Responses to Referee #1

General comments:

This manuscript presents a thorough investigation on the OC, EC, WSOC, and organic compound species in tropospheric aerosols collected at a high mountain site (Mt. Tai) in Central East China during a biomass-burning season. The authors have found that most of the organic species showed higher concentrations in early June than in late June due to the field burning of wheat straws. The data interpretation was further assisted by a comparison with published data for similar organic compounds from different locations. In my opinion, this is one of the first papers focusing on both primary and secondary organic aerosol tracers, as well as δ^{13} C of TC in high mountain aerosols, especially in China. The data presented here is interesting and convincing, and it certainly meets the scope of the journal. Therefore, this manuscript is suitable for publication in ACP after a minor revision.

<u>Response</u>: 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.

Detailed comments and technical errors:

1) Page 9081, the abstract section, line 17-23, how about the boundary layer height? Was it high enough for the transport of the plumes onto the mountaintop? If the night peak was caused by more burning activities at night?

<u>Response</u>: During the MTX2006 campaign, the height of boundary layer was 200-800 m at nighttime, while it was about 1800-4200 m at daytime (Takigawa et al., unpublished data). The summit of Mt. Tai (1534 m, a.s.l.) was present in the free troposphere at nighttime when the heights of PBL were decreased to 200–800 m. Thus, during the severe biomass-burning period (early June), the higher concentrations of organic compounds observed at nighttime should be caused by the long-range transported smoke plumes from local/regional places to the summit of Mt. Tai in the free troposphere during nighttime.

2) Page 9084, Section 2.1. Quartz fiber filters (QFFs) were used for sampling in the present study. Thus the sampling artifacts (see Turpin et al., 2000) and their potential impact on the results should be addressed briefly in this section.

<u>Response</u>: The following sentences will be added in the revised manuscript in Section 2.1. "It should be noted that the sampling artifacts might occur due to the evaporation/adsorption processes of semi-volatile organics on the filter or particle surface during sampling (Turpin et al., 2000)." (see Page 4, Line 114-116)

3) Page 9085, line 28-page 9086, line 1. ". . .whose field blank levels sometimes were high compared to real samples. . .". Please clarify how "high" the contamination level is. And how about the blank levels of other organic species?

<u>Response</u>: The sentence of "....their results showed no significant contamination except for bis(2-ethylhexyl) phthalate, whose field blank levels sometimes were high compared to real samples...." has been modified to "....their results showed no significant contamination (less than 5% of real samples) except for bis(2-ethylhexyl) phthalate, whose field blank levels sometimes were high (up to 60% of real samples)...." (see Page 5 Line 152-155).

4) Page 9086, line 8. ". . ., an aliquot (\$\$\phi14\$ mm) of . . .". What does it mean? It seems to reference a size of filter punch.

<u>Response</u>: "..., an aliquot (ϕ 14 mm) of..." has been changed into "..., a 1.54 cm² punch (ϕ 14 mm) of..." in the revised manuscript (see Page 6, Line 160).

5) Page 9089, line 14-17, in comparison to the fossil fuel combustion, biomass burning can produce more water-soluble organic compounds, (see Wang et al., AE, 45: 2473-2479 and EST 43: 6493-6499), thus WSOC and levoglucosan highly correlated each other. Page 9089, line 8. ". . . and acting as CCN." This sentence requires a reference.

<u>Response</u>: A reference of (Wang et al., 2011) has been added in the revised manuscript (see Page 9, Line 248).

A reference has been added after the sentence of "...and acting as CCN (Mochida and Kawamura, 2004)" (see Page 8, Line 239).

6) Page 9091, line 19-20, why? Many papers reported that biomass burning can produce lots of PAHs, so I think it's better to give more explanation here.

<u>Response</u>: The authors reported that biomass burning (especially wheat straw burning activities) contributed little on hopanes (not PAHs) in the lines that the reviewer mentioned (see Page 9091, Line 19-20 in the ACPD paper).

7) Page 9092, line 16-19. The positive correlation between levoglucosan and the sum of arabitol and mannitol does not necessarily indicate that biomass burning is an important source of these sugar alcohols. Some specific reference is required for this statement.

