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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric aerosol compositions and sources at two national background sites in northern and southern China" *by* Qiao Zhu et al.

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1. The introduction part is clearly not comprehensive and valuable. First, as the application of AMS in China increased significantly in recent years, the authors should do a bit more through summary of the current status; More recent studies should be mentioned, for example, AMS studies conducted in Beijing by Sun yele's group, in Lanzhou Atmos. Chem. Phys., 14, 12593-12611, 2014., and a more recent SP-AMS study in Nanjing (Environmental Science & Technology Letters, 3, 121-126,2016) and more. Secondly, if this paper intends to discuss the aerosol characteristics from background sites, the authors should summarize previous studies and major findings regarding the aerosol chemistry in background sites, and this should not be limited in

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china, but all over the world. What is the difference between the background sites and urban/polluted sites, and then what new findings do we expect in this manuscript? REPLY: We add more relevant descriptions in the introduction part. For more complete summarization of AMS studies in China, as below: Since 2006, valuable insights on the composition, sources, and evolution processes of submicron particles in China were obtained through a dozen of field campaigns using various types of Aerodyne aerosol mass spectrometer (AMS) instruments, capable of on-line measuring chemical composition of non-refractory submicron aerosol species (Canagaratna et al., 2007; Ng et al., 2011b). These previous campaigns mostly focused on much polluted areas in eastern China, such as the Beijing–Tianjin–Hebei area (Takegawa et al., 2009; Huang et al.,2010; Sun et al., 2010, 2012, 2013, 2015; Zhang et al., 2011; Hu et al., 2013), the Yangtze River Delta (Huang et al., 2012, 2013), and the Pearl River Delta (He et al.,2011; Xiao et al., 2011; Lee et al., 2013),Xu et al. (2014) reported the chemical composition, and size distribution of submicron particulate matter (PM1)in Lanzhou in northwest China. In addition, Wang et al. (2016) recently used an Aerodyne soot particle-aerosol mass spectrometer (SP-AMS), for the first time in China, to investigate the occurrence of fullerene soot in ambient air. For summarization of AMS background site studies, as below: So far, several measurements and source analyses based on AMS have been conducted at background sites around the world. Sun et al. (2009) reported the composition and size distribution of NR-PM1at the Whistler Peak in Canada. Chen et al. (2009, 2015) conducted an AMS study to characterize submicron biogenic organic particles in the Amazon Basin. Ovadnevaite et al. (2011) demonstrated the occurrence of primary marine organic aerosol plumes on the west coast of Ireland. Du et al. (2015) described the aerosol composition using an ACSM at a national background monitoring station in the Tibetan Plateau in western China. These background site aerosol studies were all conducted in remote areas, which represent for global background atmosphere rather than regional background atmosphere, while regional background atmosphere is more critical to reflect the general picture of anthropogenic emissions in a hot polluted region. In this study, we performed online aerosol mea-

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surement field campaigns at two national air background sites in both northern and southern region in eastern China, which has a population of more than one billion and is characterized by worldwide high air pollution levels under high urbanization and industrialization.

2. As there already are so many AMS papers published in the past 15 years, it is difficult to see what is the significance and novelty of this paper. This should be made more clear, the novelty should not because you did AMS measurements at sites that are different from others, but instead you should state what scientific questions and what valuable findings you gained from your measurements that can advance our current understanding on aerosol chemistry? REPLY: Although many AMS measurements were conducted in the past years, this study also offers aerosol properties at two unique sites, which are regional background sites and critical for understanding the general picture of anthropogenic emissions in a hot polluted region, i.e., eastern China, where more than 90% of population in China live in. Based on the two background site campaigns, we found clearly different regional aerosol characteristics between South China and North China, e.g., aerosol compositions and major sources. Specially, our results suggested that possible sources influencing the two background sites may not only include emissions from the Chinese mainland but also include emissions from neighboring countries, which will no doubt improve the current scientific knowledge of regional-scale air pollution in East Asia. The above scientific judgment is more clearly stated in the revised text, such as in the introduction part and the conclusion part.

