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Interactive comment

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 #1

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This study presented interesting results on how inorganic-organic interactions would influence particle partitioning, based on observation and simulation results from several models. Generally, this paper is comprehensive and well organized, while several concerns should be addressed before publishing.

Major Comments:

(1) In section 3.1, the authors attributed the discrepancy between CMAQ and observations to the inappropriate inclusion of cations from insoluble metal oxides. They further indicated that the overestimation of transition metals could not be avoided even if the dust emissions are closed. However, they did not state clearly whether the dust emis-

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sions were closed in their simulations, and to what extent that would make a difference. In fact, as shown in Fig. 2, CMAQ substantially underestimated ammonia Fp, while the RN/2S are comparable with other models. Does that arise from the overestimated total ammonia emissions, or more from the overestimated non-volatile cations that would bias the aerosol acidity and therefore the gas-particle partitioning? These mechanisms should be better described and quantified.

- (2) In section 3.1, the different RN/2S ratios from CSN and SEARCH networks are attributed to measurement errors. However, the discrepancy is over 33%, which cannot be totally explained by the <20% measurement errors from Nylon filter. Other possible error sources should be discussed, and the influence of the observation uncertainty should also be discussed in subsequent sections.
- (3) In section 3.4, when phase separation occurred, what is the acidity of each phase?
- (4) In section 3.4, the authors claimed that all the high-pH points are due to measurement uncertainty, which is not convincing. Does these points all occur at very low concentrations when the uncertainty is extremely large? Moreover, they mentioned that there were some elevated nitrite episodes, probably from sea-salts. Whether the high-pH points correspond with those episodes should be examined.
- (5) The implication of results shown in this study, and future work directions should be discussed more in-depth. For example, what is the major strength and weakness of current models shown in this study? Should the second organic phase of, say, HOAs, as mentioned in section 3.5, be added in the future?

Minor points:

- (1)In Figure 1, the colors of $0.8\sim1$ and $1.0\sim1.2$ are hardly distinguishable. If this is not on purpose, please change the colormap. Also consider adding 2 panels showing the difference between observed and modeled RN/2S and R+/-.
- (2) The relationship of IMPROVE and SEARCH network should be clarified or unified.

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All through the manuscript the "SEARCH" network is referred to, while in Fig. S1 to S5 "IMPROVE" is used.

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