General comments:

This manuscript presents an interesting study on the presence of nitrogen containing aromatic compounds and their light absorbing properties in laboratory generated biomass burning organic aerosols. I found the manuscript difficult to follow in its current form, mainly because most of important information that supports the authors' discussion is presented in supporting information (Tables S1, S2, and Figure S2). They can be moved to the main manuscript. Apart from the organization of the manuscript, I have three issues that I want the authors to address prior to the acceptance of this manuscript.

Reply:

Thanks for the reviewer's comments, and we replied to each comment as below.

Specific major comments:

1. Table S1 MAC365 for 7/14/2016 sample and lines between 199 and 202. This highlights the difficulties associated with the comparison of the data obtained from laboratory combustion experiments. As the authors suggest between the lines 199 and 202, the ambient condition appears to be very important for MAC365 values because the summertime combustion of NC forest 2 shows significantly higher values for MAC365 than the springtime combustion of NC forest 1. This can only mean higher absorption coefficients of the NC forest 2 samples than those of the NC forest 1 or lower methanol extractable mass concentrations of the NC forest 2 samples than those of the NC forest 1. Based on the higher ambient temperature of the NC forest 2 experiments, I assumed that this originates from the difference in gas/particle partitioning (i.e. higher gas phase concentrations of MAC365 compounds in NC forest 2 experiment) though it is not too clear to me if this is the case when I see the mass fractions of MAC365 products depicted in Figure 1 and Table S4. From Figure 2 and Table S6, it can also deduce that the samples from NC forest 2 combustion contained highly light absorbing compounds that are not detected in this study. Can the authors elaborate more in the manuscript? As is now, it is not too clear to me why the MAC365 values are so different when other parameters are relatively similar.

Reply:

In the revised manuscript, we added more discussions on the significant difference in bulk MAC_{365} of solvent extractable OC between the two NC forest experiments. We have considered the evaporation of more volatile OC in summer, which might not be the main reason. It is very likely that stronger light-absorbing components are generated from BB in summer than in spring, but the mechanism is unknown and warrants further study.

In pages 11–12, lines 251–263,

"The two NC forest experiments were conducted in spring and summer, respectively, with distinct ambient conditions (Table S1), and their average MAC_{365} values were significantly

($p < 0.05$) different. This could be partly ascribed to the fact that more semi-volatile OC (SVOC) will partition into gas phase in summer with higher ambient temperatures, and the SVOC is less light-absorbing than OC with low volatility (Chen et al., 2010;Saleh et al., 2014). However, if the relative abundance of EC and OC from BB emissions is similar between the two NC forest experiments, the evaporation of SVOCs in summer will lead to higher EC/OC ratios, which is not observed in Table 1. No previous study investigated the seasonal variation in BrC absorption from BB with similar fuel type. Chen et al. (2001) found that the ambient temperature might play a role in EC production from traffic by changing the air density. We suspected that the BB samples from NC forest 2 combustion in summer contained much stronger light-absorbing components than NC forest 1 combustion in spring, although the formation mechanism of these strong BrC components is uncertain and merits further study."

2. Line 215: Is there a reason for the choice of 1.7 OM/OC factor? In the original paper of Turpin and Lim (2001), 1.7 was not mentioned as a conversion factor for biomass burning organic aerosol. Values for fireplace combustion cited in Turpin and Lim (2001) were between 1.9 and 2.1 that were determined by Schauer (1998). In addition, there are several more recent values available in the literature.

Reply:

Thanks for the reviewer's comment. We used the wrong reference in the original manuscript, and we replaced Tupin and Lim (2001) with Reff et al. (2009). In the supplementary information of Reff et al. (2009), they went through a summary of the OM/OC ratio of various sources and they arrive at a value of 1.7 for biomass burning/wood burning.

3. Identification of the benzisoxazole skeleton It is not clear to me why the authors attributed the loss of CNO as the presence of the benzisoxazole skeleton instead of e.g. isocyanates for C9H9NO4 compounds. By losing CNO (actually HCNO) from the benzisoxazole skeleton, one forms highly unstable biradical product ions that aren't likely detected in MS. Can the authors shed light on how the fragments are formed in the revised manuscript?

Reply:

Yes, we should not attribute the loss of OCN to a benzoxazole structure without enough evidence. In the revised work, four standard compounds, including phenyl cyanate (C_6H_5OCN), benzoxazole (C_7H_5NO), 4-methoxyphenyl isocyanate ($CH_3OC_6H_4NCO$) and 2,4-dimethoxyphenyl isocyanate [$(CH_3O)_2C_6H_3NCO$], were analyzed using a gas chromatography (Agilent 6890) coupled to a mass spectrometer (Agilent 5975B) under electron ionization (EI, 70 ev) mode. These compounds do not have a phenol structure and cannot be detected using ESI under negative ion mode. The MS/MS spectra of 4-methoxyphenyl isocyanate and 2,4-dimethoxyphenyl isocyanate were obtained by using a modified method (ESI at positive ion mode) for NACs analysis in this work. As shown in Fig. S4a and b in the revised supplementary information (See below), the loss of OCN is observed for phenyl cyanate, but not benzoxazole. In Fig. S4c and d, the ions at *m/z* 106 and 136 can be produced from the species at *m/z* 149 and 179 through the loss of CH_3 + CO or H + NCO (43 Da). The MS/MS spectra of 4-methoxyphenyl isocyanate and 2,4-dimethoxyphenyl isocyanate (Fig. S4e,f) confirmed the loss of CH_3 + CO, and the loss of CH_3 reflected the presence of methoxy group. As such, the $C_9H_9NO_4$ compounds identified in this work is expected to contain a phenyl cyanate structure.

