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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Wintertime Spatial Distribution of Ammonia and its Emission Sources in the Great Salt Lake Region" *by* Alexander Moravek et al.

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We thank Reviewer #2 for their overall positive feedback on our manuscript. We addressed their comments as follows:

Comment: *Abstract: is there a way to explain, even simplistically, "enhancements" better in the context of an abstract? It is an atypical expression – usually emissions are compared or concentrations are compared. I'd encourage an extra sentence, if possible, for clarification for readers.*

Response: We agree that the terminology "enhancements" might not be known by every reader in this context. We therefore added a sentence which explains the terminology.

Comment: *Page 2: line 3 sources, line 15 (double parentheses)*

Response: We corrected the word "sources" and separated the chemical formula by commas from the text.

Comment: *Introduction: I'd recommend adding some information on the Cache Valley AMoN site, for context. It has been called a supervolcano of ammonia with the highest average annual NH_3 in the network (by a fairly large margin). Is it also the highest in winter (Jan/Feb) compared to the other sites in the network? If so, state this – it helps raise the importance of the work. More relevantly, here and later on in the discussion, some context of the AMoN sites in this region may be helpful during the campaign – i.e. how the 2017 Jan/Feb period compared to other years. The authors noted that the cold pools were not as consistent/frequent as in other winters, curious if AMoN was similar/different.*

Response: We agree that putting the presented data in context with the AMoN measurements is very useful. As suggested, we added a sentence in the introduction on the high NH_3 measurements compared to other regions within AMoN. As it is shown on the AMoN website (http://nadp.slh.wisc.edu/amon/; Figure: Quarterly AMoN Concentrations, 2012), the region also has the highest NH_3 measurements in the Network in the Winter months (January to March). We further added a paragraph on the AMoN measurements at the end of Sect. 3.2.1. Despite the less frequent PCAP periods, the NH_3 measurement in the January/February 2017 were comparable to other years. Furthermore, as for the presented ground site measurements, the NH_3 concentrations at the Cache Valley AMoN Site were about 10 time higher than at the

Salt Lake City site.

Comment: *Section 2.1/2.2: how many flight hours were conducted in the campaign? And how many flight hours were there NH_3 measurements?*

Response: A total of 58.3 flight hours were conducted, with 53.6 hours of NH_3 measurements. After quality control 38.7 hours (72 %) of NH_3 data were used for the analysis. We included this information in Sections 2.1 and 2.2 of the revised manuscript.

Comment: *Page 4, line 10: Is it a QC-TILDAS or cw-QC-TILDAS? QC-TILDAS is generic to Aerodyne's instruments – not sure of the proper description, but be consistent. Or just cite like Picarro is later.*

Response: In literature multiple abbreviations for quantum cascade laser instruments are used giving different levels of detail. For NH_3, the use of QC-TILDAS has been used most extensively in the past years. The Aerodyne model of the used continuous wave QC-TILDAS is called QC-mini. To be consistent with other instrumentation listed, we joined the information on the model + manufacturer and moved it to the second sentence in the paragraph.

Comment: *Section 2.2: The instrument performance and description are lacking, probably the largest weakness in this manuscript, particularly since the instrument used wasn't really similar to those used in past references. The wavelength is different, which results in different pressure and temperature dependence – more discussion is needed. The following points are introduced/discussed first but never quantified at this stage, e.g.: -P4, L13: "fast time response" and "high precision" – yet noted quantitatively at this point, nor relative to what other commercial sensors (what about research*

sensors, which are better than commercial ones?) No data were shown that the instrument was "fast response", even with the improved inlet design. Quantify the response time, t10-t90 for some representative NH_3 level observed. – also, what is the detection limit of the instrument? 3σ of the precision isn't necessarily the detection limit, if systematic errors occur from backgrounds or inlet effects.

Response: We agree with the Reviewer that more information on the instrument performance can be given. For that reason we included more details in Sect 2.2.

