

12/20/10

General Comments

This paper presents the first indirect measurements of N₂O₅ deposition to a snow-covered surface. Such observations require adherence to stringent meteorological and site-specific criteria, and the authors do an excellent job of considering these factors in their experimental design and execution. The use of heat gradients and fluxes is a particularly convincing check on the aerodynamic method. These results are essential for constraining deposition in regional and global models where hydrolysis of N₂O₅ is a dominant pathway for the loss of reactive nitrogen. The manuscript is generally well-written and is appropriate for publication in ACP. My comments and questions are primarily aimed at adding relevant details, broadening the discussion and strengthening the take-away message.

Specific Comments

p. 25330, l. 9 – I suggest providing a number for the fraction of depositional contribution to total loss (from Sec. 5, it looks like ~1/8 to 1/4).

p. 25331, l. 5 – I believe the referenced reaction should be R3b. Also, since you have not yet broken out R3 into ‘a’ and ‘b’ at this point, it would help to specifically mention that you are referring to aerosol uptake.

p. 25331, l. 9 – You might also reference (McNeill et al., 2006).

p. 25331, l. 26 – The reference to (Bertram et al., 2009) would seem more appropriate in the preceding paragraph, as it deals with measurements of N₂O₅ uptake to aerosols.

p. 25332, l. 3 – How much chemical loss was unaccounted for in these studies? 50%? 90%? Mentioning this number will help set up the importance of quantifying missing sinks.

p. 25333, l. 5 – Can these reactions not also make ClNO₂ if Cl⁻ is present in the aerosol or snowpack? I realize that Cl⁻ measurements are likely not available to explore such a possibility, but it may be worth mentioning that HNO₃ is not the sole end-product. This is also motivation for future work on the fate of the N₂O₅ in the snowpack.

p. 25334, l. 5 – This may be personal preference, but I am against citing dissertations as these could be considered “grey literature” (i.e. difficult to find). Given that you note several times that the size of the fetch could be a source of uncertainty, would it be possible to provide a brief summary of these extra fetch calculations in the supplementary material?

p. 25334, l. 13 – Can you provide a Reynolds number for your inlet conditions?

p. 25334, l. 16 – Twenty-four percent seems like a substantial inlet loss and a potentially large source of uncertainty in your concentration measurements. You might mention that such

systematic errors in concentration data would not propagate into the flux calculation as you are taking a difference.

p. 25337, l. 3 – For clarity, I suggest separating this into two equations, one for the flux and one for the exchange velocity.

p. 25339, l. 18 – The change in the N₂O₅ thermal decay lifetime for a 1°C temperature change is ~12%. Could this temperature difference affect the N₂O₅ gradient and thus the flux of N₂O₅? Given that the temperature profile is positive, I would expect this to lead to an underestimate of the actual depositional flux. You may be able to estimate such an effect using a method similar to that described by (Wolfe et al., 2009) for PAN.

p. 25340, l. 19 – This first sentence strikes me as odd when read on its own. Perhaps you are trying to say that deposition velocities are the critical input parameter for models of surface-atmosphere exchange?

p. 25340, l. 29 – Mention that this is consistent with your findings of negligibly small O₃ and NO_x gradients above the snowpack.

p. 25341, l. 22 – I agree with your argument that converting the flux to a rate constant requires accounting for the profile of that flux in the boundary layer. It is somewhat misleading, however, to frame this as an “effective layer height” since this number does not represent the height of some physical layer above the surface. Looking at the supplement, this turns out to be a correction factor of ~1/3 applied to the boundary layer height. You might consider presenting the calculation this way, i.e. $k_{3a} = v_{\text{dep}}/(bH)$, where b is a correction factor for the decaying profile of F as derived from similarity theory.

p. 25342 – the term “chemical removal rate” is used several times here to refer to both k_{3a} and k_3 . I recommend calling k_{3a} the deposition rate or something similarly descriptive.

p. 25342, l. 24 – Would you expect potential uncertainties in your estimate of z_{eff} to lead to higher or lower estimates of k_{3a} ?

p. 25343, l. 12 – What fraction of the total loss do you think is due to particles (roughly)? Do you think that you are still missing some of the loss even with deposition and particles accounted for?

p. 25344, l. 1 – This is an important point, and you might work it into your abstract as well.

Technical Corrections

p. 25334, l. 8 – delete “the”

p. 25338, l. 14-15 – Seems to be a sentence fragment.

p. 25340, l. 23 – replace “our” with “are”

p. 25341, l. 5 – replace “form” with “from”

p. S4, l. 6 – Define d.

Cited Literature

- Bertram, T. H., Thornton, J. A., Riedel, T. P., Middlebrook, A. M., Bahreini, R., Bates, T. S., Quinn, P. K., and Coffman, D. J.: Direct observations of N_2O_5 reactivity on ambient aerosol particles, *Geophysical Research Letters*, 36, 2009.
- McNeill, V. F., Patterson, J., Wolfe, G. M., and Thornton, J. A.: The effect of varying levels of surfactant on the reactive uptake of N_2O_5 to aqueous aerosol, *Atmospheric Chemistry and Physics*, 6, 1635-1644, 2006.
- Wolfe, G. M., Thornton, J. A., Yatavelli, R. L. N., McKay, M., Goldstein, A. H., LaFranchi, B., Min, K. E., and Cohen, R. C.: Eddy covariance fluxes of acyl peroxy nitrates (PAN, PPN and MPAN) above a Ponderosa pine forest, *Atmos. Chem. Phys.*, 9, 615-634, 2009.