

## ***Interactive comment on “Free Tropospheric Aerosols at the Mt. Bachelor Observatory: More Oxidized and Higher Sulfate Content Compared to Boundary Layer Aerosols” by Shan Zhou et al.***

### **Anonymous Referee #1**

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This manuscript describes high resolution aerosol chemistry measurements made at the mount bachelor's observatory (MBO) during two periods in July and August of 2013. Aerosol chemistry and size distributions were measured using a high resolution aerosol mass spectrometer, and were operated alongside instruments characterizing gas-phase properties (NO<sub>y</sub>, CO, O<sub>3</sub>), and aerosol optical properties. As a result of its altitude and strong boundary layer dynamics the MBO site is often located in the free troposphere at night, providing the authors the opportunity to characterize FT composition. The objectives of this work were to describe the chemical and physical properties of FT aerosols and compare with those from the BL. This work is well prepared and described. The data set is a bit short and variable to generally describe FT aerosol

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properties, but measurements at altitude sites are important to obtain a signature of FT aerosols.

General comments 1. How stable were the air mass sources during these two FT periods?

2. Page 5, Line 151: As discussed later on in the manuscript, this also suggest a supplementary source of sulfate aerosol particles in the FT, possibly from nucleation processes. Although no difference in the total mass of sulfate was observed during the day and night, were there differences in the signal ratios of the sulfate measured during the day (SO<sub>4</sub>\_BL) vs night (SO<sub>4</sub>\_FT).

3. The authors mention that the signal ratios of the sulfate peaks are similar to that of MSA. It is also mentioned that several sulfate peaks are associated with organic aerosols. Were there any trends in the PMF analysis that segregated the sulfate aerosol by diurnal patterns (the more oxidized OA vs the sulfate dominated OOA)? Were the signal ratios of the PMF sulfate factor similar to that of ammonium sulfate or similar to other types of sulfate.

4. From the PMF(ORG+INORG), were nitrate peaks associated with any of the organics, and was the PMF analysis capable of extracting an organic nitrate (ON) factor that could be compared to the ON extracted using the Farmer et al., method?

5. As shown in recent publications at both JFJ (Hermann et al., 2015) and at the PdD (Farah et al., 2018), the properties of FT aerosols can vary considerably depending on its last contact with the boundary layer. Similar analysis of air mass back trajectories on this data set would provide clear characteristics of FT aerosols at the MBO.

6. In the Hermann and Farah studies, the change in the size distribution for FT aerosols has been used as a tracer for FT air masses. In Figure 5, only average size distribution for the whole period are shown. Can the authors include the average size distributions for FT periods? Why are organic size distributions in the FT not included here? Can

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they be magnified? or are the signal to noise ratios too low?

Herrmann, E.; Weingartner, E.; Henne, S.; Vuilleumier, L.; Bukowiecki, N.; Steinbacher, M.; Conen, F.; Collaud Coen, M.; Hammer, E.; Jurányi, Z.; et al. Analysis of long-term aerosol size distribution data from Jungfraujoch with emphasis on free tropospheric conditions, cloud influence, and air mass transport. *J. Geophys. Res. Atmos.* 2015, 120, 9459–9480.

Farah, Antoine; Freney, Evelyn; Chauvigné, Aurélien; Baray, Jean-Luc; Rose, Clémence; Picard, David; Colomb, Aurélie; Hadad, Dani; Abboud, Maher; Farah, Wehbeh; Seasonal variation of aerosol size distribution data at the Puy de Dôme station with emphasis on the boundary layer/free troposphere segregation, *Atmosphere*, 2018, 9, 7, 244.

7. For the comparison with other “altitude” sites, the authors could also take into account measurements from:

Minguillón, M.C., Ripoll, A., Pérez, N., Prévôt, A.S.H., Canonaco, F., Querol, X., Alastuey, A., 2015. Chemical characterization of submicron regional background aerosols in the western Mediterranean using an Aerosol Chemical Speciation Monitor. *Atmos. Chem. Phys.* 15, 6379–6391. doi:10.5194/acp-15-6379-2015

Ripoll, A., Minguillón, M.C., Pey, J., Jimenez, J.L., Day, D.A., Sosedova, Y., Canonaco, F., Prévôt, A.S.H., Querol, X., Alastuey, A., 2015. Long-term real-time chemical characterization of submicron aerosols at Montsec (southern Pyrenees, 1570 m a.s.l.). *Atmos. Chem. Phys.* 15, 2935–2951. doi:10.5194/acp-15-2935-2015

8. Table S1, Figure 6: Why is there no NR-PM1 mass concentrations for pdD 2012?

9. Figure 6: O/C only 6/16 of the listed stations have values. Why so? It would be interesting to have this sort of overview of measurements? Can this information be obtained through making contact with the different stations?

10. Figure 6: It seems strange that the puy de Dome has such high concentrations

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(higher than those in E.Asia (by a factor of 3!!)). Is this representative of the station or is it a particular period that was advected by unusual air masses.

Minor comments:

Page 2, Line 35: This statement is important and useful, but can the authors include some references based on recent simulations that show the need to improve our knowledge of regional and FT aerosol in order to improve chemistry transport models?

Page 6, Line 187, and Page 7, Line 239 (and elsewhere): Can you provide the m/z values for each of these fragments.

Page 7, Line 226: The m/z 91 can also be associated with fragments of primary anthropogenic OA. How can the authors be sure that this fragment is solely biogenic? It has previously been illustrated that the situation of the f44 and f43 on the triangle plot could indicate the source/type of the aerosol, with biogenic aerosols being situated on the right hand side of the triangle (Jimenez et al., 2009; Ng et al., 2010). Can this be illustrated and commented in Figure S13.

Figure 1: Can the authors identify the FT periods in Figure 1.

Figure 2: NR-PM1 Composition. What does the y-axis represent here, I presume fractional contribution. Here we see that, at night, in the FT, more than 90% of the particle composition is SO4 aerosols. In

Figure 1, we observe this to be the case in only 2 of the 9 nights. Is this figure an average of all data shown in Figure 1?

Figure S13: The main text refers to LV-OOA and SV-OOA, but in this figure the OOA are labeled BL-OOA and FT-OOA.

Jimenez, J.L., et al. Evolution of organic aerosols in the atmosphere. *Science* 326, 1525–9. doi:10.1126/science.1180353

Page 8, Line 264: Please include the ratios expected for acidic and for neutralized

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conditions. Why not include the NH<sub>4</sub> predicted to NH<sub>4</sub> measured plots?

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