Using the NH_3 absorption line at 965.3 cm^{-1} yields a lower absorption than at the 967.3 cm^{-1}, resulting in a slightly decreased precision. However, the pressure and temperature dependency of the instrument largely depends on the fringe pattern (we clarified this in the revised manuscript in Sect. 2.2). The fringe pattern will in most cases be different at a different wavelength, but it also differs with every adjustment of the laser beam (by mirrors or changing the vibrations through wiggler) at the same wavelength. For that reason a systematic temperature/ pressure dependency which was valid for the entire dataset could not be detected.

Regarding the time response, we describe in the updated manuscript the time constant/ response time value which makes it better comparable to other studies. We use a double exponential function since this describes the time response for ammonia more adequately than the t10-t90 time.

Regarding the precision, we changed the section to include the nominal precision of the QC-TILDAS and precisions of other instruments. We removed the differentiation between commercially available and other analyzers.

The 3σ limit of detection was determined from zero air measurements. We only considered the random error for the determination of the detection limit. Systematic differences (such as due to changing backgrounds) were corrected for or data periods with large systematic differences were removed as described in the revised manuscript (see comment by Reviewer #1).

Comment: *P4, L14-15: "weight was reduced" – reduced from what? And what was the mass?*

Response: The weight reduction refers to the original QC-mini for NH_3 as it is currently sold by Aerodyne in its standard modification (the total instrument weight including vacuum pump, chiller, inlet housing and tubing and screen/ keyboard is about 100 kg). By using a different pump and new inlet design, we could reduce the total weight by about 20 kg (not accounting for additional weight of the UPS unit and winglet mounted into the aircraft roof). We included this information in Sect 2.2 of the manuscript.

Comment: *P4, L19: "within the instrument detection limit"...which was? What was the residence time of air from the tip of the inlet to the sample cell?*

Response: The detection limit is discussed further below in the same paragraph. We would find it redundant to list the detection limit here again, since this sentence is on systematic differences. The residence time of air from the inlet to the sample cell was approximately 0.1 s, which is fast enough for the 1 s sampling rate.

Comment: *P4, L26-27: "Fringes: : :are caused by optical interferences" – circular statement, fringes are optical intereferences. Maybe reword to "Optical interferences (fringes) are periodic structures in the absorption spectrum that influence precision and drift of the sensor, if the fringes are of a wavelength comparable to the absorption linewidth" or something like that.*

Response: We thank the Reviewer for the more precise definition and used it in the manuscript.

Comment: *The tenses in Section 2 are a mix of present/past tense. I'd recommend past tense, but either is fine if consistent.*

Response: We changed the tense to past tense where applicable. In general, we used past tense to describe the methodology that was implemented by the authors. However, for generally valid statements such as the description of the instruments' measurement mechanisms and the composition of the emissions inventories we still use the present tense.

Comment: *P5, L1-7: A weaker line was probed, yet the sensitivity was better (!) than the original reference (though it was noted degraded from "usual" performance)?! It seems that the past instrument/citation was similar in make/model but the specifications may be much different, and therefore it is all the more important that these details are discussed clearly. It is clear all Aerodyne instruments aren't alike. It would be helpful to see the profile of the NH_3 sensor on the ascent/descent of a missed approach, comparison to some other short-lived tracer, particularly focusing on the free troposphere – boundary layer transition (gives an idea of the sampling / response time). Another option is to compare the ascent with the descent, recognizing that there may be some spatial (horizontal) differences near the ground.*

Response: The preision of the QC-TILDAS is governed by multiple factors of which one is the absorption line strength. At the date of submission, Hacker et al. (2016) represented the only other aircraft NH_3 measurements with a QC-TILDAS, which is why we compare our instrument performance to them. However, they do not state if they used a pulsed or a continuous wave laser instrument. The former has a significantly lower sensitivity than the continuous wave laser instrument used in this study. Furthermore, critical for the precision are adjustments of the mirrors and the laser path made by the operator, which may results in a different instrument performance under different conditions.

