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Comment on acp-2022-477

Anonymous Referee #1

Referee comment on "Measurement report: Intensive biomass burning emissions and rapid nitrate formation drive severe haze formation in the Sichuan Basin, China – insights from aerosol mass spectrometry" by Zhier Bao et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2022-477-RC1, 2022

The manuscript by Bao et al. present detailed observations of the chemical composition of PM2.5 in the Sichuan Basin (SCB), and the component responsible for the formation of haze during Winter. The PM2.5 composition on site is driven by gas phase and aqueous-phase oxidation for nitrate, aqueous phase formation for sulfate, primary emission from Biomass burning and vehicle emissions and nitrate formation influenced by biomass burning. During fog events, primary organic aerosols were scavenged while secondary aerosol formation was enhanced by aqueous-phase reactions. The method applied and the case studies presented provide valuable knowledge on the species and mechanisms leading to haze and fog events in the SCB, but some restructuring and improvement in the discussion need to be addressed before publication.

General comments:

- Page 11 lines 296-316: The discussion about night time nitrate formation is a bit confusing or not well constructed. During daytime, you attribute the nitrate formation to homogeneous reaction based on the fit of NO3/SO4 and NH4/SO4 molar ratios. Then, for night time, you conclude that aqueous reactions dominate nitrate formation based on the increasing trend of NO3 with ALWC. Then you justify not considering the fitting approach used for daytime based on the fact that NOx and SO2 emissions decreased and NH3 emissions increased. Is the RH high throughout night time? Couldn't it mean that you have both homogeneous and heterogeneous reactions occurring? If you can show that "HNO3 was firstly heterogeneously formed through the hydrolysis of N2O5, then excess NH3 was uptake by wet particles and neutralised HNO3 forming ammonium nitrate" dominate nitrate formation at night, then I would simply not mention the night time NO3/SO4 vs NH4/SO4 fitting. I suggest mentioning why it is not applicable first, and then talk about the aqueous reactivity because this could lead the reader to doubt the fitting relevance during daytime as well.
- The regional transport discussion should be moved prior to the "Case studies for haze

pollution" section as the content does not provide specific details or information that contribute to a better understanding of the haze episodes. And "Evolution of chemical composition during fog periods" would probably correspond more as the second subsection of "Case studies for haze pollution".

Minor comments on manuscript:

- Page 1 lines 21-23: "The fine aerosol chemical composition was characterised by using a time-of-flight aerosol chemical speciation monitor (ToF-ACSM) with the aim of inorganic and organic aerosol characterisation and source apportionment." Please, rephrase.
- Page 1 line 25: Please choose a more appropriate word than "occupied"
- Page 3 lines 64-66: "The emission of SO2 had been reduced dramatically over the past ten years in China; however, NOx did not show a significant reduction." Please add references to support these trends.
- Page 3 line 68: "Compared to SIA, the formation process of SOA was more complicated (Chen et al., 2017)." Which formation process are you referring to? Or are you referring to SOA formation in general and therefore it includes multiple processes/pathways… As described later in the paragraph.
- Page 4 line 82: "was also suffering severe haze pollution", if it is still happening, I would use present continuous tense.
- Page 5 line 148: "~84 cc/min" for consistency with previous flow (line 141) you should either write the equivalent value in L/min
- Page 7 – Data Process section: information on the elemental analysis with the TOF-ACSM is lacking.
- Page 10 line 7: "planet boundary (PBL) height" I assume the "layer" is missing in that sentence.
- Page 10 line 261: You mention biomass burning as a source of Chloride. Any idea of fuel used or burning conditions?
- Page 11 line 275: "If $[NO_3^-]/[SO_4^{2-}]$ linearly correlated with $[NO_3^-]/[SO_4^{2-}]$ under ammonium-rich conditions", shouldn't it be linearly correlated with "$[NH_4^+]/[SO_4^{2-}]$"?
- Page 13 line 348: You mentioned that chloride is a biomass burning tracers and that these BB could be related to cooking and heating. Which of these sources would emit Cl^-?
- Page 14 lines 369-373: "The average OOA concentration did not change significantly with increasing ALWC during daytime, suggesting the less contribution of aqueous state reaction to the formation of OOA. During nighttime, the average OOA concentration showed an increasing trend when ALWC < 200 μg/m3 and kept relatively constant subsequently, suggesting the aqueous-phase reactions did not significantly affect the formation of OOA" You can maybe shorten this part by saying that aqueous reactions are not significant pathway toward OOA formation during day- and night-time.
- Page 15 lines 403-405: "Higher RH was observed for those data points within the region of aged BBOA in the f44 vs. f60 space". Although, I agree that BBOA oxidation probably occurs in the aqueous phase, in Figure 8, it seems that the RH is high for most of the points falling in the f44 vs f60 triangle, except for the data with f44 > 0.15 and 0.08>f60>0.05, where the RH seems lower. Also is there a reason behind using RH here instead of ALWC as used in the previous comparison?
- Page 16 line 428: change "Table S2" to Table S3.
- Page 17 line 460 and after: as a cluster represent several "air masses", plural form is probably more adapted, especially that you use "air parcels" later on in the paragraph.
- Page 18 PSCF discussion: more details about the threshold value used could be added

in the text or in Figure 12.

- Page 20 lines 534-540: "The average elemental O:C showed an increasing trend from pre-fog periods to post-fog/foggy periods, while H:C did not change significantly for different fog events, suggesting the OA became more oxidised. As shown in Fig. S6, the mass fractions of OOA increased, while the contribution of BBOA and HOA decreased from pre-fog periods to post-fog/foggy periods for the three fog events. As a consequence, the O:C increased in line with the increased contribution of OOA." The O:C and H:C could be added to Figure S6
- Figure 2 would benefit from a different (perhaps lighter) background as the yellow makes it difficult to distinguish between SO4, NH4 and Chl.
- Figure 3: As you discuss day/night time nitrate formation and the effect of RH at night, could you perhaps add RH diurnal variation. Or a figure with the diurnal cycles of meteorological parameters and PBL could be added in the supplement.
- Figure 6: it would be helpful to add some background to evidence the fog periods on the time series of the OA sources.

Minor comments on supplement:

- Page 3 Figure S2: It may be easier to use a lighter blue for nitrate as the mean is hard to distinguish. Could the dataset be separated between day/night time as it supports the discussion between secondary inorganic aerosol day/night formation?