

Responses to Referee #1

General comments:

This manuscript presents a field campaign to measure secondary organic aerosol (SOA) tracers at the sampling site in Mt. Tai, China. The tracer compounds reported in this study have been well documented in earlier studies from field studies and aerosol chamber experiments. The authors suggest the use of (sum of isoprene oxidation products)/(sum of monoterpene oxidation products) ($R_{iso/mono}$) as an indicator to estimate the contribution of isoprene and monoterpenes to biogenic SOA formation in various ecosystems. The idea is certainly attractive and reasonable. However, there is a fundamental problem with this approach as the authors used in this manuscript. The spectrum of compounds that the authors have detected in this study is far from complete, especially for monoterpene oxidation products. Indeed, monoterpene oxidation products that the authors have detected are known to be rather minor monoterpene oxidation tracer compounds in ambient aerosols except for pinonic acid and 3-methyl-1,2,3-butanetricarboxylic acid (MBTCA). This raises a serious question to whether this ratio has any meanings to it. In order to obtain any meaningful $R_{iso/mono}$ values, the authors need to include organosulfates and nitrooxy-(or nitrated)organosulfates originating from both isoprene and monoterpenes as they are likely to contribute significantly to biogenic SOA in ambient aerosols, especially anthropogenically influenced areas (see e.g. Lukács et al. 2009). The authors rely heavily on the use of surrogate compounds for quantification. This may be acceptable if the focus of the study is to provide a rough estimate of the concentrations of tracers that are newly identified or if the surrogate compound is a homologue of the target compound. However, the quantification is the key for this study; the whole discussion is based on the quantitative results from the chemical analysis yet large part of quantification work is performed using a handful of surrogate compounds. This leaves the impression to the readers that tentative quantification of few compounds using a surrogate compound is sufficient for publication even the focus of the study is the quantitative assessment of the contribution of a large group of compounds to local PM. In addition, the interpretation of the data is very weak. Only the most obvious observations are stated and no connection is made to the atmospheric chemistry of the region. As a result, this reviewer is not able to recommend this manuscript for publication in ACP. The detailed comments are as follows.

Response: The authors are very grateful to the reviewer for a thorough review, which provides excellent suggestions for improvements of this manuscript. We understand that the main concern is about the usage of the surrogate compounds for quantification in this study. Owing to a lack of authentic standards commercially available, almost all these SOA tracers were quantified using the same surrogates (such as *meso*-erythritol for 2-methyltetrols and C₅-alkene triols) (Claeys et al., 2004; Clements and Seinfeld, 2007; Ding et al., 2008; Hu et al., 2008; Wang et al., 2008; Kourtchev et al., 2009). In some recent studies, ketopinonic acid was used as a surrogate to quantify all these SOA tracers (Offenberg et al., 2009; Stone et al., 2009).

In order to minimize the uncertainty of the quantification of the organic tracers, we spent several weeks performing a series of experiments on the BVOCs (isoprene, α -pinene, β -caryophyllene) + OH reactions in a quartz tube under UV irradiation in the laboratory. The reaction products were analyzed using GC-MS. 2-Methylglyceric acid was obtained as the most abundant for isoprene + OH reaction product. Other SOA tracers were also detected but not abundant. This suggests that the natural atmosphere is a highly mixed system that cannot be easily simulated in the laboratory. Then, we chose those ambient aerosol samples with high concentrations of 2-methylglyceric acid, C₅-alkene triols, 2-methyltetrols, and MBTCA. The quantitative determination of these SOA tracers were performed using a capillary GC (Hewlett-Packard, HP6890) equipped with a split/splitless injector, fused-silica capillary column (HP-5), and a flame ionization detector

(FID). It was assumed that the GC-FID responses of the trimethylsilyl derivatives of the compounds and their respective internal standards were equivalent because the ionization efficiencies in GC-FID are roughly proportional with the abundance of carbon atoms in the molecules. This technique has been successfully used in the quantification of isoprene SOA tracers such as 2-methyltetrols (Claeys et al., 2004; Edney et al., 2005). In the revised manuscript, the concentrations of 2-methylglyceric acid, C₅-alkene triols, 2-methyltetrols and MBTCA will be recalculated using the response factors obtained from the GC-FID analysis. In addition, we have purchased a new standard of 3-hydroxyglutaric acid. The concentrations of 3-hydroxyglutaric acid have been quantified using the response factor of authentic standard. All the tables and figures will be modified accordingly.