As it is mentioned at the end of Sect 2.2, the data from spiraling ascents/descents data was mostly discarded for the presented analysis due to potential mixing ratio drifts. Following the comment from Reviewer #1, we clarified in the revised manuscript the procedure at the end of Sect. 2.2. However, we included in the Supplementary Material an example of a missed approach NH_3 profile (Fig. S2 in revised version), which often showed a good data quality. As mentioned also in the answer to the comment below, the example shows that mixing ratios in the ascents/descents compared quite well if horizontal heterogeneity was small.

Comment: *P5 ,line 24: extra period*

Response: Period was removed.

Comment: *P5, line 27: "some flights" – how many?*

Response: We corrected it to: "Wind data were compromised for some flights, making only partial coverage (65 – 95 %) available for eight flights and resulting in no wind data for six of the 23 flights."

Comment: *P6, "northeast", not "north east"*

Response: The changes were made.

Comment: *P7 and elsewhere: "area sources" is clear to mean agricultural/feedlots/CAFOs, so why not simply state "feedlots" or "agricultural" more generally. Focusing on their type (ag) versus point-vs-area is more important. A general statement can be made in the introduction that the agricultural sources are not simply point sources like exhaust but rather occur through the scale of a feedlot, field, or feeding*

pen. For the context of the analyses (emissions/STILT), these are effectively numerous, point sources from the airplane's perspective (i.e. lots of CAFOs in a general grid domain).

Response: It is true that areas sources represent mainly emissions from agriculture. However, area sources may also include emissions from other sectors than agriculture, such as residential wood combustion. Especially in Salt Lake County, the contribution of these other sources may be significant depending on the area source definition. To be consistent with the inventory description and to be most precise in the terminology, we decided to keep "area sources" when it is referred to the emission inventory. We agree with the Reviewer that the terminology of area sources should be more clearly defined. We therefore added a general definition of area sources in Sect. 2.5.

Comment: *P8, L25-30: Given that one has meteorology and can use deposition velocities, what are the deposition loss terms? More justification is needed to consider NH_x as a passive tracer, or at least the caveats of assuming this.*

Response: Since no reliable vertical wind velocity measurements could be obtained from the aircraft, an accurate determination of the deposition velocity is not viable. However, in Sect. 2.6, we mentioned that the used approach does not account for dry and wet deposition. Due to the bi-directional nature of NH_3 exchange the determination of the dry deposition loss terms which would impact the modelled NH_x enhancements is not straight forward. The actual dry deposition will depend on the above surface NH_3 mixing ratio and surface resistance term towards the NH_3 uptake. The latter largely depends on the surface's ability to adsorb or uptake NH_3 and varies largely by the surface type.

Comment: *P9, L5: the 1st percentile seems reasonable, but perhaps in the SI one could provide some sensitivity to that choice (vs. 0.1 %, 2 %, etc.)*

Response: Following the suggestion from the Reviewer we performed a sensitivity analysis which evaluated the differences between choosing a 0.1, 1 (as used in this study) and 2 percentile. We found that the differences in the obtained baseline using these values are small. For the example as shown in Fig. S15 (Fig. S14 before revision) the standard deviation between the three scenarios is 0.056 ppbv (calculated for each altitude layer and then averaged over the vertical profile). The median standard deviation of the three scenarios was 1.8 % and for NH_x values above the instruments detection limit the deviation was only 1 %. This underlines that the sensitivity of the baseline correction on the choice of the percentile was not very strong. For the presented example, this can be explained by the fact that the amount of data used in each bin, was typically too low to retrieve different values for the 0.1, 1 and 2 percentiles. We added a sentence with the conclusion of this analysis at the end of Sect. 2.6.

Comment: *P10, L9: nighttime vs. night-time*

Response: We corrected it all to "night-time".

