Responses to Referee #1

We are grateful to the reviewer for the helpful comments and suggestions. Below are point-by-point responses with reviewer's comments in blue and authors' responses in black.

This manuscript addressed the polar organic compounds measured by aircraft during summer and spring in central China. Although the dataset is a little old for the sampling period is in ten years ago, some new insights can be found by the discussion and conclusions. I recommend publishing this paper after addressed the following comments.

1) The detail QA/AC for filter keeping and chemical experiments should be added to show the dataset quality, for it was nearly 10 years past after the sampling.

Response: Actually we finished the analysis for the aerosol samples in February 2008, just a few years after the sampling. As mentioned in the text, after the sampling, each filter was placed in a pre-combusted (450 °C for 6 h) glass vial with a Teflon-lined cap and stored at −20 °C in the dark prior to analyses (see Page 4, Line 98-100). Under such a same filter keeping condition, we once checked one aerosol filter sample (QFF298) collected at Alert in the Arctic in 1991. We analyzed dicarboxylic acids in 1994 and reanalyzed in 2010, and found that the concentrations of major organic acids are equivalent within an analytical error of 15% (Kawamura et al., 2010). Thus, the data reported in the present study should be solid enough.

The following sentence has been added in the revised manuscript. "The GC-MS measurements of polar organic compounds were completed in 2008." (see P5L131-132).

2) The seasonal variation of meteorological condition (for example the mixing layer height and RH) and ground vegetation cover should be described when analysis the datasets.

Response: It's a pity that there were no information on the mixing layer height and relative humidity available during the aircraft campaign.

As suggested by the reviewer, we provided a figure of detailed vegetation cover in central

China, which was derived from a Multi-source Integrated Chinese Land Cover (MICLCover) map (Ran et al., 2012). The below figure was added into the revised manuscript as Figure 7.

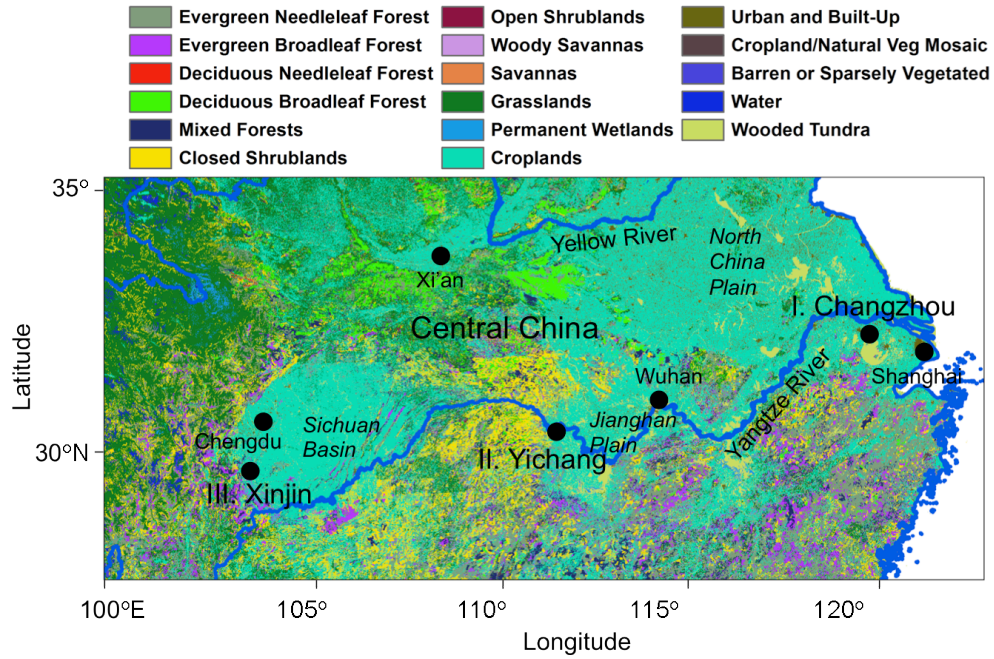

Fig. 7. Spatial distribution of vegetation cover in Central China. The original data were derived from a Multi-source Integrated Chinese Land Cover (MICLCover) map (Ran et al., 2012).

Accordingly, the following paragraph was added in the revised manuscript:

"Terrestrial vegetation emits a large mount of BVOCs into the atmosphere, including isoprene, monoterpenes, sesquiterpenes, as well as oxygenated hydrocarbons (e.g., acetone and methanol) (Guenther et al., 1995). Here, we present the annual vegetation cover in Central China (Figure 7), which was derived from a Multi-source Integrated Chinese Land Cover (MICLCover) map (Ran et al., 2012). The main vegetation types in Central East and South China are evergreen broadleaf forest, evergreen needleleaf forest, and shrubs. However, most parts of the North China Plain, Jianghan Plain and Sichuan Basin are croplands. Broad-leaf trees (e.g., *Quercus*, *Populus*, and *Eucalyptus*) are strong emitters of isoprene; needle-leaf trees (e.g., *Picea* and *Abies*) can emit both isoprene and monoterpenes (Pacifico et al., 2009); crops and herbs are generally weak emitters of isoprene and monoterpenes, instead, they are one of the major contributors to the sesquiterpene emissions. It should be noted that sesquiterpene emissions, as same as isoprene and monoterpene emissions, vary geographically depending on vegetation type and other

factors such as temperature, light, soil moisture, plant developmental stage, as well as atmospheric pollutants (Duhl et al., 2008).

