

## ***Interactive comment on “Ammonia at Blodgett Forest, Sierra Nevada, USA” by M. L. Fischer and D. Littlejohn***

### **Anonymous Referee #2**

Received and published: 18 December 2007

#### General Impression.

The paper by Fischer and Littlejohn presents a short time series of measurements of NH<sub>3</sub> concentrations and fluxes made with a QCL absorption spectrometer at the Blodgett Forest Research Station. There are few direct measurements of ammonia fluxes over North American ecosystems, and thus any addition to the sparse database potentially makes a significant contribution to the literature. Unfortunately, both the measurement period and the data analysis are fairly short and do not provide the information needed to improve the description of dry deposition processes in atmospheric models. Instead, the authors focus in their conclusions on the controls of the ammonia concentrations by linking the measurements to a somewhat tentative modelling exercise. The application to such a clean site is pushing the detection limit of the QCL. There appears

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to be instrument issues with the QCL which are reflected in a different noise pattern during different periods and poor agreement between QCL and denuder sampler. It is therefore questionable whether the measurements support the conclusions or further analysis of the fluxes suggested here.

#### Major Scientific Points:

The description of the instrument setup is insufficient to judge whether it was suitable for flux measurements. For example, was the inlet sufficiently far away from the bluff body of the QCL and scissor lift? The measurement height (10 m) is certainly low compared with the tree height (10 - 12 m), and measurements were made well within the roughness sublayer, possibly within the canopy. Fig. 1 should be modified to illustrate the entire setup, not just the calibration manifold.

It sounds like the PTFE filter is subjected to 50 Torr. Surely, this would lead to evaporation of all available ammonium nitrate (and ammonium chloride) and subsequent detection as ammonia. Thus the measurement relates to the sum of gaseous ammonia and volatile ammonium aerosol. This needs to be discussed and considered throughout the manuscript.

It is not clear whether or how ammonia chemistry was considered in the back trajectory modelling and STILT. Having been developed for CO<sub>2</sub>, it is likely that chemistry is not considered. This would make the model inappropriate for NH<sub>3</sub>. Although there was no potential for aerosol formation at Blodgett, there may well have been potential for aerosol formation in the emission areas which would have changed the transport distance and atmospheric lifetime of total reduced nitrogen.

The difference between QCL and denuder samples could be due to problems with either method, but the authors need to resolve this problem to lend support to the remainder of the paper. What are the dimensions of the denuders that they are effective at 100 lpm? Have they been confirmed to be 100% efficient?

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The comparison between measured and predicted equilibrium gas phase concentration products of  $\text{NH}_3 \times \text{HNO}_3$  adds little information to the paper as the concentration of  $\text{HNO}_3$  was guessed (taken from a different measurement period).

It seems counterintuitive that the power spectrum of  $w'\text{NH}_3^2$ ; shows a weaker slope than sensible heat. A damped signal should show a steeper slope. Hence, Fig. 7 raises more questions than it answers and provides little support that the frequency response of the system was sufficient. More discussion is needed. Are the unexpected slopes due to the effect of measuring well within the roughness layer (rather than the inertial sublayer)?

The comparison of  $V_d$  and  $V_{d\max}$  (Fig. 9) only demonstrates that the measurements are not in contradiction with  $V_{d\max}$ . It would be more helpful to show a time-trace of  $R_c$  instead, which is a parameter that models can use. In general it would be helpful to see a table listing summary statistics (range, mean, median, standard deviation etc.) of concentrations, fluxes,  $V_d$  and  $R_c$ . In the analysis, the authors ignore the entire European literature on  $\text{NH}_3$  fluxes, in which this kind of analysis has been performed for 20+ years. The conclusions on the deposition rate (P14153, L2-5) are not clear and highly qualitative.

In addition, there are references to TDL/QCL fluxes that should be included (e.g. Famulari et al., Water Air Soil Pollution Focus 1, 17-27, 2005; of interest is also the paper: Whitehead, J.D., Twigg, M., Famulari, D., Nemitz, E., Sutton, M.A., Gallagher, M.W. & Fowler, D.: Evaluation of laser absorption spectroscopic techniques for eddy covariance flux measurements of ammonia. Environmental Science & Technology, in press).

Technical Comments:

P14143, L1: Should be: "... during a two-day period ... with a filter sampler " as the intercomparison has not previously been introduced. Also LBNL should be introduced on first use.

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P14143, L17: incomplete sentence: "The mean ammonia flux ...".

P14147, L9: There seems to be a 'minus'; missing:  $V_d = - F/\text{conc}$ , judging by the sign convention used in the paper.

P14147, L13. It is well known that  $R_c$  is composed of two parallel pathways: stomatal uptake and cuticular uptake. In very dry conditions (probably dominating during this period),  $R_c$  may approach  $R_s$ .

P14148, L5. This equation needs a reference.

P14148, L12. Delete 'and'.

Figures. Please explain what the error bars represent in the figure captions, where appropriate. Why did the author decide not to connect the symbols of Figs. 4, 5 and 8?

Fig. 10. Additional 'of' in caption.

Fig. 6. Mismatch between legend (with symbols) and trace (no symbols) of red trace.

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Interactive comment on Atmos. Chem. Phys. Discuss., 7, 14139, 2007.

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