These results and discussions were added in the revised manuscript (Pages 14–15, lines 311–331).

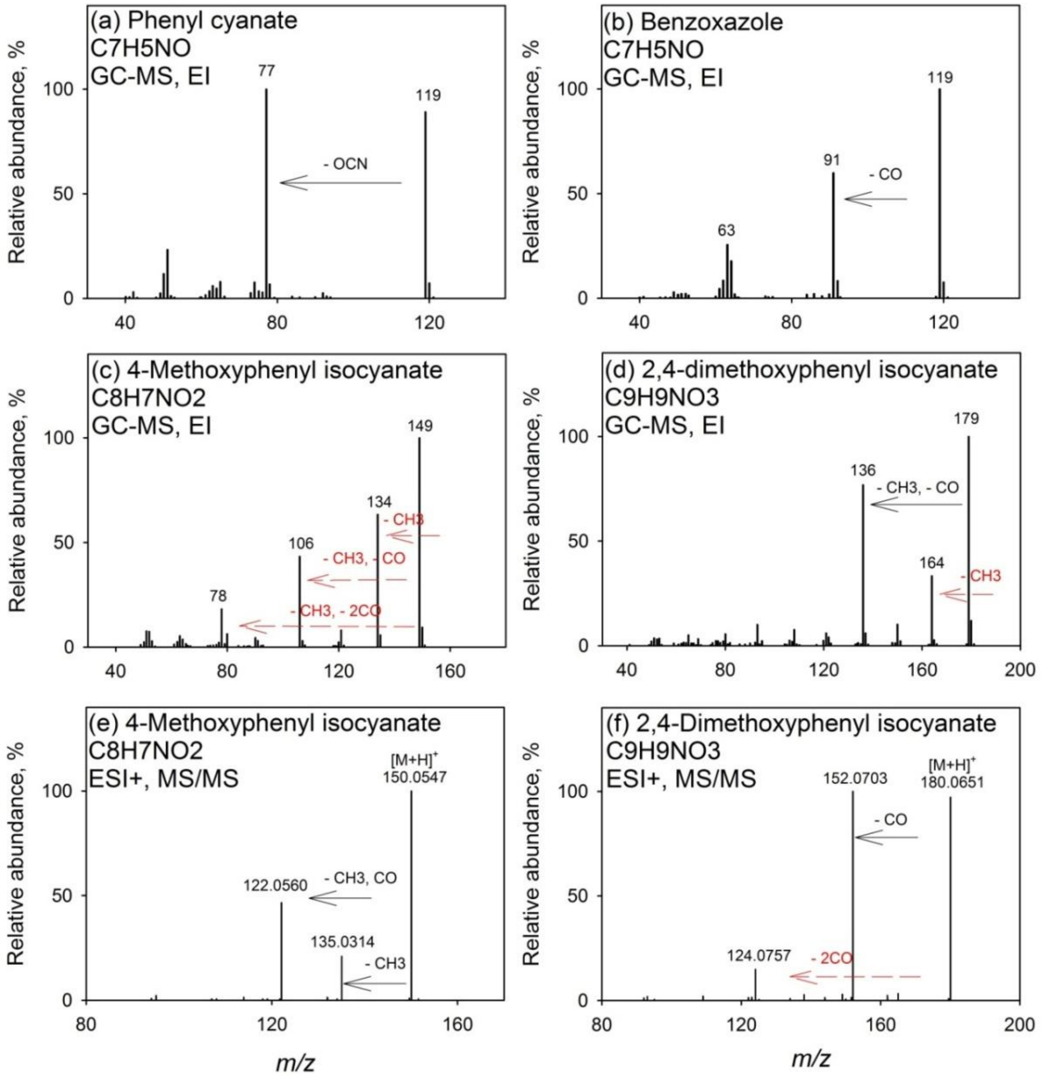

Figure S4. Mass spectra of (a) phenyl cyanate, (b) benzoxazole, (c) 4-methoxyphenyl isocyanate, and (d) 2,4-dimethoxyphenyl isocyanate with EI mode; MS/MS spectra of (e) 4-methoxyphenyl isocyanate and (f) 2,4-dimethoxyphenyl isocyanate with ESI positive ion mode.

References

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