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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Constraining Ammonia Emissions in Vehicle Plumes Utilizing Nitrogen Stable Isotopes" *by* Wendell W. Walters et al.

Anonymous Referee #1

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This paper by Walters et al. entitled "Constraining ammonia emissions in vehicle plumes utilizing nitrogen stable isotopes" describes several measurement campaigns that occurred either in Northeastern US or in China aiming at measuring NH3 and pNH4+ (= NHx) mixing ratios and isotopic composition. The authors used an active collection technique (filters + denuders) previously tested in laboratory to ensure the complete collection of NHx. They compare results from this technique from observations using passive collection, both from this study and from the literature. They also discuss the variables that could explain the observed spatio-temporal variability in d15N-NH3, and finally provide an updated range of d15N-NH3 from vehicle emissions. I find that overall the paper is well written, and the findings are definitely worth of publication, provided that the authors address the mostly minor comments that follow. I

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Discussion paper

especially appreciated the care taken data quality check, and the detailed field and lab operating procedures.

My main concern relates more to the structure/length of the article. I think that overall it is a very long paper to highlight the main result, which really is the range of d15N-NH3 from vehicle emissions. I think this could be a much shorter paper, which could emphasize more on the importance of characterizing NH3 isotopic composition from vehicle to be used in prospective NH3 source apportionment studies, which will likely become the norm in the close future, as has been the case over the past 20 years for HNO3. I realize that this comment is probably not very constructive, but I was thinking that the whole active/passive collection technique comparison, while very interesting and certainly useful, could be the subject of a separate article that the authors could refer to here. That alone would considerably lighten the results/discussion, and would help the reader to follow more easily the different campaign results and subsequent discussion.

Detailed Comments:

I think the title is somewhat misleading. You don't really constrain the vehicle ammonia emissions using N isotopes. The title as is suggests a source apportionment study, which is not the case. It should read: "Characterizing the isotopic composition of ammonia from vehicle plumes" or something like that.

Your abstract makes no mention of the comparison between active and passive collection techniques, which supports my previous point that you could remove that from your manuscript and have it in a separate paper. It reads as a sideways discussion in the present format, and distracts the reader from the main findings. I am not saying it is not interesting and useful, just that it could be its own paper.

L. 37-39: I thought soil acidification is mostly due to HNO3. How can an alkaline compound like NH3 cause acidification?

L. 47: Helpful if you could indicate here NH3 atmospheric lifetime.

L. 48-49: The Templer group in Boston has more recent studies highlighting large vehicle contribution to urban NH3 budget. Check out the Decina et al. papers, particularly relevant since you drove to Boston for this study.

L. 61: Can you quantify here the contribution as a % at the global scale? L. 65: Once again, check the work lead by the Templer group in Boston about N deposition in urban areas.

L. 66-67: how are "fuel-combustion" and "vehicle" sources different? Isn't the latest included within the first?

L. 90-91: Didn't you just say that these techniques were shown to not accurately capture the d15n-NH3, based on work by Skinner et al.? This seems contradictory.

L. 158: How long is the inlet line?

L. 209: Did you characterize potential inlet loss, and induced fractionation on NH3, to see if tit was indeed negligible?

L. 214: Any chance the denuders could trap a portion of the particulate phase as well, on top of the gas phase?

L. 214: Can you give quantify your detection limits?

L. 219: pNO3-, but what about pNH4+ ?

L. 241: What do you use the ethanol for?

L. 380: Does it mean that the urban background NH3 has the isotopic composition of vehicle emissions?

L. 395-401: I understand that you can't estimate f(NH3) accurately, but why can't you calculate the concentration of pNH4+ here? Were the Nylon filters also saturated? There is no mention of that aspect it, and it should be expanded on.

L. 409: An introduction sentence about what ISORROPIA is would be nice.

L. 421: I think it would be useful and interesting to provide, maybe in the SI, the isotopic composition for each component, especially the nylon-collected pNH4+. And maybe expand on the different isotopic composition of NHx and pNH4+, if such is the case (and I expect it to be).

L. 431: Section title should be revised; it is the same ast the previous section title

L. 481: Please recall here what are elevated NH3 concentrations.

L. 521-523: Maybe recall that your f(NO3) is approximate in this case.

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