

## ***Interactive comment on “Size-resolved simulations of the aerosol inorganic composition with the new hybrid dissolution solver HyDiS-1.0 – Description, evaluation and first global modelling results” by F. Benduhn et al.***

### **Anonymous Referee #1**

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This study presents the development and application of a new solver that is used to describe the partitioning of inorganic compounds between gas and particle phase. The novelty of the solver is the fact that it can save computing power by deciding on dynamical and equilibrium approaches. Its application to a global model clearly shows that the new solver can capture the dynamical, time-dependent features of nitrate partitioning that are not properly reproduced by equilibrium assumptions. The manuscript is well written although it is somewhat lengthy at some places. However, I do not have any clear suggestion on how to considerably shorten it since all parts seem essential and show the logical steps from development, over sensitivity studies in a box model

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to application in a global model. I recommend the manuscript for publication after consideration of the comments below.

#### Main comments

- Throughout the manuscript, the particle phase is referred to as ‘liquid phase’. Many recent studies have suggested that there might be multiple liquid phases (aqueous/organic) in an aerosol particle. Therefore, I suggest changing ‘liquid’ to ‘aqueous’ in the text.
- Eq. 4 and p. 8, l. 15: Usually the effective Henry’s law constant  $H^*$  for acids includes the acid dissociation constant  $K_a$ , i.e.  $H^* = H (1 + K_a/[H^+])$ . Eq1 Does the fact that you assume  $H_2SO_4$  as being completely dissociated lead from Eq1 to Eq-4? Would your approach be applicable using Eq1 for acids that are not completely dissociated?
- Currently, the number of interacting species is limited to three. Will it be possible to rigorously extend the solver to more compounds, given the complex composition of ambient atmospheric aerosol?
- The discussion of the model results (both box and global) in Sections 4.2 and 5.3., respectively, are poorly connected to the figures. All figure panels should be labeled a, b, c, etc and the discussion should be tied more closely to the individual panel so that it is easier for the reader to follow. - It is not clear what the ‘entire microphysical box model’ (e.g., p. 28, l. 32; and Table 1) includes. How far is a comparison of the microphysical model meaningful to the chemical model that includes the new solver? Does using the new solver in the microphysical model lead to redundancy?
- The comparison to the efficiency of previous models is only touched on briefly (end of Section 5.4). Given the extensive length of the current manuscript already, I am not asking for a detailed discussion/ However, a brief statement of the ability of the model by Zaveri et al. (2008) to reproduce the dynamical features as opposed to equilibrium assumptions should be added. In general, how well did the model by Zaveri et al., to

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observations?

Minor comments

p. 2, l. 11: At most RH values, the amount of water exceeds the amount of solute mass, e.g. (Liao and Seinfeld, 2005). Do you mean here the increase of particle size due to solute mass and its associated water?

p. 6, l. 3: Do you mean 'the concentration of the dissolving species in the aerosol aqueous phase'?

p. 11, l. 17: This is unclear, in particular for the conditions described here. 1) Make clear that the aqueous phase 'is predicted to lose protons'. 2) One would assume that under the acidic conditions described here, there is a huge excess of protons.

p. 14, l. 26: Are these parameters (activity coefficients, liquid water content, and dissociation) held constant throughout the simulation or during one time step?

p. 16, l. 21: What is the extent to which the pH varies within the iteration? Are these extreme values or are they realistic over the course of a time step, when aerosol might be exposed to ambient conditions?

p. 17, l. 9: The wording is ambiguous, I think. The convergence criterion is stricter; however, the value is DEcreased from 0.1 to 0.01. If I misunderstood this, clarify.

p. 18, Eq.29: Does 'a' have a physical meaning? What is the range of meaningful values for this parameter?

p. 19, l. 18: What are the physical/chemical parameters that determine equilibration time of a dissolving species? Is it e.g. solubility (Henry's law constant) or something else? Could threshold values be given above/below which equilibration is achieved in short time, relatively to the model time steps?

p. 20, l. 13: Does 'a' here have the same meaning as in Eq. 29? If not, choose a different symbol to avoid confusion.

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p. 23, l. 23: Not clear what 'its' refers to here.

p. 23, l. 22 and 24: Spell out ADDEM, MANIC

p. 24, l. 1: What are the particle sizes for the various modes?

p. 24, l. 31: Is the temperature dependence of HNO<sub>3</sub> dissolution very different than that of NH<sub>3</sub>? Can you support this trend by numbers (T-dependence of H)?

p. 26, l. 3ff: Are there any simultaneous measurements of NH<sub>3</sub> partitioning in gas and particle phase that show similar trends of subsaturation?

p. 27, l. 14/15: What is different in terms of (physico)chemical properties of chloride and nitrate vs ammonium that could explain their different behavior?

p. 27, l. 31: 'Particle concentrations' usually refers to mass or number concentration of particles. Do you mean 'the concentration of ammonia and chloride in the aqueous phase'?

p. 29, l. 20: What is included as sulphur chemistry? Both gas and aqueous phase processes? When sulphate is formed in the aqueous phase, does it contribute to the species that are equilibrated between the phases?

p. 32, l. 15: Reword: "... the ambient concentration of ammonia over the Southern Ocean is predicted to be lower by 10-25% than predicted by the equilibrium approach" or similar in order to clarify that you compare two model results.

p. 32, l. 28: Is this very low nitric acid concentration in the Arctic in agreement with observations?

Figure 1, caption: 1) Is the particle size the wet radius? At what RH?; 2) Could you mark the steps 1-3 in the figure (e.g. by shading or vertical lines)?

Figures 6, 7 and 9, 10 should have a somewhat more detailed caption. What does the 10 ppb etc refer to?

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Figure 11: Some of the text in the figure is very blurry, e.g., 'HCl', 'soluble nucl.' In the top panel and 'OH, NO<sub>2</sub>' (?) in the bottom panel

Figure 12: In the caption only two panel (top and bottom) are described, but there are three panels (top, middle, bottom) shown. Clarify and improve caption.

Technical comments

p. 1, l. 15: 'developed' misspelled

p. 3, l. 14/15: acidic property → acidity

p. 3, l. 26: 'serves as the factor that' can be omitted

p. 10, l. 25: should this read '... relative variation of the surface pressure'?

p. 18, l. 27: 'increments' misspelled

p. 15, l. 7: Should it be 'Equations (24)'?

p. 20, l. 8: Should that be 'Eq 32'?

p. 23, l. 12: 'hydrophilic' misspelled

p. 26, l. 29: spell out 'approximately'

p. 28, l. 15: 'concents' = 'concentrations' or 'contents'?

p. 29, l. 5: 'the number' redundant

p. 30, l. 3: 'hydrophilic' misspelled

p. 32, l. 18: 'superposition of..' – something seems missing here

Figure 8, caption: 'nucleation' misspelled

Additional reference

Liao, H., and Seinfeld, J. H.: Global impacts of gas-phase chemistry-aerosol interac-

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tions on direct radiative forcing by anthropogenic aerosols and ozone, *Journal of Geophysical Research: Atmospheres*, 110, D18,D18208, 10.1029/2005JD005907, 2005.

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Interactive comment on *Geosci. Model Dev. Discuss.*, doi:10.5194/gmd-2015-264, 2016.

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