Comment: *P10, L16-31, on the vertical profiles of NH_3 near the ground: NASA DISCOVER-AQ data in California in Jan/Feb in the San Joaquin Valley also had very strong inversions, and the two different airborne NH_3 instruments showed dramatically different profiles up vs. down – and the vertical profiles were certainly not monotonically decreasing. While I agree with the interpretation that the concentration of NH_3 should be highest at the ground, and this could be a reason for differences between aircraft/ground sites, I wonder how much sampling/response times of the inlet/instrument affect these values. Going from cleaner regions up above to very high levels on the missed approach will result in surface adsorption effects buffering the actual concentrations measured by the instrument. A reverse effect may occur going upward, though*

not necessarily symmetric – do the ascent/descent profiles agree on average?

Response: In general we do not see a systematic bias of the mixing ratios collected during the descents/ascents of the mixed approaches which might be attributed to time response effects of the NH_3 measurement. If there were significant adsorption/desorption effects in the inlet system, one may expect that the NH_3 mixing ratios during ascends are on average higher due to the higher NH_3 levels at the ground. This was not the case. We added this observation at the end of Sect 2.2 together with two examples in the SI of missed approaches at the Logan airport. Figure S2 a) shows an example where horizontal heterogeneity played a dominant role as mixing ratios steadily increased when the Twin Otter was flying northwards over the Logan airport runway. In Fig. S2 b), the vertical NH_3 profiles match very well during the descent and ascent below an altitude of 1450 m.

Comment: *The discussion of the various emission inventories (USU, UDAQ) and how they are implemented (diurnal/weekly/monthly) is well developed. However, this manuscript had relatively few comparisons to other papers that also showed emissions are lower than what observations suggest (a general trend). This manuscript represents another convincing case study that NH_3 emissions are vastly under-reported in most inventories, and some context of prior work should be noted (e.g. a paragraph). Are the magnitudes that the inventories are "off" – for ag and mobile sources – consistent with other studies in the literature? I wouldn't expect them to be identical (or necessarily even close, due to differences in season/location/etc.), but trying to put some context would be helpful. Were other studies off by factors of several for feedlot regions? Or mobile emissions off by 30 %?*

Response: We agree with the Reviewer that the comparison with other studies can be improved. We therefore added a paragraph at the end of Sect. 3.3.3 discussing previous studies that compared NH_3 emissions from inventories with those derived from measurements. As stated, other studies an even higher underestimation of

emission from agriculture depending on the area and the used inventory.

Comment: *Fig. 1: labels are very tiny (and missing bracket on the lower one for [ppbv]*

Response: We increased all labels and added the missing bracket.

Comment: *Fig. 2: Add lengths between the 16 Lpm flow and aerosol impactor and impactor to cell*

Response: Both lengths were minimal (< 10 cm, including the size of the PFA fittings). We added this information in the caption of Fig. 2.

Comment: *Fig. 3: caption reads (a) Univ. Utah (b) Cache Valley but figure panels are reversed from that*

Response: Thank you for noting this inconsistency, the labels in the figure panels are correct. We corrected the caption accordingly.

Comment: *Fig. 5: legends are incredibly small to read, both #s and units*

Response: We increased the all axes and legend markers and labels.

Comment: *Overall, this is a very good manuscript with detailed analyses from novel flight measurements. The conclusions are sound and well-justified, just additional (straightforward, I believe) clarifications are needed to improve it further / make things clearer to the reader.*

Response: Following the suggestions and valuable input of the Reviewers, the revised version of the manuscript includes more technical details and clarifications. Still, none

of the revisions have changed the overall conclusions of the manuscript.

Reference

Hacker, J. M., Chen, D., Bai, M., Ewenz, C., Junkermann, W., Lieff, W., Mcmanus, B., Neininger, B., Sun, J., Coates, T., Denmead, T., Flesch, T., Mcginn, S. and Hill, J.: Using airborne technology to quantify and apportion emissions of CH4 and NH3 from feedlots, Anim. Prod. Sci., 56, 190–203, 2016.

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