We proposed the $R_{\text{iso/mono}}$ value to evaluate the relative contribution of isoprene and monoterpenes to SOA simply because these polar organic tracers (e.g., 2-methyltetrols and pinic acid) have been reported in aerosols collected from different geographical locations, which makes it possible to calculate and compare the $R_{\text{iso/mono}}$ values through other studies. The reviewer suggested that organosulfates should be included for the calculation of $R_{\text{iso/mono}}$ values. Although organosulfates and nitrooxy-(or nitrated)organosulfates are reported recently as both isoprene (Surratt et al., 2007a; 2007b; Gómez-González et al., 2008) and monoterpenes (Liggio and Li, 2006; Iinuma et al., 2007) SOA in ambient aerosols, most of these studies used different methods (e.g., ESI-MS). Meanwhile, there is still a lack of quantitative information of organosulfates because of the lack of authentic standards or surrogate compounds for quantification. To the best of our knowledge, up to date, there has been no study reported both organosulfates and biogenic SOA tracers from isoprene to β -caryophyllene simultaneously. Therefore, we think it is reasonable not to include organosulfates for the comparison of $R_{\text{iso/mono}}$ values in the current study.

The introduction itself will be modified in order to better state the goal of the study. The current study is the first attempt to report the SOA oxidation products from biogenic VOCs at high altitudes over East Asia. We believe that our results are very helpful for readers to better understand the chemical composition of organic aerosols and the contributions of biogenic VOCs to SOA at high altitudes.

Please see our detailed responses as follows.

Specific comments:

Pp. 16943, line 16: What do the authors mean by “among the highest in the aerosols studied in different regions”? Do the authors mean this study, other studies that the authors are involved or available literature data? Furthermore, how reliable is this ratio when the quantification in this study relies heavily on the use of surrogate compounds? In addition, the authors have determined only five monoterpene oxidation traces that are far from complete. Better quantification attempt is needed for both isoprene and monoterpene oxidation traces as detailed synthetic methods are described for some of major compounds that the authors have detected. I suggest removing this sentence completely.

Response: We mean that the $R_{\text{iso/mono}}$ values in this study are higher than those calculated using available literature data of the aerosols from different ground surface sites. All those studies used the same surrogates for isoprene SOA tracers and the same spectrum of polar tracers listed in Table 2 in the submitted manuscript, so it is comparable with our results. In addition, we have re-calculated the data using improved methods as mentioned above.

Pp. 16943, lines 17-18: It is not too clear how high O₃, NO_x and OH concentrations relate to high isoprene

oxidation product/monoterpene oxidation product ratios. Especially, how does high NO_x lead to high isoprene oxidation tracer concentration in the particle phase? Here, the ratio of $\text{VOC}/\text{NO}_2/\text{NO}$ rather than the total " NO_x " is important for the distributions of both isoprene and monoterpene oxidation products. How have the authors determined the isoprene flux? What about monoterpenes?

Response: We fully agree with the comments. Information on the temporal variations of NMHCs/ NO_x will be added, and the corresponding figure will be modified in the revised manuscript. The mixing ratios of NMHCs were made with canister samplings (performed on 30 occasions during the campaign) followed by analysis using gas chromatography-flame ionization detection (GC-FID) and gas chromatography/mass spectrometry (GC/MS) (Suthawaree et al., 2009).

Pp. 16943, lines 18-19: What is the tracer based method? Is it simply the sum of all detected compounds? If this is the case, it is misleading to call this number as secondary organic carbon (SOC) as the compounds that the authors have determined do not cover a spectrum of compounds that are considered to be both anthropogenic and biogenic secondary organic compounds. Furthermore, a term "monoterpenes" is not exactly correct when the authors have detected mostly α -pinene oxidation products.

Response: The tracer-based method, first proposed by Kleindienst et al. (2007), has been successfully used to assess the secondary organic carbon (SOC) and SOA mass contributions of isoprene, monoterpenes and β -caryophyllene to ambient OC and organic matter (OM) (Hu et al., 2008; Offenberg et al., 2009; Stone et al., 2009). By using the smog chamber experimental data of different hydrocarbon/ NO_x mixtures, Kleindienst et al. (2007) derived the mass fraction of tracer compounds in SOC (f_{soc}) generated from individual precursor, which was defined as

$$f_{\text{soc}} = \frac{\sum_i [\text{tracer}_i]}{[\text{SOC}]} \quad (1)$$

where $\sum_i [\text{tracer}_i]$ is the sum of the carbon fraction of the selected suite of tracers for a same biogenic VOC precursor in $\mu\text{g m}^{-3}$, and $[\text{SOC}]$ the measured SOC concentration in $\mu\text{g m}^{-3}$ in chamber studies. The laboratory-derived f_{soc} for isoprene, α -pinene and β -caryophyllene were 0.155 ± 0.039 , 0.231 ± 0.111 , and 0.0230 ± 0.0046 , respectively. This approach to SOA attribution is a simplification of a truly complex system of the real atmosphere, which is a good tool available currently (Stone et al., 2009).

