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Biogeosciences Discussions

Interactive comment on "Exchange of reactive nitrogen compounds: concentrations and fluxes of total ammonium and total nitrate above a spruce forest canopy" *by* V. Wolff et al.

V. Wolff et al.

veronika.wolff@mpic.de

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We thank the reviewer for the comment and reply to it after quoting it in italic for convenience.

Anonymous Referee # 2: This paper investigates the dry deposition of total ammonium and nitrate to a spruce forest canopy using a semi-continuous wet chemistry technique. The paper is well written and the subject matter is of interest to a wide audience from ecologists to atmospheric chemists and therefore suitable for publication in Biogeosciences. Though I have only a few comments, collectively they constitute a major revision. However, I believe the authors can easily address the necessary

changes without a large increase in the length of the manuscript. I recommend publication subsequent to treatment of these suggestions. This manuscript deals primarily with fluxes of total ammonium and nitrate and the authors state that individual fluxes (NH_3, $NH4^+$, HNO_3, NO_3^-) will be analyzed in a forthcoming paper. I feel strongly that this manuscript should be modified to incorporate an analysis of the individual fluxes. If the authors had a full year of observations I might go along with the idea of publishing one paper on total deposition and a second, more process oriented paper, focusing on individual compounds. However, one month of observations is not enough to draw any substantive conclusions about total N deposition to the site without a fairly rigorous model-based scaling exercise. The simple scaling exercise to derive annual deposition described in section 4.2 is not appropriate and should be removed from the manuscript (along with Figure 8). In this case, I think a more detailed analysis and discussion of concentrations and fluxes of total ammonium/ nitrate and individual compounds yields a much more interesting and useful paper. I suspect there is already significant overlap between the current manuscript and the forthcoming paper on individual compounds (section 3.4, much of section 4.1, points b and c in the Conclusions section). Indeed, the total ammonium/nitrate fluxes cannot be fully interpreted without examination of the individual fluxes.

» In the submitted manuscript we report deposition loads of reactive nitrogen of the NH_3-HNO_3-NH_4NO_3 triad by lumping gaseous and particulate nitrogen compounds. A timescale analysis showed that measured vertical concentration differences of individual compounds were most likely affected by phase changes (chemical divergence) driven by vertical gradients in temperature and relative humidity above the forest canopy. The constant flux layer assumption (Fowler and Duyzer, 1989) is therefore violated such that a calculation of exchange fluxes of individual compounds of the NH_3-HNO_3-NH_4NO_3 triad using the aerodynamic gradient method is not allowed. In order to assess qualitatively or even quantitatively, the effect of the phase changes on the measured differences and therefore derive a realistic exchange flux of individual compounds, a detailed analysis on equilibrium and phase changes with height would

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be necessary, which include modelling exercises. Such an evaluation is beyond the scope of this paper, which focuses on ecosystem input of nitrogen as pollutant and nutrient. Potentially, the inclusion of individual fluxes and associated analysis would lead to a change of the title of the paper and would increase the paper length considerably.

Nevertheless, there is, of course, an overlap between the total deposition and the equilibrium processes in the triad which is the reason for addressing that issue at several points in the manuscript (section 3.4, much of section 4.1, points b and c in the Conclusions section). In the revised version of the manuscript we emphasise the reasons why individual fluxes could not be calculated using the aerodynamic gradient method. We agree with the reviewer that a one month measurement period is too short to compare with annual estimates/measurements. However, we found large deposition fluxes that require a comparison with other studies. Very few studies on direct, micrometeorological exchange fluxes are reported in literature. Following the reviewers suggestions, we shortened the controversial comparison with annual budgets (and deleted figure 8), highlighting monthly comparisons and we only shortly address other annual deposition estimates.

Reference:

Fowler, D., and Duyzer, J. H.: Micrometeorological techniques for the measurement of trace gas exchange, in Exchange of trace gases between terrestrial ecosystems and the atmosphere, edited by M. O. Andreae, and Schimel, D. S., Wiley, Chichester, pp. 189-207, 1989.

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