

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2022-419

Anonymous Referee #1

Referee comment on "Quantifying the importance of vehicle ammonia emissions in an urban area of northeastern USA utilizing nitrogen isotopes" by Wendell W. Walters et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-419-RC1>, 2022

General Comments

The Authors present a detailed foray into stable isotope source apportionment of NH_x in a urban area to ascertain the relative importance in accounting for vehicle and industry NH₃ emissions when setting air quality policy. They combine a robust observational dataset and isotope mixing model with archived NH_x observations to unravel the sources contributing to ambient levels in Providence, RI. Overall, the manuscript presents a strong case for the prevalence and significant contribution of vehicular NH₃ to urban air. Pending minor revisions below, the manuscript will be suitable for publication in Atmospheric Chemistry and Physics.

Minor Revision

- The most substantial oversight in interpreting the long-range transport component of the NH_x observations comes from the inferred agricultural regions of Canada that are upwind. While these areas are agriculturally intensive, the Authors seem to have overlooked that one of the 3 largest cities in North America, Toronto, is in this same fetch. It is a potent source of vehicular and industrial NH₃ emissions. Similarly, Montreal and additional industry along the St. Lawrence River in Quebec are overlooked. The Authors should consider these and probably revise their stress on wildfire (biomass burning) emissions towards those from more urban and industrial sources.

Technical Comments

Page 6, Line 165-168: Given the sampling setup, it seems strange that you'd find NO₂- in these extracts. Are the sources for this known? Is it an impurity in the reagents used for coating and sample preparation? A bit of elaboration on this could help reduce confusion on why this issue arises. Maybe give the breakdown of the criteria that were unsatisfied here (e.g. XX% of rejected samples were because of criterion 2)? This way the propensity of each issue is clear.

Page 9, Line 238: Should 'fertilization' be fertilizer?

Page 11, Line 294: Should % instead be ‰?

Page 11, Lines 308-310: A technical oversight here that there could also be increased physical losses driving the observations. The dry deposition loss of NH₃ to the Earth's surface is sustained during transport, and this would happen more readily in the winter. Suggest revising with the alternative process also used to provide context. There is plenty of literature on the subject.

Page 12, Line 332: 'sewerage' should be 'sewage' and appropriate references need to be added at the end of this sentence.

Page 12, Section 3.5: Implement consideration of the minor revision regarding urban and industrial areas in Ontario and Quebec throughout.

Page 12, Line 350: Deposition and revolatilization is not limited only to agricultural NH₃, but any emitted NH₃, which will then hop downwind and fractionate along the way. Suggest revising here.

Page 12, Line 354: Wildfires are not that common. Suggest retuning for urban emissions upwind.

Page 13, Lines 368-370: This is likely mixed in with volatilized NH₃ from Canada. Revise the discussion in this section to be a bit more aware of sources in the region.

Page 13, Line 375: It isn't clear how SCR differs from vehicle sources of NH₃? Here and below these are mixed and there doesn't seem to be a definition on why these are considered separate sources? I eventually found a description in the SI, but that example should be given here. From the SI it is not clear whether all power generation in the US is equipped with SCR by law? Couldn't there be quite a lot that is not? If that is the case, should SCR be used so specifically in the main manuscript? Consider revising for clarity.

Page 13, Line 377: Indoor sources requires an appropriate reference(s).

Page 14, Lines 409-411: In summer, your source region is strongly influenced by the heavily urbanized Toronto and shoreline area.

Page 14, Line 424: This line of reasoning about wildfires doesn't have much factual justification. Is this speculative or can quantitative metrics be brought to bear on this? Suggest removing throughout if it is speculation as a major Canadian source. Biomass burning/wildfire events intruding into the US are largely confined to those originating in Alberta and British Columbia.

Page 30, Figure 7: Can an accurate representation of the great lakes be added to these figures? Since there's discussion regarding Ontario and Quebec linked to this, the major geographical features that make them identifiable should be properly plotted here.

Supporting Information

Page 4, Figure S3: Given the specific commentary on sources of NH₃ from Canada, have the authors looked into acquiring CAPMoN or NADP datasets to include the source region more completely in this visualization?

Page 7: 'may closer represent' should be 'may more closely represent'

Page 7: '.... Representative of all industry-related NH₃ emissions'. Since steel industry uses coal in the coking process, this isn't too surprising to see a similar trend to the values found from fuel combustion. Values from natural gas or oil-fired facilities would provide a greater level of confidence that all fossil fuel fired power or industrial activities yield a

similar NH3 signature.

Page 7: why does '14' follow the per mil value?