) and in 2010 (this study), N dry deposition decreased from 3.9 kg N/ha in 2002 to 3.6 kg N/ha in 2010, which represents an 8% reduction at this site. If this trend is extrapolated to 2018, the predicted N dry deposition in 2018 would be about 3.4 kg N/ha (~ 12% reduction). Similar trends are expected for LON and BRA sites as they are also near agricultural areas (Table S1), where ambient NH_3 is strongly influenced by agricultural activities. However, it is likely not applicable to other CAPMoN sites because they are farther from agricultural sources; the trend in N dry deposition would be less affected. One exception is the impact of wildfire driven NH3 emissions on the west coast site. It is also important to note NH3 dry deposition estimates were likely overestimated because bidirectional flux has not been considered. Therefore, we can state that the average percentage decrease in N dry deposition was smaller than 50% if NH_3 was included, but the magnitude of the decrease is still uncertain. We added the following sentence. "The percentage decrease in N dry deposition over the two periods was likely below 50% if changes in NH_3 and NO_2 dry deposition were included (see detailed discussion below)." (paragraph after Table 1)

Zhang, L., Vet, R., O'Brien, J. M., Mihele, C., Liang, Z., and Wiebe, A., 2009. Dry deposition of individual nitrogen species at eight Canadian rural sites. *J. Geophys. Res. Atmos.*, 114, D02301, doi:10.1029/2008JD010640.

L303-304: conceivably these contributions of HNO3, pNH4 and pNH3 to dry N deposition would be different if NH3 dry deposition was included. The particulate bound N dry deposition contributions could in fact be much smaller?

Response: Relative contributions of HNO_3, pNO_3^- and pNH_4^+ to N dry deposition at the EGB site based only on the routinely-monitored N species (no NH_3, NO_2 and unknown NO_y) during 2000-2018 were 46%,

21% and 33%, respectively. Based on additional N measurements at EGB in 2002 (Zhang et al., 2009) and in 2010 (this study), the average relative contributions of HNO_3, pNO_3^- and pNH_4^+ to N dry deposition were 18%, 6% and 12%, respectively. During 2002-2005, the corresponding percentages at non-agricultural sites were 29%, 1% and 16% (Zhang et al., 2009). If we considered all the sites, relative contributions of HNO_3, pNO_3^- and pNH_4^+ to dry N deposition were likely smaller if the dry deposition of other N species were included. But the magnitude is uncertain because of spatial gradients in ambient NH3 and bidirectional exchange (see above response). Also, NO_2 dry deposition at non-agricultural sites likely decreased in the recent period because of NOx emissions reductions, which means the proportion of other N species would increase. We added a sentence to address this comment. "As discussed below, relative contributions of HNO_3, pNH_4^+ and pNO_3^- to N dry deposition would be smaller if NH_3 and other N species were included in the dry deposition budget." (paragraph after Table 2)

L306: It is possible that pNO3 decreased in response to HNO3 decreases because there was sufficient NH3 available. The relative amounts of oxidized N to total N deposition could in fact be different if the atmospheric NHx budget increased and that became a larger fraction of the N dry deposition. Should probably caveat this discussion by this important missing component.

Response: At EGB, average proportions of oxidized and reduced N in dry deposition based only on the routinely-monitored N species during 2000-2018 were 67% and 33%, respectively. Based on additional N measurements at EGB in 2002 (Zhang et al., 2009) and in 2010 (this study), the corresponding percentages were 52% and 48%, respectively. If NH_3 were considered in the N dry deposition budget, the proportion of oxidized N in dry deposition would be smaller. But the magnitude is uncertain because of spatial gradients in ambient NH_3 and bidirectional exchange (see above response). We added a caveat similar to that of the previous response. "As discussed below, relative contributions of HNO_3, pNH_4^+ and pNO_3^- to N dry deposition would be smaller if NH_3 and other N species were included in the dry deposition budget. Similarly, the proportion of oxidized N dry deposition was likely smaller with NH_3 dry deposition considered." (paragraph after Table 2)

L305: how representative is the fraction of non-routinely monitored N-species to the dry deposition budget (70%) at EGB of other locations?

