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Interactive comment on “Modelling the partitioning of ammonium nitrate in the convective boundary layer” by J. M. J. Aan de Brugh et al.

Anonymous Referee #2

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This paper addresses two related topics, the (lack of) appropriateness of instantaneous equilibrium partitioning for describing the gas-aerosol distribution of nitrate, and the hygroscopic growth and optical properties of aerosol particles with significant nitrate. As the authors state, the relatively increasing importance of nitrate as a constituent of aerosol motivates a better understanding of its properties and behaviour. This is an ambitious paper on a useful topic, however the analysis would be strengthened if the authors could better account for some of their assumptions.

The authors put forth a hypothesis that the lack of consistency between the observed gas fraction of total nitrate, and that predicted by an equilibrium partitioning model, is explained by the atmospheric constituents not being at equilibrium. While this is a plausible situation, they have not eliminated other potential explanations. For exam-

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ple, inaccuracies in the measurements of total ammonium could significantly influence the predicted partitioning. The presence of organic material in the particles, which is not accounted for in ISORROPIA, may influence their water content, and change the gas fraction. Most importantly, the presence of mineral cations, e.g. Ca, Na, K, could strongly influence the gas fraction of nitrate and has not been considered. The difference between the aerosol nitrate measurements from the ground-based AMS and MARGA (Fig 7) strongly suggest that refractory nitrate may have been present. To what degree can the equilibrium modelling be improved by including these cations?

The gas fraction predictions described in Section 3.2.2, resulting from delayed equilibrium (Fig 4) and including vertical mixing (Fig 5), are not improved enough to convince the reader that this can be the only explanation. The comparison of the ground-based and aircraft measurements is complicated by the fact that not only are the particles sampled by the AMS and MARGA representative of different size fractions, but they also may have different cations. Thus the information derived from Section 3.2.3 and Figure 7 is unclear. In this section, the authors state that the amount of particle nitrate aloft should be at least as high as that at the surface. However this might not be the case in there is a strong vertical gradient in total ammonium.

Section 3.4 presents results on the optical scattering of particles through the vertical column on the boundary layer. Aircraft observations are used to compare to predictions of the vertical dependence of scattering based on condensable water and ammonium nitrate. The authors argue that a model where the condensation of ammonium nitrate depends on particle surface area rather than volume matches better with observations, but judging from Figures 8 and 9, neither model is especially representative. It is also not clear why the equilibration time scale for water would be significantly faster than for ammonium nitrate. As the authors mention, there are other constituents in the aerosol, e.g. organics, which may also be influencing the vertical distribution of scattering and have not been accounted for. Given these uncertainties, how confidently can the authors use the observations to show that a D2 dependence provides further

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evidence for a lack of equilibrium for ammonium nitrate?

Technical comments The figures are very difficult to understand. The lines representing the data series are too thin and are difficult to tell apart. The fonts for the legends are too small in Figs 4,5, 8 and 10. In Figure 8, there appear to be more lines than are explained in the legend or caption.

P28274, L24 – ‘coarser resolution’ in what sense?

P 28275, L 19 – ‘depends stronger’ should be ‘depends more strongly’

P28275, L21-22 I think it would be more correct to say that nitrate resides in both the gas and aerosol phase while sulphate resides exclusively in the aerosol phase. Ammonium can also reside in both phases.

P28276, L7-8 – There are approaches other than denuder filter packs that can successfully separate the gas and aerosol nitrate, e.g. Trebs et al., ACP, 2004

P28276, L8-9 ‘For continuous measurements, these labour intensive are hardly used.’ The wording of this sentence is unclear. Do you mean that they are rarely used, of that they are hard (difficult) to use?

P28279, L12-24 It seems like these sentences are mostly about the model rather than the observations, so it should be moved to section 2.2

P28280, L7-8 should read ‘The impactor upstream of the DMA...’

P28280, L18 ‘Aerodyne’ should be capitalized

P28289, L21 – ‘At some days’ should be ‘On some days’

References

I. Trebs, F. X. Meixner, J. Slanina, R. Otjes, P. Jongejan, and M. O. Andreae, Real-time measurements of ammonia, acidic trace gases and water-soluble inorganic aerosol species at a rural site in the Amazon Basin, *Atmos. Chem. Phys.*, 4, 967–987, 2004.

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