Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-266-RC2, 2019

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.

Anonymous Referee #2

Received and published: 7 October 2019

Moravek et al., ACP, "Wintertime spatial distribution of ammonia. . ."

The manuscript analyzes airborne measurements in Jan/Feb in northern Utah where PM2.5 levels are often non-compliant for particulate matter. The focus of the study was to understand ammonium nitrate formation, and precursor sources, for both cold-pool (strong valley/winter inversions) and non-cold pool conditions that occur frequently in winter. The authors analyze the measurements in the context of STILT footprints and examine enhancements of NH3 and NHx compared to those derived from various emission inventories. The key finding is that NH3 emissions are significantly under-estimated in each valley region (compared to observations), particularly in the Cache

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Valley region where large agricultural (feedlot) emissions are occurring. Another significant result is that the large NH3 plume in the Cache Valley does not appear to influence the more populous Salt Lake City area (NH3 largely locally generated for that valley), though NOx emissions from outside valleys do appear to influence the Cache Valley (where ammonium nitrate formation is NOx-limited). The results/findings have large implications for understanding, and ultimately mitigating, PM2.5 in northern Utah, and the manuscript is very well-suited for ACP. I recommend publication upon considering/revising the points outlined below.

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.

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

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 NH3 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.

Section 2.1/2.2: how many flight hours were conducted in the campaign? And how many flight hours were there NH3 measurements?

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.

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 NH3 level observed.

– also, what is the detection limit of the instrument? 3sigma of the precision isn't necessarily the detection limit, if systematic errors occur from backgrounds or inlet effects.

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

-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?

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.

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

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 NH3 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.

P5 ,line 24: extra period

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

P6, "northeast", not "north east"

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).

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 NHx as a passive tracer, or at least the caveats of assuming this.

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.)

P10, L9: nighttime vs. night-time

P10, L16-31, on the vertical profiles of NH3 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 NH3 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 NH3 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?

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 NH3 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%?

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

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

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

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

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.

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