3. As the two measurements were conducted at different years, they have very different meteorological parameters, the discussion should consider and discuss more the meteorological effects, while current version is clearly lacking of such discussion. No meteorological data are shown. If so, the findings might be only for these two cases, having very limited scientific values for future and other studies. REPLY: The reviewer might ignore the meteorological parameters shown in Table 1. Actually, whether the two campaigns were conducted in the same year is not that important, because

the two sites have a distance of about 1900 km and thus cannot be influenced by the same weather system even in the same year. The difference of general air pollution characteristics between the two sites should be more determined by extensive regional emissions rather than by short-term local meteorology. In section 2.1, we give the information that no matter in 2013, 2014 or 2015, northern China all had a much higher PM2.5 concentration level than that in southern China, confirming that regional emissions are more important. In this study, we pay more attention to regional scale meteorology instead of local meteorology due to little local emission at the background sites. Therefore, back trajectories are more useful than local wind parameters to discuss meteorological effects in this study. We applied the TPSCF model, a statistical method based on back trajectories, to identify potential major source areas, which are not case-dependent and certainly useful for other aerosol studies in East Asia.

4. Regarding the quantification of organic nitrates, do the authors consider influences of metal nitrates? The AMS can measure nitrate that associated with sodium etc., although it is difficult to measure metals. Previous studies also pointed out that metal nitrates can have higher NO/NO2 ratios, so your estimation method is incorrect without considering this point. REPLY: At SCB, the concentrations of measured metals are very close to 0 because the very small "open-closed" difference mass spectra for V and W modes of Na, K, Al, Cu, Zn, and Pb. So the interferences of metal nitrates can be excluded. We make a clarification of this point in the revised manuscript as below: “On the other hand, significant existence of metal nitrates at SCB could be excluded due to non-detectable amounts of metals in the mass spectra.”in Section 2.4.

5. The Vk diagram is quite limited in describing the formation processes of ambient OA in the reviewer’s viewpoint. As there are so many possibilities that can influences the O/C and H/C ratios of ambient OA. The variation of O/C and H/C may not reflect the evolution processes at all. It maybe useful for chamber studies but should be discussed with cautions for ambient data. REPLY: We agree the reviewer’s point and have made a clarification in the revised text to highlight that many other factors can

influence the slope in V-K diagram for ambient data, the Van Krevelen diagram is still useful for constraining the reactions that are responsible for the aging of OA (Hayes et al., 2013). And we modify some descriptions in Section 3.4 as below: “Heald et al. (2010) proposed using the Van Krevelen diagram to illustrate how reactions involving addition of functional groups fall along straight lines for ambient aerosol. Actually, many other factors may also lead to a variety of slopes in the Van Krevelen diagram in the case of ambient field measurements, while the Van Krevelen diagram is still useful for constraining the reactions that are responsible for the aging of OA (Hayes et al., 2013).”

6. Why a 3-factor solution was chosen for NCB site? It seems like 2-factor solution is also fine. In the supplement, it seems like the 2-factor solution is similar as the SCB site. Then why for NCB you chose 3-factor solution? Also, the Mass spectra of OOA1 and OOA2 in your 3-factor solution are quite similar, their diurnal patterns are similar too. Even if the authors insist to keep a POA factor for NCB site, this reviewer thinks OOA1 and OOA2 can be combined as one OOA factor. REPLY: There are two reasons why choosing the 3-factor solution for NCB as a better choice. Firstly, if we chose the 2-factor solution, the one with lower O:C ratio of 0.39 should be regarded as SV-OOA because it has a higher f44 of 6.5%, and thus we will miss HOA. However, high BC concentrations were observed at NCB, implying an HOA component should exist at NCB. Actually, in the quick review reports of this manuscript, it was suggested to split HOA from OOA at NCB by other reviewer. Secondly, although the mass spectra of OOA1 and OOA2 are similar, their time series are totally different, and their source areas are also different according to the TPSCF model analysis, indicating they are factors with different origins despite of similar O:C values.

7. A few technical comments (there maybe more, please check the MS carefully): page 2, line 15: "Valuable insights into the composition, sources, and evolution processes of was mostly found to be a submicron in China were obtained by the powerful on-line tools". What meaning? Please re-write this sentence. Page 5, line 7: a flow rate of 80 l min-1? should be 80 ml min-1. REPLY: All corrected.

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