Response: In our previous study which was conducted during 2002-2005, the percentage of non-routinely monitored N-species in dry deposition were comparable between agricultural (58%) and non-agricultural sites (53%) (Zhang et al., 2009), though these short-term deployments did not always capture the growing season. In this study, we showed that this percentage was 70% of the N dry deposition budget at EGB (agricultural site) in 2010. The findings at EGB can be extrapolated to the LON site, which is another agricultural site in the same region (Table S1). However, this percentage is not expected at all other locations because NH3 dry deposition is smaller at non-agricultural sites. Thus, the percentage of non-routinely monitored N-species in dry deposition is predicted to be below 70%. At the BRA site, oil and gas emissions in the region may contribute to higher NO_2 dry deposition, thereby the fraction of non-routinely monitored N-species to the dry deposition could be even higher than at EGB.

The following explanation is provided in the paragraph after Figure 2. "Based on additional N measurements at EGB in 2010, the relative dry deposition of routinely versus non-routinely monitored N species was 30% and 70%, respectively, on an annual basis. The findings at EGB can be extrapolated to the LON site, which is another agricultural site in the same region (Table S1). BRA and FRE are also in agricultural regions and may have comparable contributions from NH_3. The percentage of non-routinely monitored N species in dry deposition for non-agricultural sites is expected to be somewhat smaller because of the lower ambient NH3 with the exception of wildfire-impacted regions. At the BRA site, oil and gas emissions in the region may contribute to higher NO_2 dry deposition, thereby the fraction of non-routinely monitored N-species to the dry deposition could be even higher than at EGB."

L476-479: would the oxidized N deposition be greater than that of reduced N if NH3 dry deposition estimates were also available? Perhaps this conclusion should be caveated since a potentially large contributor to the reduced N deposition is not accounted for. It is possible that some of the sites in Table 5 are not in proximity to NH3 sources so that NH3 dry deposition may not influence the conclusion – if that is the case it would be helpful to state that more explicitly.

Response: Yes, in the early 2000s (in which this sentence is referring to), total oxidized N deposition was greater than that of total reduced N if NH_3 dry deposition was included. Additional N measurements collected during 2002-2005 at a greater number of CAPMoN sites showed that total oxidized N deposition was greater than total reduced N deposition by a factor of 1.3 to 1.7 for most of the sites (Fig. 7 in Zhang et al. 2009).   Total oxidized N deposition was nearly equivalent to reduced N deposition at the BRA/BRL site in the early 2000s.

L551: could the lack of trend in total deposition of reduced N be due to missing dry deposition of NH3?

Response: The 2nd paragraph in section 3.3.4 is specifically referring to total deposition of reduced N (excluding dry deposition of NH3); this was clearly stated in the first sentence. To clarify further, we changed total reduced N to total NH4+ deposition in the text. "The lack of trends in total NH_4^+ deposition can be explained as follows. First, total NH_4^+ deposition was dominated by the wet deposition of NH_4^+, which exhibited a lack of trend in the eastern U.S. and eastern Canada (Feng et al., 2021). Second,…"

L70: the large variations in the contributions of dry deposition to total N-deposition across the U.S. convey significant spatial variability that likely arises from a combination of effects associated with spatial heterogeneity in emission source distribution, land-use and precipitation – it may be useful to state that explicitly.

Response: The explanations are now stated in the Introduction (2nd last paragraph).

L99: Is there a formal way to assess whether a site is "regionally representative" or is it just based on the rural settings of the site?

Response: Regionally representative CAPMoN sites were assessed using the criteria in Olsen et al. (1990). A site is considered regionally representative if it is not strongly affected by local pollution

sources, including local towns or suburban areas; major SO_2 or NO_x emission sources; nearby surface storage facilities, transportation, agricultural activities, buildings, trees and dusty areas. We clarified the meaning of regionally representative sites and added the reference in the 1st paragraph of section 2.1 as follows. "All sites are considered regionally representative, following the criteria in Olsen et al. (1990), meaning that local pollution sources do not have an outsized influence."

Olsen, A.R., Voldner, E.C., Bigelow, D.S., Chan, W.H., Clark, T.L., Lusis, M.A., Misra, P.K. and Vet, R.J., 1990. Unified wet deposition data summaries for North America: data summary procedures and results for 1980–1986. Atmospheric Environment. Part A. General Topics, 24(3), 661-672.

Equations 1-5: while a reference to the work of Zhang et al. (2008) is provided for the basis of these equations, for the sake of completeness it may be useful to briefly state (if possible) the assumptions on aerosol mixing state (if any) used in this formulation. It would also be useful to state what size fraction pSO4, pNO3, and pNH4 represent – I assume it is PM10?

