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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 #1

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General comments

This paper does a fairly good job of describing the discrepancies in gas-particle partitioning from equilibrium aerosol model and surface and aloft observations and discuss how this impacts the modeled boundary layer optical properties. I have the following concerns in the study that should be discussed and quantified when possible; (1) The authors did not consider the potential interaction of other base cations with HNO₃ and it is unclear how this would impact the model biases. (2) The recommendations regarding the use of an aerosol equilibrium model are empirical/or stochastic corrections for applications in the planetary boundary layer. Local conditions are important in a modeling exercise like this and modeling the boundary layer with temperatures and relative humidity as a function of height is more appropriate than picking one representative

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value for each of these variables and should be taking into account when modeling aerosol optical properties in the boundary layer.

Could the discrepancy between the modeled and aerosol observations be due to more stable forms of nitrate that were not included in the model such as NaNO_3 and $\text{Ca}(\text{NO}_3)_2$? Schaap et al 2011 ACP estimated that approximately half of the NO_3 aerosol deficit in the summer at this site could be due to NaNO_3 . ISORROPIA did not have the capability to simulate these more stable aerosols but ISORROPIA version 2 does include this capability. The MARGA system can measure these base cations and this paper could address some of the uncertainty in the model estimates and build upon the work of Schaap et al 2011 by adding this to the modeling exercises.

The results in this manuscript are primarily discussed in a qualitative manner. The authors should discuss these results in a more quantitative manner. Specific examples have been highlighted below.

Specific comments

Page 28275 Lines 23 to 24: The nitrate aerosol equilibrium dependence on relative humidity and temperature was already stated in lines 18-20.

Page 28279 Line 6: What about other base cations that can partition with HNO_3 like Na , Cl , and Ca that may be present in the observations?

Page 28279 Line 11: "we refer to above mentioned reference" Why not just cite it here?

Page 28279 Line 14-15: "to avoid implementation of advection..." Are you assuming that the advection, deposition and emissions are implicitly included in the observed concentrations?

Page 28279 Line 20: "if advection varies with altitude" Advection will vary with altitude near the surface and the top of the boundary layer. How does this introduce error into your model and does that have any implications on your results?

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Page 28280 Line 8: "upper size limit" What is this limit?

Page 28281 Lines 15-16: "For simplicity, this study does not employ a size-resolved aerosols scheme" One would expect the coarse aerosols to reach equilibrium more slowly than the fine mode aerosols. Do you think that this is reflected in your model results?

Page 28282 Line 7: "Evaporation" should be transpiration or "evapotranspiration" and "crop" should be either "canopy" or "stomatal".

Page 28283 Line 9: "the humidity was generally high" How high? Can you provide a range of modeled humidity?

Page 28289 Line 12: Schaap et al. 2010 should be updated here and in the references to reflect the final revised paper.

Page 28290 Line 5: "does never" should be "never"

Page 28290 Line 18: "represented very well" It is difficult to interpolating general statistics from figures. Could you provide some metrics of what is considered "very well", e.g. coefficient of determination, biases, and/or some other descriptive statistic.

Page 28290 Lines 19-20: "underestimate" and "overestimate" - please quantify the model biases here.

Page 28291 Line 2: "in good correspondence with" - How good? Please quantify.

Page 28292 Line 7: "The improvement ... is impressive" By what metric?

Page 28292 Line 10: "diurnal cycles ... are disturbed" by what?

Page 28293 Line 11: "Similar to the observations" How was this quantified?

Page 28293 Line 21-23: "Captured very well", "not as large" and "remarkably low". Please quantify these.

Page 28293 Lines 24-26: Could the low HNO₃ concentrations observed by the MARGA C13238

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be due to a concentration gradient at the surface due to rapid dry deposition of HNO₃?

Page 28295 Line 4: "scattering coefficients are stronger" By how much and compared to what?

Page 28295 Line 12: "mainly due to the lack of organic aerosols in our model" How was this determined?

Page 28295 Line 14: "if we disregard the general underestimation" This should be quantified to show a systematic bias and the model precision in capturing the general trends.

Page 28297 Lines 16-22: "very close to 1.0", ""too high", and "lowest (i.e. worst)" Should be quantified.

Page 28298 Lines 19-20: ISORROPIA was run for an atmospheric column using the surface temperature and RH and values that are more representative of the atmospheric columns resulted in better results. Would running ISORROPIA with the vertical temperature profile result in similar improvements?

Page 28300 Line 25: "quite abundant" How abundant?

Page 28301 Lines 3-4: "With our optical analysis ..." The evidence supporting this statement does not support such a general conclusion. This is an appropriate conclusion for conditions similar to the three days analyzed.

Figure 7: It is difficult to visually interpret how well the model performs from the scatter plot of the observations. This figure could be improved by binning the aloft observations and plotting them as box plots as was done in Figure 8. This will give the reader a better understanding of the distribution of the aloft observations and how that compares to the modeled sensitivities.

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