

Interactive comment on “Coupling of organic and inorganic aerosol systems and the effect on gas-particle partitioning in the southeastern United States” by Havala O. T. Pye et al.

Anonymous Referee #2

Received and published: 14 August 2017

General Comments

This study examines the partitioning of inorganic and organic species in the south-east US summer using information from a large number of observational data sets and explores several different modeling frameworks to describe the partitioning of these species with varied levels of sophistication and agreement with observations. The inclusion of organics and phase separation as well as the organic compound composition is shown to be important for determining aerosol acidity and ammonia partitioning. This work is an important contribution to the modeling community's treatment of inorganic and organic aerosol species. The paper is well written and highly detailed, and below

[Printer-friendly version](#)

[Discussion paper](#)



are specific comments to be considered in the revision of this manuscript.

Specific Comments

Page 3, lines 1-3: NAAQS are set for certain particulate precursors (SO₂, NO₂), so is the reference specifically to NH₃ and organic gases?

Page 3, line 18: Please cite NEI version used in CMAQ simulations and clarify “specific when available.”

Page 3, lines 27-29: It would be helpful for the authors to distinguish between the use of forward and reverse modes of ISORROPIA for clarity and consistency with thermodynamic modeling literature.

Section 2.2: How do the AMS measurements used in this study compare to the other AMS measurements made at the same site during the measurement campaign (Hu et al., 2015), which do include nitrate (and chloride). How do SOAS measurements compare with SENEX and SEAC4RS measurements?

Page 5, line 32-page 6, line 1: Clarify the trigger condition; what is the ammonium associated with if $\text{NH}_4^+ \geq 2 \times \text{SO}_4^{2-}$?

Section 2.2 SEARCH ammonia observations: Are these measurements hourly or 24-hr averages and are they only coincident with SOAS AMS observations? Are the measurements sensitive to diurnal variations and possible measurement interferences at cold temperatures/low relative humidity?

Section 3.1: It would be helpful to include a discussion of model biases in ammonium and sulfate, even if RN/2S is okay. Why is there a disagreement in the model bias between IMPROVE and CSN sites for sulfate? Is sea salt sulfate an important contributor to total sulfate in the model and observations along the coasts?

Section 3.1: It is unclear why Na, Ca, K, and Mg used in ISORROPIA's calculations in CMAQ shouldn't be. If the problem is that their concentrations in CMAQ are overesti-

Printer-friendly version

Discussion paper



mated as is pointed out in the text, what can be done? It is a concerning conclusion to draw that CMAQ can't be used to diagnose species partitioning.

Figures S2-S4: It would be helpful to add SEARCH observations to these plots as a supplement to the ratios shown in Figure 1.

Page 7, lines 22-23: Why a 10% overestimate in sulfate if in line 16 there is a 4-5% overestimate cited?

Page 7, line 31-page 8, line 1: Why is NH_x Fp so similar and RN/2S very different between the two ISORROPIA runs?

Page 8, lines 12-14: Visually it doesn't look like NH_x Fp for the two CMAQ particles sizes looks very different. Stating values in the text could help make that point clearer.

Page 8, lines 15-17: If ISORROPIA was not sensitive between two different particle sizes, why was CMAQ?

Page 8, lines 20-21: Could a full CMAQ model run with only NH_4 , SO_4 , NO_3 , Cl included in the thermodynamic calculations be done?

Figure 2: This figure is slightly difficult to interpret given the large amount of information concentrated in it. What significance does the different gray shading have for the box plots – that should be defined in the figure caption. NH_x could be shown on a separate panel to separate some of the information.

Page 8, lines 31-32: If organosulfates were not present in large enough concentrations to explain a 20% bias, are there any other explanations? Also, should cite other organosulfate measurements from SOAS (Budisulistiorini et al., 2015; Hettiyadura et al., 2017).

Page 9, lines 17-20: What is the reason for the differences between this work and Pye et al. (2017), and which work compares better to observations in terms of expected frequency of phase separation?

[Printer-friendly version](#)[Discussion paper](#)

Section 3.4: In discussion of aerosol pH, are there notable differences in aerosol liquid water content between models that would further impact the pH calculations?

Page 12, lines 9-11: The discussion of improvements required for CMAQ is not sufficient, especially because those errors prevented the use of the full CMAQ results in this study. If the current CMAQ thermodynamic partitioning is not usable, it is important to note if so and why.

Technical corrections

Eastern should not be capitalized on page 2 line 28 for consistency with the rest of the text and US should be U.S. on page 8 line 6.

Works Cited

Budisulistiorini, S. H., et al. (2015), Examining the effects of anthropogenic emissions on isoprene-derived secondary organic aerosol formation during the 2013 Southern Oxidant and Aerosol Study (SOAS) at the Look Rock, Tennessee ground site, *Atmos. Chem. Phys.*, 15, 8871-8888, <https://doi.org/10.5194/acp-15-8871-2015>.

Hettiyadura, A. P. S., et al. (2017), Qualitative and quantitative analysis of atmospheric organosulfates in Centreville, Alabama, *Atmos. Chem. Phys.*, 17, 1343-1359, <https://doi.org/10.5194/acp-17-1343-2017>.

Hu, W. W., et al. (2015), Characterization of a real-time tracer for isoprene epoxydiols-derived secondary organic aerosol (IEPOX-SOA) from aerosol mass spectrometer measurements, *Atmos. Chem. Phys.*, 15, 11807-11833, <https://doi.org/10.5194/acp-15-11807-2015>.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2017-623>, 2017.

Printer-friendly version

Discussion paper

