Answer to Referee # 2

We thank Anonymous Referee # 2 for their comments. Here below are the Authors' answers point by point)in blue).

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 PM2.5 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, kow qicly HNO3 would be formed from NOx 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 NOx and NH3 emission maps.

The Referee is correct. Identifying the sources was indeed one of the objectives of the UWFPS measurement campaign. However, the scope of this manuscript was to 1) characterize the chemical composition of the aerosol particles and 2) investigate the effects of the decreases of total nitrate and total ammonium in the region. In other words "to achieve a better understanding of the processes that drive the conversion of precursor vapors into aerosol particles," as we wrote in the introduction (page 3 line 26).

Other manuscripts such as Moravek et al., (in preparation) will discuss the sources of NH3 in the region, showing the NH3 emission maps from the inventory and comparing them to the Twin Otter measurements. The paper will focus on NH_3 sources, but will also touch on NOx sources in the region. In addition, a second manuscript by McDuffie et al (in preparation) will investigate the contribution of nocturnal heterogeneous reactive nitrogen chemistry to particulate matter formation. A third paper by Womack et al (under review) explores NOx and VOC control as mitigation strategies for PM. Unfortunately, all these manuscripts are not published yet and therefore not possible to cite properly in the current paper.

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.

We have ion chromatography measurements of gas and aerosol phase at the University of Utah site that show presence of Ca^{2+}, Na^{+} and K^{+} in $PM_{2.5}$ (Mg^{2+} was always close to the detection limit ~0.01 ug/m^3). The average concentrations were 0.14 $\mu g\ m^{-3}$ (max 1.2 $\mu g\ m^{-3}$) 0.09 $\mu g\ m^{-3}$ (max 3.6 $\mu g\ m^{-3}$) and 0.25 $\mu g\ m^{-3}$ (max 1.6 $\mu g\ m^{-3}$) respectively. The mass fractions of these species were always less than 10% of the total PM2.5 when the PM2.5 was > 20 ug m^{-3}). Ca2+ was always less than 3% and usually less than 1% for the high PM2.5 mass periods. The results of those measurements will be presented in detail in a separate manuscript by Hrdina et al. (in preparation).

How do meteorological conditions affect the isorropia calculated equilibria ? E4xtend the discussion would be interesting. Is aerosol expected to be liquid or solid ?

We agree with the Referee that it would be interesting to explore how the meteorological variables and aerosol phase affect the equilibria from the ISORROPIA model. However, we think that such a discussion is out of the scope of the paper. Cold temperatures and high relative humidity generally favor the formation of ammonium nitrate in the aerosol phase in both the model and our observations. In the area studied during UWFPS the median RH and temperature were 0.66 and 271 K, respectively, and the phase of the aerosol is unknown at these conditions.

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.
The referee is correct the temperatures presented in Figures 7 are 8 air temperatures, not potential temperatures. We corrected the labels in both figures. In Figure 8 different profiles are shown because those are averages over polluted periods, not single missed approaches as in Figure 7.

Minor remarks :
Introduction: In Europe Petetin et al., 2016 performed a similar study as the one pre-sented, although based on daily measurements and for an urban environment. This could be cited.
We added the citation

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.
We changed the sentence into “It is important to note that during the campaign 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) were not measured.”

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?
The reasons to the uncertainty being so large at times is related to a minor pressure-dependence in the background zeros in the NO channel. We added that information in the text. Inlet effects, such as HNO3 sticking on inlet walls, would not be relevant for the NO, NO2, O3 channels, as those channels are “blind” to HNO3.

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, PM2.5 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 ?
We added at page 9 line 5: “”These variations among valleys in peak PM2.5 concentrations are characteristic and are due to variations in sources and meteorological processes (Baasandorj et al, 2018) (Figure S2).”
We further modified the prior sentences, taking surface based measurements during non-flight times into account, to: “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 ~90 µg m-3 respectively) and the lowest in Utah Valley (~10 µg m-3 and ~25 µg m-3 respectively). During the second pollution episode, the highest mass concentrations observed at the ground sites in Cache Valley were up to 100 ug m-3, in Utah Valley were ~70 ug m-3, and in Salt Lake Valley were up to 60 ug m-3.”
Page 8, line 30: Please define Â´n total aerosol mass Â˙z, probably PM1 non-refractive species measured by AMS.
We modified the sentence to “For the purposes of this analysis, we define clean conditions, when the total aerosol mass ($NR\text{-}PM_1$) is ≤ 2 µg m^{-3} and …”

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 ?

That is correct. However, ammonium nitrate is mentioned here because it is the largest component of the aerosol as the AMS measurements show.

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 (6±0.6%) and low ammonium mass fraction (3±0.6%).” What are the typical emissions of such a factory ?
The U.S. magnesium plant it is the largest producer of primary magnesium in North America. It has a natural gas power plant on site and a processing plant. According to our measurements, it emits a large amount of aerosols, chloride and other halogens in the gas phase.
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 ?
As written, we meant total inorganic aerosol. We changed the sentence to be more precise into “Reduction in total nitrate led to approximately a proportional decrease in total inorganic aerosol concentrations during polluted conditions and across all locations”

Page 15, line 6 : “The PM2.5 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 PM2.5 or that PM2.5 was derived from AMS. Please clarify from which type of measurement PM2.5 loadings have been calculated here (Surface ?).
The referee is correct. It should be NR-PM$_1$. We corrected the sentence.

Wording , typos :
Page 2, line 2: “lead” => “leads”
Corrected

Page 5, line 28 : I would say Â´n reactive nitrogen species Â˙z instead of ”nitrogen oxides Â˙z. oxide
We decided to leave the sentence unchanged, as by using “reactive nitrogen species” the sentence would result less specific.

Page 8, line 19: Please correct Â´n (, which is surrounded Â˙z
Corrected

Page 9, line 30: Â´n C2H6 Â˙z plus put indices.
We changed the sentence from “… showed a contribution from organic fragments, probably CH_2O or C2H6.” To “… showed a contribution from organic fragments.”

Page 11, line 33 : Â´n slightly Â˙z => “slight”
Corrected

References

Baasandorj, M., Brown, S. S., Hoch, S., Crosman, E., Long R., Silva, P. Mitchell L., Hammond I., Martin, R., Bares R., Lin, J., Sohl J.,Page, J., McKeen, S., Pennell, C., Franchin, A., Middlebrook, A., Petersen, R., Hallar, G., Fibiger, D., Womack, C., McDuffie, E., Moravek, A., Murphy, J., Hrdina, A., Thornton, J., Goldberger, L., Lee, B., Riedel, T., Whitehill, A., Kelly, K., Hansen, J., Eatough, D., 2017 Utah Winter Fine Particulate Study Final Report, 2017.https://www.esrl.noaa.gov/csd/groups/csd7/measurements/2017uwfps/finalreport.pdf (Last accessed June 2018)