Response: The sentences of "Their temporal patterns are also similar to that of levoglucosan (Figure 5a), suggesting that biomass burning can also emit a certain level of sugar alcohols. This idea is supported by the correlation ($R^2 = 0.54$, n = 37, figure not shown) between levoglucosan and the sum of arabitol and mannitol. Another possibility is that airborne fungal spores can be adsorbed onto smoke particles and then co-transported to the summit of Mt. Tai." have been modified as "Their temporal patterns are also similar to that of levoglucosan (Figure 5a), suggesting that biomass-burning activities can enhance the emission of sugar alcohols at a certain level. This idea is supported by the correlation ($R^2 = 0.54$, n = 37, not shown as a figure) between levoglucosan and the sum of arabitol and mannitol. Recently, Yang et al. (2012) observed elevated fungal tracers in atmospheric aerosols due to biomass burning in Chengdu City, China. One potential process is that airborne fungal spores can be adsorbed onto smoke particles and then summit of Mt. Tai." (see Page 11, Line 341-343)

8) Page 9093, the section 3.3.4, Wang et al have reported the impact of the summertime wheat straw burning on the chemical composition and size distribution of organic aerosols in a Chinese mega-city, I recommend to make comparison with their results, which might be very interesting for readers to see the difference between the lowland urban area and the elevated mountaintop.

<u>Response</u>: Thanks. The following sentence has been added in the revised manuscript: "Similar molecular distributions of *n*-alkanes and fatty acids were also reported in urban aerosols collected during a severe haze event in Nanjing, East China (Wang et al., 2009). Such an urban haze event was also caused by wheat straw burning in the North China Plain in summer 2007." (see Page 13, Line 379-382)

9) Page 9094, line 19-21, what is the reason that an extremely high value of benzoic acid (57 ng m-3) was observed in early June?

Response: Sorry. We made a mistake here. The sentence of "Higher levels of benzoic acid were observed in late June (2.4-12 ng m⁻³) than those in early June (0.88-57 ng m⁻³)." has been changed as "Higher levels of benzoic acid were observed in late June (0.88-57 ng m⁻³, mean 9.8 ng m⁻³) than in early June (2.4-12 ng m⁻³, 5.8 ng m⁻³)." in the revised manuscript. (see Page 13, Line 399-401)

10) Page 9098, line 1-9, how about fatty acid C16:0, a compound classified to the category (b), which is abundant in cooking emission.

<u>Response</u>: As mentioned in the manuscript, there are many sources of low molecular weight fatty acids such as C16:0. Cooking emission should be one of the major sources of C16:0 fatty acid in urban regions. In the present study, C16:0 was grouped into category (b) "marine / microbial source", because the influence of cooking emission should be minor at the summit of Mt. Tai.

11) Page 9100, line 16-18, I am not sure this conclusion is strong enough, because 13C value of ambient aerosols including the background is always variable.

<u>Response</u>: Stable carbon isotopic compositions of ambient aerosols should vary from different regions. Here the authors just estimated the stable C isotopic composition of background aerosols over Mt. Tai in summer using the equation obtained in this study, which should be acceptable.

12) Page 9122, Figure 11. "Isiprene SOC" should be "Isoprene SOC".

Response: Corrected. (see Page 39, Figure 11).

References

- Mochida, M. and Kawamura, K.: Hygroscopic properties of levoglucosan and related organic compounds characteristic to biomass burning aerosol particles, J. Geophys. Res., [Atmos], 109, D21202, doi:10.1029/2004JD004962, 2004.
- Turpin, B. J., Saxena, P. and Andrews, E.: Measuring and simulating particulate organics in the atmosphere: problems and prospects, Atmospheric Environment, 34, 2983-3013, 2000.
- Wang, G. H., Kawamura, K., Xie, M. J., Hu, S. Y., Cao, J. J., An, Z. S., Waston, J. G. and Chow, J. C.: Organic molecular compositions and size distributions of Chinese summer and autumn aerosols from Nanjing: Characteristic haze event caused by wheat straw burning, Environ. Sci. Technol., 43, 6493-6499, 2009.
- Wang, G. H., Chen, C. L., Li, J. J., Zhou, B. H., Xie, M. J., Hu, S. Y., Kawamura, K. and Chen, Y.: Molecular composition and size distribution of sugars, sugar-alcohols and carboxylic acids in airborne particles during a severe urban haze event caused by wheat straw burning, Atmos. Environ., 45, 2473-2479, 2011.
- Yang, Y., Chan, C.-Y., Tao, J., Lin, M., Engling, G., Zhang, Z., Zhang, T. and Su, L.: Observation of elevated fungal tracers due to biomass burning in the Sichuan Basin at Chengdu City, China, Sci. Tot. Environ., 431, 68-77, 2012.