In the past decade, considerable efforts have been devoted to understand biogenic SOA formation because it is an important component in the Earth's atmosphere (Hallquist et al., 2009 and references therein). East China is considered as one of the most important source regions of isoprene emission in the world during summertime (Guenther et al., 1995). Hence, biogenic SOA formation from isoprene oxidation may play an important role in atmospheric chemistry and climate in this region." (see P8L218-237).

3) Do the authors supply fire spot map in spring and summer to support the conclusion that biomass burning was more abundance in spring than in summer? In addition, literatures related biomass burning on ground observation in central China should be cited to support your conclusion.

Response: We provided the fire spot map (see Fig. 4) and added the following discussion in the revised manuscript.

"For example, the concentration of levoglucosan was up to 305 ng m^{-3} in the sample collected over Changzhou city (flight H13 in Figure 1b), which should be affected by the active wheat-straw burning activities in the North China Plain during late spring to early summer (Fu et al., 2008). While in late summer, concentrations of levoglucosan and its isomers were lower than those in spring (Figure 2). Then, we checked the fire spots provided by a Moderate-resolution Imaging Spectroradiometer (MODIS) (https://earthdata.nasa.gov/data/near-real-time-data/firms) (Figure 4). Obviously, in late summer, fire spots could be occasionally observed in Central East China (Figure 4a). While during the spring 2003 campaign, active fire spots were located in the south part of the North China Plain as well as in regions near Changzhou and Shanghai (Figure 4b), suggesting a heavy impact of biomass burning to the regional air quality in Central East China." (see P6, L157-168).

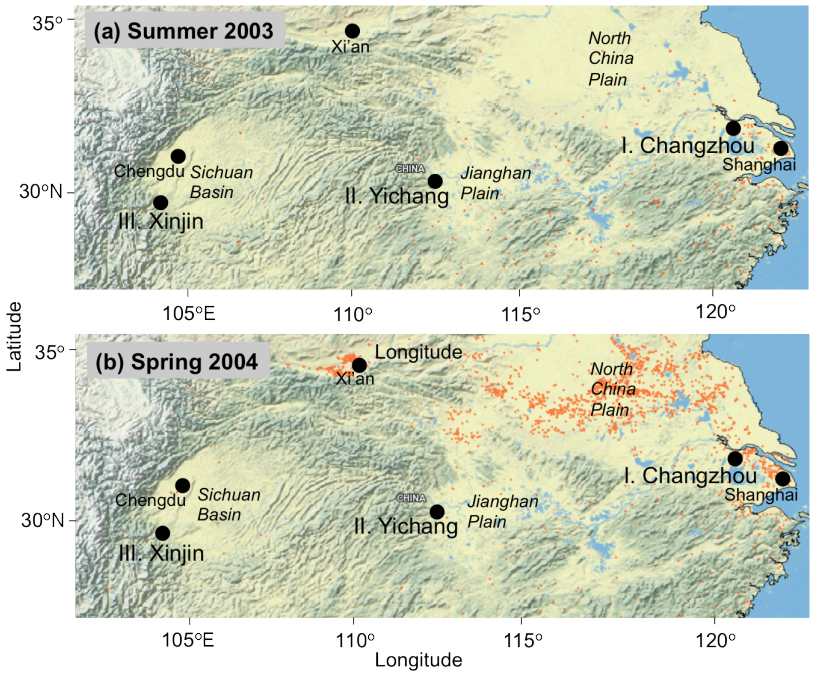

Fig. 4. Spatial distribution of fire spots during **(a)** late summer (5 August–13 September, 2003) and **(b)** late spring (16 May–10 June, 2004) over Central East to West China. The fire data were derived from a Moderate-resolution Imaging Spectroradiometer (MODIS) (https://earthdata.nasa.gov/data/near-real-time-data/firms). (see Page 29).

4) One of conclusions: Concentrations of the measured organic species decreased with an altitude, suggesting that they are emitted from primary sources and/or produced by secondary oxidation of their precursors on the ground surface. I suggest to cite some literatures about ground observation results to demonstrate your assume.

Response: In the Sections of 3.2, 3.4, 3.7 and 3.8, we have cited many previous works on both POA and SOA tracers in different locations on the ground surface including China to improve such an assumption in the text. Thus, we think it is not necessary to cite literatures in the Section of Conclusions.

5) Does the levels of general chemical species (such as dust trace elements, water soluble ions, and OC and EC) were determined or not? Such results may also favor the conclusions of this study.

Response: We did not analyze trace elements et al. for the sample sets. In addition, as shown in the title of the manuscript, our present study mainly focuses on polar organic tracer compounds in the PM_{10} samples. Thus, we consider that the trace elements and water-soluble inorganic ions could be another subject of research.

