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Interactive Comment

Interactive comment on "High resolution vertical distributions of NO₃ and N₂O₅ through the nocturnal boundary layer" by S. S. Brown et al.

Anonymous Referee #2

Received and published: 7 November 2006

Recent advances in instrumentation, including those made by the present research group, are now making it possible to probe the spatial variability in reactive trace gases on the same scales as they occur in the atmosphere itself. This work reports field observations of the vertical distributions of NO3 and N2O5 on an unprecedentedly fine scale, and so is a significant advance in the study of variability in nighttime oxidation chemistry. The work shows that concentrations of NO3 and N2O5 can change abruptly for just a ten metre increase in altitude; vertical profiles were also observed to evolve on a time scale of minutes.

The authors show convincing evidence that a large fraction of NOx above the nocturnal boundary layer - in some cases the majority of NOx - is tied up as NO3 + N2O5. This is an important conclusion because, as they say, such a reservoir could be expected to



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store or transport NOx until sunrise (page 9440). But there is an alternative, potentially equally important interpretation. NOx stored in this way might not necessarily be recycled into the atmosphere because N2O5 is the "halfway-house" in the heterogeneous removal of NOx. Thus formation of large amounts of N2O5 could prime the system for rapid NOx deposition if, for example, such an air mass encountered a region of high sulfate aerosol loading (e.g. Brown et al, Science 2006)

I agree with Referee #1's assertion that variability in the anthropogenic NOx source terms are likely more important than variations in NOx deposition & soil emissions. Line 28 of page 9441 states that NOx occurred in "relatively discrete layers", and this is suggestive of NOx plumes impacting the measurements site.

This work should be published with minor revisions.

The authors note the limited duration of their study. Perhaps they could comment on why their measurements were restricted to just one night (4th-5th Oct 2004). I imagine there were significant logistics involved in deploying their instrument from the tower, so I'm curious why measurements didn't continue on other nights (especial when interesting data was being gathered).

Page 9432 line 16 ...influenced by a number of factors. Too vague

Page 9432 line 24 ... NOx is oxidized in the presence...

Page 9433 line 1: Additional references are required (I doubt Brown et al 2006 were the first to propose that NO3 and [mainly] N2O5 can act as nocturnal reservoirs for NOx).

Page 9433 line 16: Friedeberg et al is missing from the reference list.

Page 9435 line 19: Is the uncertainty of the commercial O3 instrument 2% or 2ppbv (or the bigger of the two)?

Page 9436 line 22: The authors need to explain very briefly why potential temperature

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provides a better descriptor of the profiles than the actual measured temperature (page 9435, line 20)? What are the uncertainties in theta?

Page 9437 line 15: the timings given in the text do not correspond with those in Fig 6's caption. Likewise for page 9440 & Fig 7, and 9441 & Fig 8.

Page 9445 line 8: ...with an average difference in relative humidity of 29% between these two heights...

Figure 2: Would the NO3 concentration be better multiplied by a factor of 10 to make it more visible above the baseline?

Figure 2 and elsewhere: is there a significant difference in the time of "sunset" at the top & bottom of the 300m high tower?

The first panel of Figure 4 shows significant NO3 and N2O5 aloft before sunset - comments?

Figure 4: the panels for the first half of the night show distinctly different profiles for NO3 and N2O5. These are discussed together with Figs 6-8, in part, in terms of variations in the NO2 concentration. However the NO3 and N2O5 profiles appear quite similar after 03:30, implying [N2O5] is approximately 10 x [NO3] at all altitudes: why?

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 9431, 2006.

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