We thank the referees for their thoughtful and constructive comments. We have addressed the comments (numbered, below), with referee comments in quotes and italics, and our responses in plain text.

**Referee #1**

1. *"p. 2, line 9: "nitric acid, ammonia and nitrate, ammonium" is awkward. The phrasing used on line 29 (and elsewhere) is much clearer*."

   We have revised this to "$HNO_3$-$NO_3^-$, $NH_3$-$NH_4^+$" to be consistent with line 29 and elsewhere.

2. *"p. 3, line 20: delete extraneous word "details""*

   Deleted.

3. *"p. 4, line 11: is "time of light mass spectrometer" correct here, or should it be "time of flight"?"*

   Yes, the typo is corrected.

4. *"p. 4, line 19: define PFA*"

   The definition of PFA is added and the revised sentence is, "A 12 m heated (40 ± 2°C) and insulated 3/8 inch perfluoroalkoxy (PFA) line (Clayborn Lab, Truckee, CA, USA) connects a short (10 cm; 8 m above ground) custom-designed quartz inlet to the QC-TILDAS."

5. *"p. 5, line 5: insert "negative" before "logarithm""*

   Inserted.

6. *"p. 9, line 12: change "if" to "whether the" (or "whether the shift of one pH unit...")"*

   We have revised the sentence to "An analytical calculation of $HNO_3$-$NO_3^-$ partitioning can also be used to assess whether the shift of one pH unit caused by $Na^+$, $Cl^-$, $K^+$ is consistent with observed nitric acid partitioning for $PM_{2.5}$".

7. *"p. 10, line 24: "highly scatter data" is ungrammatical; please revise"*

   "Highly scatter data" is revised to "highly uncertain data".

8. *“p. 12, line 3: could the authors provide a reference or any data on coarse mode inorganic aerosol concentrations during CalNex?”*

   During the CalNex ground study, up to ~3 μm particle composition was resolved by PALMS, but not high enough for evaluating overall coarse mode. A three-year size-resolved $PM_{10}$ study, also conducted in the Los Angeles basin, is added as the reference to support our analysis. The revised sentence is “Of the various locations where we have investigated pH, this study has the highest coarse mode inorganic aerosol concentrations (Sardar et al. (2005) reports levels of ~ 20 μg $m^{-3}$ year round) and is the only one where we have observed this bias (not observed in the eastern US reported by Guo et al. (2016)).”

9. *“p. 15, line 7: Alduchov and Eskridge is not cited in the text.”*

   This reference is removed from the reference list in the main text. It is cited in the supplemental material.

10. *“*Figure 5: consider deleting the word "Measured" from the y-axis label, since theoretical S curves are also shown.”

    Yes, removed.

**Referee #2**

11. *“Before publication, however, one aspect of this area that could benefit from a brief discussion is the non-linear interplay between other factors that could be captured in a regional model that might benefit from this validation. Given the discussion that follows from the approach in section 4.2, this study would really value from such comparisons. For example, the competition between different sizes and role of dilution might deliver interesting insights into the value of these measurements. That in itself presents an interesting paradox given the complexity prescribed to aerosol composition and evolution. I appreciate some aspects of complexity, including LLPS, have been raised, but the general benefit a regional prediction would bring needs discussion or referencing. To decipher exposure to fine particle pH, or even assess impacts on aerosol-cloud interactions, for example, require this next step.*”

    We with several other investigators are working implementing pH calculations in region models, e.g. CMAQ. The thermodynamic model, ISORROPIA-II, is embedded in these regional models so this suggestion is possible, and as the reviewer points out could be very insightful. However, at this point we are simply working on trying to understand the observations to develop datasets for evaluating regional or global models with metrics that have never been used before. At this point we feel this is beyond the scope of this paper.

12. *"Section 4. The discussion on potential for evaporative loss is interesting. I find it odd that characterization of this problem is not more apparent in the literature. Are there any studies that try to quantify this effect using controlled environments? One might consider some focus laboratory studies would add weight to these discussions. Also, if higher temperatures encourage semi-volatile loss, what is the interplay between propensities to form solids over maintaining equilibrium with the gas phase in these conditions?"*

    Thermodenuder studies may be the most relevant to this issue. However, the residence time, temperature gradient (ΔT), and studied species (most as organic species) in those studies are more extreme than this study and typical indoor/outdoor contrasts. The thermodynamic runs based on ambient and heated conditions provide insights on the sample loss degrees, which turned out not to be the main cause for prediction bias. We have added the following text with two references on Page 11 Line 24, "This is consistent with the limited inlet heating and residence times in this study compared to some thermodenuder studies utilizing more extreme conditions (Huffman et al., 2009; Riipinen et al., 2010)." We agree studies at more realistic conditions would be useful.

    Ammonium sulfate aerosols effloresce around 38% RH (Tang and Munkelwitz, 1994). The large amounts of nitrates present in the aerosol and other dissolved electrolytes and organics depress crystallization RH (Seinfeld and Pandis, 2006). Therefore, particles tend to maintain equilibrium with the gas phase before crystallization during sample line heating, which could cause the loss of semi-volatile particle-phase species. Crystallization is not favored during this study considering the high RH even after heating, on average ~52% (campaign average RH was 79%).

13. *"Page 6, Line 21. The authors note that 'The likelihood for phase separation decreases at higher RH and only has a weak dependence on T (Schill and Tolbert, 2013; You and Bertram, 2015).' Is this strictly true? There are some studies that suggest partitioning across all ranges of sub-saturated humidity's: Zuend, A. and Seinfeld, J. H.: Modeling the gas-particle partitioning of secondary organic aerosol: the importance of liquid-liquid phase separation, Atmos. Chem. Phys., 12, 3857-3882, doi:10.5194/acp-12-3857-2012, 2012."*

    The reviewer seems to be referring to SOA formed from α-pinene ozonolysis which exhibits a liquid-liquid phase separation over 30% to 99% RH range (Zuend and Seinfeld, 2012). As this study points out, O:C ratio of the SOA components are ~0.5, which can lead to phase separation up to very high RH. However, the likelihood for phase separation decreases at higher RH; this is especially true for O:C ratios between 0.5 and 0.8. To make our statement more accurate, the sentence is revised to "The likelihood for phase separation decreases at higher RH ($0.5 < O:C < 0.8$) and only has a weak dependence on T (Schill and Tolbert, 2013; You and Bertram, 2015)".

    You and Bertram (2015) found that the SRH (liquid-liquid phase separation RH) varied by 9.7% at most for the 290 K to 244 K temperature variation. Therefore, SRH is only weakly dependent on temperature.

14. *“Please check the resolution of all figures, I found them difficult to read at times.”*

Image quality was compressed saving from word to pdf. The setup has been changed to the highest fidelity.

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