Review of "Influence of mineral dust and sea spray supermicron particle concentrations and acidity on inorganic NO_3^- aerosol during the 2013 Southern Oxidant and Aerosol Study" by H. M. Allen, et al.

General Comments

This study describes the results of gas and aerosol sampling in the southeastern U.S. during the SOAS campaign. The focus is on aerosol nitrate and its occurrence during several mineral dust/sea salt events. I think that overall, this work is highly novel and will be of interest to many in the atmospheric chemistry community. Among their findings, those that stood out as particularly interesting were: 1) the importance of dust and sea salt in the southeastern US – a region not typically thought of as having high contributions from either source, and 2) the observation that differences in measured aerosol nitrate concentrations between techniques may be influenced by minor differences in the cut point sample inlets due to the presence of nitrate in the coarse mode. The second point, especially, seems quite novel (at least to this reviewer), although the authors do not give it a great deal of discussion. I think with a little more analysis, this could have more substance (for example, comparison of their MARGA nitrate measurements ($PM_{2.5}$) to AMS nitrate (PM_1) measurements).

These plaudits aside, there are some issues with the article that need to be addressed before it is suitable for publication in *ACP*. These are detailed in both sections below:

Specific Comments

1. I have two concerns with the sampling methodology used for the MARGA system. First, why were two cyclones deployed in series? I am not familiar with such a sampling configuration. Cyclones can have a relatively high pressure drop, so I wonder if this could affect the cut point efficiency of the second cyclone? Are the results in Figure S6 taken with both cyclones deployed?

2. My second concern is more significant: pg. 13833 ln 25-26 describes a 1.5 m segment of polyethylene tubing used for the MARGA sample inlet line. I would expect this material to cause significant particle losses in this segment of the sample line. Did the authors consider this effect, and if so, what was the magnitude/impact on the aerosol measurements?

3. I have two questions about the inorganic modeling (Section 3.6):
    a. for the E-AIM simulations, how were the components not included by the model accounted for? If the calcium/magnesium/potassium equivalents were input as H^+ (to meet the model's requirement of charge balance on input), I would expect that to have a very different impact on predicted nitrate than the same simulations treating crustals as equivalent sodium, as other studies have done. This may help to explain the significant difference in E-AIM predictions of aerosol nitrate.
    b. Perhaps I am just misinterpreting Figure 7d, but there seems to be a major disconnect with the E-AIM model results. The total ($HNO_3 + NO_3^-$) measured nitrate concentration appears to be significantly higher than the total modeled nitrate concentration. Even if E-AIM is partitioning a disproportionate fraction to the gas phase, why is there not a material balance on $HNO_3 + NO_3^-$? Note, for ISORROPIA, the total modeled and measured nitrate appear to be quite similar.

4. In Section 3.5, the authors support their claim for nitrate production on the surface of the dust particles by noting that the rate of HNO_3 uptake is controlled by the enhanced surface area contributed by the larger particles. However, the authors should also discuss how the *rate* shown in Figure 6d connects to the observed aerosol nitrate concentration in Figure 6a. Further, the authors state that "higher rates of aerosol NO3- production track more closely with higher Sa than with other factors that contribute to NO3-" – this would be supported by showing gas-phase NH_3 data. Similarly, in the instances where the authors suggest chloride displacement was the major route for HNO_3 uptake – e.g., Section 3.3 – the argument would be strengthened by showing and discussing the MARGA HCl data.

5. Pg. 13844, line 16-19: there are many instances throughout the paper (this sentence is one example) where the authors need to be more careful with their notation. This sentence gives the impression that 'coarse mode particles' is applied to PM > 1 μm.

6. Pg. 13831, line 6: in 'highly acidic environments' with excess sulfate, I would not say that the NH_4NO_3 dissociates, as much as the limited NH_3 preferentially associates with sulfate, leaving HNO_3 in the gas phase.

7. Pg. 13832, line 2-3; Pg. 13841, line 6-9: the authors are referred to other studies which suggest much longer equilibration timescales than the authors assume here. For example, Fridlind and Jacobson (*JGR*, 2000) model HNO_3 equilibration timescales with 2-3 μm sea salt particles that are ~5-20 hours, with considerable variation dependent on α.

8. Section 3.4: the discussion in this section gives the impression that all particles/particle types were externally mixed? Is that the case?

9. Pg. 13846, line21-23: what is meant by this sentence?

Technical Corrections

1. Abstract, line 3-4: suggest removing "1 June to 15 July"

2. Abstract, line 5-7: suggest removing "an ion chromatograph coupled with a wet rotating denuder and a steam-jet aerosol collector for monitoring of ambient inorganic gas and aerosol species"

3. Pg. 13830, line 6: what is meant by 'a dominant pollutant'?

4. Pg. 13830, line 7-8: run-on sentence

5. Pg. 13830, line 10: I would say regulators or regulatory agencies are also quite interested.

6. Pg. 13830, line 23: Temperature and aerosol water also strongly affect aerosol nitrate concentrations.

7. Pg. 13832, line 12: change 'a glimpse' to 'insight'

8. Pg. 13834, line 18: say ‘chemical suppressor’ instead of ‘suppressor ion exchange columns’
9. Pg. 13835, line 24: do the authors mean ‘biomass burning’?
10. Section 2.2.2: give the range of particle size analyzed by this method
11. Note that Guo et al. (2014) reference is now an ACP article
12. Pg. 13839, line 26: note that the cited values from Guo et al. are modeled using ISORROPIA, not ‘measured’.
13. Pg. 13840, line 1-2: say ‘inorganic ionic species’
14. Pg. 13840, line 26: see Specific Comment #5 above
15. Pg. 13841, line 21: change ‘suggest’ to ‘suggests’
16. Pg. 13842, line 3-5: are the fractions (45.2% and 41.8%) statistically different?