Response: No information on aerosol mixing state is available. Mixing state is not important for calculating Vd, since Vd is a function of the particle size distribution. The formulas are simply based on fine/coarse ($PM_{2.5}$/$PM_{2.5-10}$) mass fraction measured at multiple CAPMoN sites and seasons, as presented in Zhang et al. (2008). Although there is no size cutoff for CAPMoN particulate ion measurements, pSO_4^{2-}, pNO_3^-, and pNH_4^+ mass are assumed to be mostly in PM_{10}. This is now clarified before equations 1-5.

L255: Do the west coast sites also see influence of intercontinental transport of airborne S and N species and their subsequent contributions to deposition? Could such contributions also lead to the noted weaker trends despite reduction in local emissions?

Response: The west coast site is also affected by occasional long range transport from Asia. McKendry et al. (2008) observed episodes of elevated sulfate and crustal dust at another Canadian west coast site (Whistler, British Columbia), which is attributed to long range transport from Asia. At the west coast site (SAT), sulfate dry deposition was much smaller relative to SO_2 dry deposition and sulfate wet deposition (Figure 10) likely because of the small dry deposition velocity of sulfate. It is possible that the enhancement of sulfate contributed to sulfate wet deposition at the west coast site. Increasing contributions from intercontinental transport (McKendry et al., 2008) could lead to weaker trends in airborne S and N at the west coast site. We have added some explanations, which read "Weaker trends in atmospheric S and N at the west coast and prairie locations reflected the emission trends and levels in western Canada and the western U.S. The Canadian west coast is also affected by episodes of elevated sulfate and crustal dust associated with long range transport from Asia (McKendry et al., 2008). The increasing contributions from intercontinental transport could result in weaker trends in airborne S and N at the west coast site." (paragraph before section 3.2)

McKendry, I. G., Macdonald, A. M., Leaitch, W. R., van Donkelaar, A., Zhang, Q., Duck, T., and Martin, R. V.: Trans-Pacific dust events observed at Whistler, British Columbia during INTEX-B, Atmos. Chem. Phys., 8, 6297–6307, 2008.

L255-265: Are trends in seasonal values similar to those shown for the annual estimates?

Response: Trends in cold and warm season concentrations of airborne N and S species were similar to those of the annual trends. For most of the species, correlation coefficients between seasonal and annual concentrations were above 0.8-0.9. There were a few exceptions. Correlations between warm season concentrations and annual concentrations were lower at the prairie site for all chemical species. Correlations between warm season concentrations and annual concentrations of nitrate were also lower at remote sites and Atlantic sites.

L272-274: I was not sure what the physical significance of providing mean values (across all sites) of the deposition fluxes is?

Response: In addition to providing mean values across all sites, the range of the values was also provided to describe the spatial variability. The sentence in section 3.2.1 (1st paragraph) states, "For all 15 sites, the mean dry deposition was 0.6 kg N ha^{-1} yr^{-1} and 0.9 kg S ha^{-1} yr^{-1} (non-sea salt), where N ranged from 0.1 (MIN) to 1.9 (LON) kg N ha^{-1} yr^{-1}, and S ranged from 0.2 (MIN) to 3.5 (LON) kg S ha^{-1} yr^{-1} (Table 1 and Table S3)."

Figure S3: please clarify in figure caption (here and elsewhere) whether the units are for ug-N/m3 and ug-S/m3.

Response: The units for the airborne concentrations of the trace gases and particulate ions are μg of ion/m^3. We have added these to the captions in Figure S3 and S4.

L34-342: It would be helpful if the authors could explain why the relative proportion of pSO4 dry deposition increased?

Response: The significant decrease in SO$_2$ dry deposition over time led to an increase in the proportion of pSO4 dry deposition. An explanation was already provided before section 3.2.2 as follows. "The relative dry deposition of pSO$_4^{2-}$ increased over the years to 11-55% and exceeded that of SO$_2$ at five sites in the 2014-2018 period (Fig. 2). Feng et al. (2020) showed that the relative fraction of pSO$_4^{2-}$ to total S in ambient air rose by 50.6% as SO$_2$ declined in the eastern U.S. and eastern Canada for the 1989-2016 period."

L415: what is the significance of comparing the annual rate of decrease in S and N dry deposition? Should also probably qualify this statement to indicate that the rate of decrease is expressed as percent.

