Responses to Referee #3

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.

General comments:

This study presents an airborne investigation of tropospheric aerosols over central China. The PM_{10} aerosol samples collected during summer 2003 and spring 2004 are analyzed for polar organic compounds using a technique of solvent extraction/BSTFA derivatization/gas chromatography-mass spectrometry. The spatial and vertical trends of polar organic tracer compounds are investigated based on their potential sources. The authors demonstrate that primary biological aerosols, biogenic secondary organic aerosols, and organic nitrogen species are important components of organic aerosols in the troposphere over Central China. Overall, this work is interesting and is certainly relevant within the scope of ACP. I recommend this paper to be published in ACP after minor revisions noted below.

Minor suggestions:

1. Section 2.2, Page 24486, Line 20-25: It would be useful for readers if the authors can provide more explanation on the identification of the organic species here.

Response: The following information has been added in the revised manuscript.

"Individual compounds (TMS derivatives) were identified by comparison of mass spectra with those of authentic standards or literature data (Claeys et al., 2004; Simoneit et al., 2004a; Jaoui et al., 2007; Hu et al., 2008; Lin et al., 2012). Following fragment ions were used for quantifications: *m/z* 217 and 204 for sugar compounds, *m/z* 297 and 312 for vanillic acid, *m/z* 239 for dehydroabietic acid, *m/z* 129 for sterols, *m/z* 262 for 3-MeTHF-3, 4-diols, *m/z* 219 and 203 for 2-methylglyceric acid, *m/z* 219 and 277 for 2-methyltetrols, *m/z* 231 for C_5-alkene triols, *m/z* 171 for pinonic and pinic acids, *m/z* 349 for 3-hydroxyglutaric acid, *m/z* 405 for 3-methyl-1, 2, 3-butanetricarboxylic acid (MBTCA), *m/z* 383 for β-caryophyllinic acid, *m/z* 193 for aromatic acids,

m/z 147 for hydroxy acids, *m/z* 377 for tricarballylic acid, *m/z* 189 for urea, and *m/z* 190 for oxamic acid." (see P4-5, L116-125).

2. Section 3.4, Page 24490: Many field observations have shown that 2-methyltetrols (IEPOX-derived species) dominate over 2-methylglyceic acid (MPAN-derived) in summer (Kleindienst et al., 2007; Lewandowski et al., 2007). How about the concentration ratios of 2-methyltetrols to 2-methylglyceric acid in summer and spring campaigns?

Response: As suggested by the reviewer, we calculated the concentration ratios of 2-methyltetrols to 2-methylglyceric acid in both summer and spring campaigns, and the results are presented in the revised manuscript as follows (see P9-10, L272-277): "Thus, the concentrations ratios of 2-methyltetrols to 2-MGA were found to be several times higher during the summer campaign (2.8–23, mean 10) than those (0.9–6.4, 2.8) during in spring (Table 2). The higher ratios observed in summer than in spring could be explained by the aerosol acidity, NO_x conditions, pathways leading to the production of 2-methyltetrols and 2-MGA, and the relative amounts of their gas-phase precursors such as ISOPOOH and MAE (Pye et al., 2013)."

In addition, the sentence of "2-MGA can be formed by the oxidation of methacrolein and methacrylic acid, two major gas-phase oxidation products of isoprene, and is considered as a methacrylic acid epoxide (MAE)-derived product under high-NO_x conditions (Lin et al., 2013)." (see P9, L267-270).

3. Section 3.4: Do the authors have data on NO_x during the aircraft campaigns? It should be interesting to see the effect of NO_x on the formation of isoprene SOA tracers in the troposphere.

Response: We fully agree that the effect of NO_x on the formation of isoprene SOA tracers in the troposphere is an interesting topic. It's a pity that we didn't have NO_x data during the whole campaigns. So, in the present study, we just briefly mentioned the effects of NO_x and other parameters on the formation of isoprene SOA as follows:

"Thus, the concentrations ratios of 2-methyltetrols to 2-MGA were found to be several times higher during the summer campaign (2.8–23, mean 10) than those (0.9–6.4, 2.8) during in spring (Table 2). The higher ratios observed in summer than in spring could be explained by the aerosol acidity, NO_x conditions, pathways leading to the production of 2-methyltetrols and 2-MGA, and the relative amounts of their gas-phase precursors such as ISOPOOH and MAE (Pye et al., 2013)." (see P9-10, L272-277).

4. Page 24491, Line 26-27: "...inorganic tracer of biomass burning." References are needed here.

Response: References added. "nss-K^+ is an inorganic tracer of biomass burning (Falkovich et al., 2005)." (see P10, L299-300).

Another sentence was also added in the revision: "In fact, in addition to the emissions from biomass burning, nss-K^+ is also representative of natural sources such as dust soil resuspension and biogenic emissions (e.g., plant exudates) (Sella et al., 2006)." (see P10, L304-306).

5. Page 24493, Line 2: The volatility of urea is generally low. So the primary or direct emission of urea into the atmosphere due to agricultural activities may be insignificant. Please clarify this point.

Response: In the revision, the sentences of "Urea could be primary emitted from agricultural activities in Central China. However, urea may also be a secondary product from atmospheric reactions (Simoneit et al., 2004a)." have been changed into "However, the vapor pressure of urea is very low (Bernhard et al., 2011); it is much less volatile than water. During the ACE-Asia campaign, Simoneit et al. (2004b) reported that urea was not observed in the ground surface aerosol samples. In a previous study, 65% of urea was present in the coarse mode in the marine atmosphere, while large fractions of urea were found in the fine mode (<2.1 μm) in coastal or continental aerosols (Shi et al., 2010). Thus, except for the emissions of urea from primary

sources such as agricultural activities, soil dust and the sea surface through bubble-bursting processes (Shi et al., 2010; Violaki and Mihalopoulos, 2011), the secondary production of urea from atmospheric reactions could also be among the major sources. Such a point warrants further studies. Interestingly, although it is hygroscopic and highly water soluble, urea seems to be active as cloud condensation nuclei (CCN) in the atmosphere because it has an endothermic heat of solution that makes it take on a water film and then freezes part of the water (Byers, 1965)." (see P11-12, L332-344).

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