

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

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We are grateful to the anonymous second reviewer for his or her important feedback. We will utilise this feedback to improve our manuscript for submission to ACP.

The reviewer proposed several explanations for the mismatch between the equilibrium and the observed partitioning: (i) Inaccuracies in the measurements of total ammonium, (ii) Presence of organic material in the particles changing the water content and influencing the aerosols, (iii) the presence of other cations

In the answer to the other reviewer, we already addressed the presence of organic material or other cations. Inaccuracies in the measurements of total-tracers are not expected to be large. As pointed out in the introduction ("Research into ammonium . . . in

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the Netherlands (Morgan et al., 2010)", P28276, L1–11), measurements of total-tracers face less technical difficulties than separated gas and aerosol measurements. Also, we did not conclude a very sensitive behaviour of the partitioning to the concentrations of the total-tracers ("There is no . . . and the partitioning, P28289, L20–21). The concentration of total-ammonium is expected to be the least important in the Netherlands, because there is always an excess of ammonium.

The reviewer asks more discussion in Sects 3.2.2 and 3.2.3, about other explanations that would explain the observations.

In the answer to the other reviewer, we shortly discussed rapid deposition of nitrate by outgassing and quick deposition velocities of nitric acid, for which we found a reference to a presented paper (Nemitz et al., 2009), now cited in Sect. 3.2.3. We will add a discussion about the interaction with other cations at the end of Sect. 3.2.2:

"It should be considered that there may be a systematic overestimation of the nitrate gas phase fraction in our model, because the model does not take the interaction with sea salt into account (where chloride is displaced by nitrate, see Sect. 2.2). Though it is difficult to quantify this effect, this might suggest that a timescale of 1800 s produces good results on 6 May with a well-timed maximum gas phase fraction around 18:00 UTC that is overestimated by 17%."

The reviewer remarks that in Figs 8 and 9, neither model is especially representative. Also, it was asked why water is more quickly equilibrated than ammonium nitrate and how confidently we can state that our ($\sim D^2$)-model is superior to the ($\sim D^3$)-model.

As already discussed when answering to reviewer 1, we think that Fig. 9 indicates that ($\sim D^2$) is better than ($\sim D^3$), because of the variability in b -values and the good match of the a -values.

We expect that water is indeed more quickly equilibrated than ammonium nitrate. The timescale can be defined as the number of transferred molecules required for equilib-

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rium divided by the flux. The number of transferred molecules is in the same order of magnitude for water and for ammonium nitrate, because the aerosols generally contain about as much water as ammonium nitrate. But because the ambient water concentration is much higher than the ambient ammonium nitrate concentration, the water fluxes are expected to be much higher than those of ammonium nitrate. This all results in a much quicker timescale for water equilibration than for ammonium nitrate equilibration.

The reviewer asks to enlarge figures and thicken lines. Especially Figs. 4, 5, 8 and 10. The figures were created for full-width A4 display. Unfortunately, the ACPD-format resulted in much smaller figure. We will increase the line thickness in the graphs with many different lines.

P28274, L24: The reviewer asks in what sense the resolution is coarser in the model mentioned here.

It can be either the spatial grid or the model time step. In both cases, the model is not able to resolve the process we are analysing, so the model relies on parameterisations. Usually, models with a coarse spatial grid also have a long time step. Those are generally global circulation models. For instance, TM5 has five vertical levels between the surface and 850 hPa and a time step of 22.5 min in the most refined zone.

P28275, L19: The phrase 'depends stronger' should become 'depends more strongly'. This has been corrected.

P28275, L21–22: The reviewer remarks that it is more correct to say: "Nitrate resides in both the gas and aerosol phases while sulphate resides exclusively in the aerosol phase. Ammonium can also reside in both phases". Our manuscript uses a salt-approach (ammonium nitrate and ammonium sulphate) and the reviewer prefers an ion-approach (ammonium, nitrate and sulphate).

This part of the introduction ("There are two ... properties seems appropriate", P28275, L17–27) is a comparison between (ammonium) nitrate and (ammonium) sul-

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phate. Ammonium is considered less important, because in our situation (high ammonia concentrations), any cation is neutralised by ammonium, so 'nitrate' can be interpreted the same as 'ammonium nitrate'. This is the reason that we stick to the salt-approach, though the ion-approach is more correct in principle.

P28276, L7–8: The reviewer remarks that there are approaches other than denuder filter packs that can successfully separate (Trebs et al., 2004).

This is a good remark. The MARGA also uses a wet annular denuder instead of a denuder filter pack. The idea of our text is that a denuder is required to correctly separate gas and aerosol. We will also add Trebs et al. (2004) as reference.

"Correct separation between ... denuder filter packs (Schaap et al., 2002)" (P28276, L7–8) becomes:

Correct separation between aerosol and gas phase is only possible with denuders in combination with a filter pack or a Steam Jet Aerosol Collector (Slanina et al., 2001; Schaap et al., 2002; Trebs et al., 2004).

P28276, L8–9: The word 'hardly' caused a confusion, because it looks like hard (difficult).

We will substitute it with 'rarely' to prevent this kind of confusion.

P28279, L12–24: The reviewer proposed to move this to the model section instead of the observations.

This is a good point. We will just say that we use the observed total concentrations to prescribe the model and that we use the observed partitioning to evaluate the model. The abovementioned lines will be added at the end of Sect. 2.2.2, together with the revisions discussed in the reply to the other reviewer.

P28280, L7–8: The word 'of' was forgotten in the phrase "The impactor upstream of the DMA". Has been corrected.

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P28280, L18: The word 'Aerodyne' should be capitalised. This is done.

P28289, L21: "At some days" should be replaced with "On some days". This has been corrected.

References

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 11, 28273, 2011.

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