6) There are too much assuming in conclusions. For example, "Most of the POA and SOA tracers were less abundant at higher altitudes, suggesting they are of ground surface origin, either being directly emitted from anthropogenic/natural sources on the ground surface, or rapidly formed through photooxidation of their precursors emitted from the ground surface and then diluted during uplifting into the troposphere". Even they are beyond the scope of this study, prior literatures can be cited to demonstrate you opinions.

Response: We added several references (Simoneit et al., 1991; Gao et al., 2003; Feng et al., 2006; Fu et al., 2010; Wang et al., 2013) in the revised manuscript. (see P12L361-362).

7) For the last sentence of the abstract, the author should be cautioned that the levels of primary biological aerosols and biogenic SOA influenced heavily on the ground vegetation cover, so their seasonal distribution should be evident, that's their levels should be high in late spring, summer, and early autumn, but relative lower in cold seasons.

Response: Suggestion is taken. We rephrased the last sentence of the Abstract "This study demonstrates that primary biological aerosols, biogenic SOA, and organic nitrogen species are important components of organic aerosols in the troposphere over Central China" to "This study demonstrates that primary biological aerosols, biogenic SOA, and organic nitrogen species are important components of organic aerosols in the troposphere over Central China during warm seasons." (see P2L40).

References

Duhl, T.R., Helmig, D., Guenther, A., 2008. Sesquiterpene emissions from vegetation: a review. *Biogeosciences* 5, 761-777.

Feng, J., Hu, M., Chan, C.K., Lau, P.S., Fang, M., He, L.Y., Tang, X.Y., 2006. A

comparative study of the organic matter in PM2.5 from three Chinese megacities in three different climatic zones. *Atmos. Environ.* 40, 3983-3994.

Fu, P.Q., Kawamura, K., Okuzawa, K., Aggarwal, S.G., Wang, G.H., Kanaya, Y., Wang, Z.F., 2008. Organic molecular compositions and temporal variations of summertime mountain aerosols over Mt. Tai, North China Plain. *J. Geophys. Res., [Atmos]* 113, D19107, doi:10.1029/2008JD009900.

Fu, P.Q., Kawamura, K., Kanaya, Y., Wang, Z.F., 2010. Contributions of biogenic volatile organic compounds to the formation of secondary organic aerosols over Mt. Tai, Central East China. *Atmos. Environ.* 44, 4817-4826.

Gao, S., Hegg, D.A., Hobbs, P.V., Kirchstetter, T.W., Magi, B.I., Sadilek, M., 2003. Water-soluble organic compounds in aerosols associated with Savanna fires in southern Africa: Identification, evolution, and distribution. *J. Geophys. Res.* 108, 8491, doi:8410.1029/2002JD002324.

Guenther, A., Hewitt, C.N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., McKay, W.A., Pierce, T., Scholes, B., R., S., Tallamraju, R., Taylor, J., Zimmerman, P., 1995. A global model of natural volatile organic compound emissions. *J. Geophys. Res.* 100, 8873-8892.

Hallquist, M., Wenger, J.C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N.M., George, C., Goldstein, A.H., Hamilton, J.F., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M., Jimenez, J.L., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, T.F., Monod, A., Prévôt, A.S.H., Seinfeld, J.H., Surratt, J.D., Szmigielski, R., Wildt, J., 2009. The formation, properties and impact of secondary organic aerosol: current and emerging issues. *Atmos. Chem. Phys.* 9, 5155-5235.

Kawamura, K., Kasukabe, H., Barrie, L.A., 2010. Secondary formation of water-soluble organic acids and α-dicarbonyls and their contributions to total carbon and water-soluble organic carbon: Photochemical aging of organic aerosols in the Arctic spring. *J. Geophys. Res., [Atmos]* 115, D21306, doi:10.1029/2010JD014299.

Pacifico, F., Harrison, S.P., Jones, C.D., Sitch, S., 2009. Isoprene emissions and climate. *Atmos. Environ.* 43, 6121-6135.

Ran, Y.H., Li, X., Lu, L., Li, Z.Y., 2012. Large-scale land cover mapping with the integration of multi-source information based on the Dempster-Shafer theory. *Int. J. Geograph. Inform. Sci.* 26, 169-191.

Simoneit, B.R.T., Sheng, G.Y., Chen, X.J., Fu, J.M., Zhang, J., Xu, Y.P., 1991. Molecular marker study of extractable organic-matter in aerosols from urban areas of China. *Atmos. Environ., Part A* 25, 2111-2129.

Wang, G.H., Zhou, B.H., Cheng, C.L., Cao, J.J., Meng, J.J., Li, J.J., Tao, J., Zhang, R.J., Fu, P.Q., 2013. Impact of Gobi desert dust on aerosol chemistry of Xi'an, inland China during spring 2009: differences in composition and size distribution between the urban ground surface and the mountain atmosphere. *Atmos. Chem. Phys.* 13, 819-835.