

Interactive comment on “Airborne and ground-based observations of ammonium nitrate dominated aerosols in a shallow boundary layer during intense winter pollution episodes in northern Utah” by Alessandro Franchin et al.

Anonymous Referee #2

Received and published: 7 October 2018

This paper deals with airborne and ground-based measurements of aerosol concentrations, chemical composition and gas phase precursors in three valleys in northern Utah (U.S.A.), obtained in winter 2017. It shows that increases in total aerosol mass “above $\sim 2 \mu\text{g m}^{-3}$ were associated with increases in the ammonium nitrate mass fraction, clearly indicating that the highest aerosol mass loadings in the region were predominantly attributable to an increase in ammonium nitrate.” The study shows a generally nitric acid (HNO₃) limited regime for the Cache and Utah Valleys, and a mixed regime (HNO₃ and NH₃) limited regime downwind of Salt Lake city. The inorganic species

C1

were compared with the ISORROPIA thermodynamic model. Total inorganic aerosol mass concentrations were calculated for various decreases of total nitrate and total ammonium showing the combined total nitrate and total ammonium decreases were most efficient to reduce ammonium nitrate (so despite the prevailing HNO₃ limitation).

The paper is a valuable contribution to ACP, because it gives a clear and sound estimate of the ammonium nitrate contribution to winter PM_{2.5} and of its limiting precursors.

The paper could be further improved if the following remarks were taken into account :

In the Introduction (page 3, line 5), the authors write “those studies suggested that nighttime nitrate formation within the residual layer was a major contributor to surface level PM_{2.5} concentrations.” I understood that investigating this type of processes was one of the goals of the campaign. But little is said in the paper about the origin of ammonium nitrate precursors, within the study region, or outside. During winter time, how quickly HNO₃ would be formed from NO_x emissions. May be the observational data set does not allow such a discussion, or it is foreseen for another paper. Authors should please state something about that. If the origin within the study region is most important, it would be interesting to show NO_x and NH₃ emission maps.

The authors state that other cations as Ca²⁺ or Mg²⁺ could bound a major part of nitrate. Although this is excluded of total nitrate as defined in the paper, it would be interesting to know what Ca²⁺ or Mg²⁺ levels could be typically expected.

How do meteorological conditions affect the isorropia calculated equilibria ? Extending the discussion would be interesting. Is aerosol expected to be liquid or solid ?

In Figure 7b, Salt lake city: The potential temperature profile decreases with altitude. This is not possible in this extent. Are potential temperature and temperature mismatched? In figure 8, another profile is shown.

Minor remarks :

Introduction: In Europe Petetin et al., 2016 performed a similar study as the one pre-

C2

sented, although based on daily measurements and for an urban environment. This could be cited.

Page 5, line 4 : "It is important to note that refractory species such as sea and / or lake salt (mostly sodium chloride), road salt (mostly magnesium chloride), dust (mostly alkali salts and silicon oxides), and black carbon (from diesel exhaust or wood combustion) are not routinely measured with the AMS" The sentence is not clear. I think you mean that they have not been measured during the campaign.

Page 6, line 27: "but zero uncertainty can be as large as 0.2 ppbv" I wonder what are the reasons for this ? Could it be due to HNO₃ sticking on inlet walls, and getting desorbed ?

Page 8, line 24 "During the first pollution episode, both the aircraft and ground-based aerosol mass concentrations were the highest in Cache Valley (~70 μg m⁻³) and the lowest in Utah Valley (~10 μg m⁻³). During the second pollution episode, the highest aerosol mass concentrations were observed in Utah Valley (~70 μg m⁻³) and varied for Salt Lake and Cache Valley over the course of the episode (40 – 90 μg m⁻³). During UWFPS, PM_{2.5} was on average around 70% of PM₁₀ in the three valleys (Figure S3)." How to explain these differences between valley concentrations for different episodes be explained ?

Page 8, line 30: Please define \hat{A} total aerosol mass \hat{Z} , probably PM₁ non-refractive species measured by AMS.

Page 9, line 20: "The scatterplot gives an average slope of 1.8 g cm⁻³, which is comparable to the density of ammonium nitrate (1.72 g cm⁻³), within the combined measurement uncertainty." But it could be consistent, within the measurement uncertainty, with the density of other aerosol species too, isn't it ?

Page 9, line 23: "Those correspond to measurements in the plume of the U.S. Magnesium plant (Figure S1) where we observed a high organic (74.0±1.2%), high chloride

C3

(6±0.6%) and low ammonium mass fraction (3±0.6%)." What are the typical emissions of such a factory ?

Page 13, line 27 "Reduction in total nitrate led to approximately a proportional decrease in total aerosol concentrations during polluted conditions and across all locations." Is total aerosol or total nitrate meant ?

Page 15, line 6 : "The PM_{2.5} mass loadings for the region varied from below the AMS detection limit of 0.38 to 72.4 μg m⁻³ (2nd and 98th percentiles)." This suggests that AMS measures PM_{2.5} or that PM_{2.5} was derived from AMS. Please clarify from which type of measurement PM_{2.5} loadings have been calculated here (Surface ?).

Wording , typos :

Page 2, line 2: "lead" => "leads"

Page 5, line 28 : I would say \hat{A} reactive nitrogen species \hat{Z} instead of "nitrogen oxides \hat{Z} . oxide

Page 8, line 19: Please correct \hat{A} (, which is surrounded \hat{Z}

Page 9, line 30: \hat{A} C₂H₆ \hat{Z} plus put indices.

Page 11, line 33 : \hat{A} slightly \hat{Z} => "slight"

References : Petetin, H., Sciare, J., Bressi, M., Gros, V., Rosso, A., Sanchez, O., Sarda-Estève, R., Petit, J.-E., and Beekmann, M.: Assessing the ammonium nitrate formation regime in the Paris megacity and its representation in the CHIMERE model, *Atmos. Chem. Phys.*, 16, 10419-10440, <https://doi.org/10.5194/acp-16-10419-2016>, 2016.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2018-629>, 2018.

C4