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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The influence of gas-particle partitioning and surface-atmosphere exchange on ammonia during BAQS-Met" *by* R. A. Ellis et al.

Anonymous Referee #1

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This paper presents unique data on measurements of gas and particle phase ammonia (along with other species) at a rural site. The observations are compared to a model and discrepancies observed. At high sulfate loadings the model under-predicts the ammonia gas fraction relative to total gas plus particle, and at low sulfate the model over-predicts the gas fraction. The authors conclude that this is due to a source/sink not included in the model whereby plants emit or absorb NH3 in response to ambient NH3 concentrations. Apparently this idea has been raised by other investigators and so it seems plausible. However, is this really the only viable explanation? I believe the authors should rule out all other possibilities before asserting that this is the missing process.

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Discussion Paper

For example, can the authors rule out any measurement uncertainty or bias that may explain the discrepancies? It is noted that no discussion is provided in this paper on the uncertainty or quantitative accuracy of the measurement data.

Is it possible that there is a significant component of ammonium associated with the coarse particle mode, which could influence observed gas phase NH3 levels and hence the measured gas fraction.

Could it not be argued that based on Fig 9, which compares the ammonia gas fraction versus sulfate, that the measurements and model discrepancy is systemic and that the authors should not just focus on the discrepancies at either sulfate concentration extreme (which is the basis for the bi-directional flux from/to plants as a function of gas ammonia concentrations). One could argue that the model overall does a poor job and thus is likely lacking in many respects. It is noted that even when a bi-directional flux is included (though it may be a crude representation), the improvements were marginal.

As a side question, why do plants attempt to keep gas phase ammonium in a certain concentration range? Could the proposed missing source realistically provide the amount of missing ammonia needed to bring models in agreement with observation during high sulfate conditions?

Specific comments.

Pg, 21901 line 10, give the calculated cut size of the inertial separator used to remove particle interferences. The NH3 inlet configuration is also not clear; does the complete inlet only consist of a 10cm quartz tube, or are there additional sections. How high was the inlet located above ground level and how was it located relative to activities around the site?

Pg 21902, clarify the sentence: $\frac{1}{2}$ OD Teflon-coated aluminum tube breaching a 4 diameter PVC pipe. Also, I assume this was the same inlet used for particles. Include an estimate of particle sampling looses (or lack of them). Are there issues sampling

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particles through a teflon-coated tube due to electrostatic effects. No information is provided on the AIM accuracy (eg, was it compared to filters etc), measurement LODs or uncertainties.

Pg 21904, line 9, clarify what is meant by exact overlap, what time scales were associated with the miss-match? Line 26, give distance to road.

Pg 21905, lines 4 and 5, but was there a direct correlation between the field work and measured NH3, this is implied but not explicitly stated. Line 24, missing "be" (to be expected?).

Pg 21908, regarding the discussion of variability between 10:00 to 15:00, numbers are needed to justify the statement that partitioning contributed to NH3 decrease during this time (line 15). In Fig 5a, it appears that there is a 10% decrease in NHx, is this significant relative to the decrease in NH3?

Pg 21910 line 18-20, what about changes in BL height and variability in the vertical concentration profile of NHx as a process that influences NHx concentrations in each model grid cell.

Pg 21911 and 21912 regarding the discussion of bi-directional flux of NH3 involving ambient NH3 concentrations and plant apoplastic fluid. The argument is that plants attempt to keep some form of equilibrium between apoplastic fluid and ambient NH3 levels and that this explains the discrepancy between the model and observations. Is it reasonable to assume that the plants can adjust that quickly and with sufficient emissions to account for the large differences observed?

In the Conclusions definitive statements are made but were not completely proven in the body of the paper. Eg, Pg 21913 line 5, I don't believe that spikes at night were proven to be from cars, I thought it was only speculated. Other statements should be considered and qualified if need be.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 21895, 2010.