Response: The significance of comparing the trends in S and N dry deposition is to provide insight on which key component of acid deposition is decreasing faster. This has implications on pollution control strategies because S and N are not affected by the same sources. For example, as shown in Figure S5a, SOx emissions in eastern Canada are mainly from ore and mineral production, whereas NOx emissions in eastern Canada are mainly from transportation and mobile equipment. We reported both the absolute annual rate of decrease as well as the percentage decrease in section 3.2.3 (3rd paragraph). It now reads, "The rates of decrease varied from -0.016 (BON) to -0.35 (LON) kg S ha^{-1} yr^{-1} (percentage decreases of 4.2% to -7.1% per year; Fig. S6 and Table S4)."

L470: I agree that quantifying the contribution of NO2 and NH3 dry deposition is important and that the current suite of measurements do not allow for their quantification. What other methods may be available to fill these critical data gaps? Can modeled deposition estimates of different species be used to fill in the gaps in the interim – are such estimates available from models such as GEM-MACH?

Response: The gaps in monitoring can be filled using estimates of NO_2, NH_3 and other N species from models, such as ECCC's GEM-MACH model, though further model validation needs to be conducted before using the model output for dry deposition calculations. Model-measurement fusion of airborne concentrations of N and S species can improve estimates of dry deposition fluxes on a regional scale if there is sufficient monitoring data available (beyond CAPMoN stations). NH_3 dry deposition estimates across North America for one season in 2013 have been produced using satellite-generated concentration data and dry deposition velocity data from the same model as the one used in this study (Kharol et al., 2018). There are currently some NH3 passive sampling data at selected CAPMoN sites as well. Once the long-term data are made available, a more detailed analysis of dry and total N deposition can be conducted.

We have discussed these methods as follows. "The importance of agricultural NH_3 to local and downwind total N deposition is evident (Warner et al., 2017; Walker et al., 2019a; Hu et al., 2021; Pan et al., 2021); however, quantifying its role remains elusive in natural lands in Canada because of limited measurements. Satellite generated surface-level air concentration data of some important nitrogen species (e.g., NH_3 and NO_2), once available for multiple years, could be potentially useful in filling the data gaps described in this study (Kharol et al., 2018). Passive NH_3 measurements, which are part of the National Atmospheric Deposition Program's Ammonia Monitoring Network, are also ongoing at selected CAPMoN sites. Estimates of NO_2, NH_3 and other N species from chemical transport models and model-measurement fusion products are suitable for quantifying total N deposition over broader scales beyond CAPMoN stations." (paragraph after Figure 6)

Kharol, S. K., Shephard, M. W., McLinden, C. A., Zhang, L., Sioris, C. E., O'Brien, J. M., Vet, R., Cady-Pereira, K. E., Hare, E., Siemons, J. and Krotkov, N. A., 2018. Dry deposition of reactive nitrogen from satellite observations of ammonia and nitrogen dioxide over North America. *Geophys. Res. Lett.*, *45*(2), 1157-1166.

L474: What does comparing the deposition fluxes of N and S tell us? Is there an ecological exposure significance of the relative mass fluxes of S and N?

Response: In terrestrial ecosystems, critical loads (CL) exceedance is derived from a critical loads function (Figure S9 in the revised supplement). There is no unique combination of S and N deposition to achieve or maintain non-exceedance of terrestrial CL. In some cases, changes to S or N deposition individually could result in a change of exceedance status in soils, while in others, it may be easier to achieve change through varying reductions in both S and N. In this study, we found that wet SO_4^{2-} deposition alone resulted in terrestrial CL exceedance at five stations in the early 2000s (see section 3.5), suggesting wet SO_4^{2-} deposition was the major driver of ecosystem damage in the earlier period.

L638-640: Please elaborate why emission reductions were more effective in decreasing dry N deposition than wet N deposition? Some discussion of possible reasons leading to the noted "super-linear" (>100%) response in dry N deposition to emission reductions in the southeastern US should also be provided. Do contributions from transboundary transport play a role here? If so, which species would cause such behavior since HNO3 deposits readily and would not be expected to undergo long-range transport?

Response: We made several clarifications in section 3.4 of the revised paper. The following explanation was added in the 1st paragraph to clarify the interpretation of the response ratio. "Based on previous studies, the closer the ratio is to 1 (or the percentage is to 100%), the greater the efficacy of precursor emissions reductions on decreasing N or S deposition. A percentage between 80% and 120% suggests good efficacy, whereas a percentage below 80% suggests precursor emissions reductions were ineffective at decreasing N or S deposition (Tan et al., 2020)." Thus, the response in dry N deposition for southeastern Canadian sites of 103-106% is an ideal response but it is not super linear. Note in this sentence the response in deposition is relative to NOx emissions decrease in eastern Canada and the eastern U.S., the latter mostly includes northeastern and mid-Atlantic U.S. and some states in the Midwest (southeastern U.S. was not the focus).

Potential reasons for the greater response in dry deposition compared to wet deposition and other factors that can affect the response in deposition are provided (last paragraph of section 3.4). It reads, "changes in emissions likely result in a more immediate change in dry deposition of S and oxidized N, since dry deposition is more localized. Changes in wet deposition are less regionally coupled with emissions because contaminants are removed from higher in the troposphere and higher up for some intense thunderstorms, thereby including contaminants from long-range transport. Process-based drivers can also have an effect on the N or S deposition response, such as variability in precipitation amounts, air mass patterns, aerosol chemistry as the relative amounts of ambient SO_2, NO_x and NH_3 change, and bidirectional flux of NH_3."

Tan, J., Fu, J. S., and Seinfeld, J. H., 2020. Ammonia emission abatement does not fully control reduced forms of nitrogen deposition. *Proc. Natl. Acad. Sci.*, *117*(18), 9771-9775.

 L659-660: It is not clear to me how the CL for the lakes were estimated - brief description or references to other work would be useful. Please also provide some discussion of what the max and min values of CL in Table S10 represent and how to interpret these in context of the discussion in L684-697.

Response: CL for the lakes surrounding selected CAPMoN sites were estimated using the Steady-State Water Chemistry (SSWC) model (Jeffries et al., 2010; Aherne and Jeffries, 2015) as described in section 2.4. We have included interpretations of the CLminN, CLmaxN and CLmaxS for calculating terrestrial CL exceedance in section 3.5 (2nd paragraph) as follows.

"Five of the 14 stations (ALG, CHA, CPS, LED, SPR) received total N+S deposition in exceedance of their terrestrial CL of acidity at some point in time across the study period (Fig. 11). The SPR site represented the highest initial terrestrial CL exceedance (beginning in 2003) and steepest decline into non-exceedance of the five exceeded sites; ALG followed a similar trend. The CPS site was only briefly in exceedance during a period of high deposition in 2000 and 2002. Wet deposition was the main driver of

the majority of the acidic deposition, and wet SO_4^{2-} alone resulted in terrestrial CL exceedance at these five stations. Note that there is no unique combination of S and N deposition to achieve or maintain non-exceedance; in some cases, changes to S or N deposition individually could result in a change of exceedance status, while in others, it may be easier to achieve change through varying reductions in both S and N. At four stations that did not experience exceedance over the time period, sensitivity to S deposition alone (that is, very low $CL_{max}S$) was estimated at BAB (208 eq ha^{-1} yr^{-1}) and $CL_{max}S$ values below 500 eq ha^{-1} yr^{-1} were estimated for MIN, CPS, and LED (Table S10). Poor weathering rates characterized the CL at these four sites, which have shallow soils (i.e. 30 cm or less) underlain by acidic parent material. The three southern stations (LON, EGB, FRE) had the highest $CL_{max}S$ (that is, they are tolerant to higher levels of S deposition, up to 2,936 eq ha^{-1} yr^{-1} at LON; Table S10); these stations are situated in areas with deeper soils (up to 50 cm) and alkaline parent material. The BON site showed high estimated tolerance to N deposition ($CL_{max}N$) at 2,589 eq ha^{-1} yr^{-1}, which is likely driven by poorly drained soils (and therefore a high denitrification fraction); other sites ranged between 398 and 1,719 eq ha^{-1} yr^{-1} (Table S10), with BAB being very sensitive to acidifying N (that is, $CL_{max}N$-$CL_{min}N$ is the smallest among the sites)…"

L731: The sentence "the critical load analysis presented in this paper is a case study" is vague? Should a case study not be representative of conditions? Reasons for continued acidic state of lakes in eastern Canada should be mentioned briefly.

Response: The term case study was used to clarify that it is not an exhaustive assessment of a vast lake population in eastern Canada, though the selected lakes cover a wide range of acid-base status. This is now clarified in section 4 (2nd paragraph). The following sentence was also added to explain why some lakes remain acidic despite the recent improvements in precipitation chemistry. "It is likely due to the long-term legacy of acidic deposition that decreased the size of the exchangeable base cation reservoir in the soils." The concept of legacy was already mentioned in lines 729-730 of the ACPD paper, but a clear explanation was not provided.