A brief introduction of this approach and its uncertainties will be added in Section 3.5 in the revised manuscript as "...It should be noted that the tracer-based method is a simplification of a truly complex system of the real atmosphere (Stone et al., 2009). The uncertainties of this approach range from 20 to 48%, based on the standard deviation of f_{soc} mentioned above (Kleindienst et al., 2007a)."

The term "monoterpenes" will be changed as " α -pinene" accordingly.

Pp. 16943, line 23: This reviewer feels that this statement is unwarranted for above mentioned reasons. The detected compounds do not cover a spectrum of compounds that originates from the oxidation of isoprene and a variety of monoterpenes (including isoprene and monoterpene originating organosulfates and nitrooxy-organosulfates).

Response: Here the percentage of SOC to OC was calculated not from the total concentration of SOA tracer, but from the tracer-based method mentioned in Section 3.5. Although it is not currently known if there is a linear relationship between the tracer species and SOC yields, the tracer-based method was the best tool

available at this moment to evaluate the percentage of SOC to OC (Stone et al., 2009).

Pp. 16944, line 8: This is rather vague. How much is the estimated emission of biogenic VOC globally?

Response: This sentence has been modified as “On a global scale, emissions of BVOCs (1150 Tg yr⁻¹) are suggested to be one order of magnitude larger than those of anthropogenic VOCs (Guenther et al., 1995)”.

Pp. 16944, lines 9-16: This part of introduction is rather poorly written. I feel it reads better if the authors start from the BVOC emissions including isoprene first then move to SOA.

Response: We will carefully rewrite the section Introduction according to the reviewer’s suggestion.

Pp. 16944, lines 15-16: I failed to grasp the meaning of this sentence. How can 'SOA yields' enhance the 'predicted SOA formation'? Do the authors mean that isoprene oxidation products may contribute significantly to SOA even its SOA yields is typically lower than monoterpenes because of its large emission? The authors need to clarify this point.

Response: The sentence has been removed in the revised version.

Pp. 16944, line 22: References are far from complete. I suggest to cite a recently published review by Hallquist et al, 2009 here.

Response: We will cite this review paper and several other references (Hoffmann et al., 1997; Iinuma et al., 2007; Ng et al., 2007; Gómez-González et al., 2008; Sato, 2008) here.

Pp. 16945, line 5-6: I failed to grasp the meaning of this sentence. Does this mean “modeled isoprene concentration is higher at higher altitude than other biogenic SOA precursors” or “modeled isoprene oxidation product concentrations are higher than other biogenic SOA compounds at higher altitude”? The authors need to clarify this point.

Response: We will revise this sentence as follows: “Henze and Seinfeld (2006) reported that inclusion of isoprene as a source of SOA in a global model substantially increases SOA concentrations in the free troposphere, because isoprene, and especially isoprene’s oxidation products, has much greater concentrations at higher altitudes than other biogenic SOA precursors”.

Pp. 16946, line 18: Have the authors synthesized cis-norpinic acid? If so, please provide a reference for the procedure. As far as I know, trans-norpinic acid was available from Sigma-Aldrich in the past but not a cis isomer. The supplier information can be helpful.

Pp. 16946, line 18: If it is trans-norpinic acid, how have the authors achieved the positive identification? Are the retention times for both standard and ambient samples the same? The authors need to clarify this point.

Pp. 16946, line 26: Here, the authors state that trans-norpinic acid is used for the recovery test. Have the authors determined cis or trans-norpinic acid in the samples? This reviewer is not aware of mechanisms forming a trans isomer of norpinic acid from the atmospheric oxidation of monoterpenes. The authors need to clarify this point.

Responses: We appreciate the reviewer’s comments. We just got the authentic *trans*-norpinic acid. After derivatization with BSTFA, the total ion chromatogram of the standard of *trans*-norpinic acid shows two close peaks with almost identical mass spectra. The major peak appears later, which should be *trans*-norpinic acid. The earlier one is minor. The retention times for ambient samples are identical to the earlier peak of the

standard. We will change “norpinic acid” to “*cis*-norpinic acid” in the revision. Supplying information will be added in the revised version.

Pp. 16946, line 19-21: At the very least, the quantification and identification of methyltetrols and MBTCA MUST be performed using the authentic standard compounds. There is no reason to rely on 'surrogate' compounds for these two as the synthetic procedures for methyltetrols and MBTCA are described well and they are not overly complicated. It is not acceptable to use 'surrogate' compounds for the quantification when the target compounds can be readily synthesized (or purchased for that matter) using standard laboratory apparatuses.

Pp. 16946, line 20: Why have the authors decided to use pimelic acid and pinic acid as surrogates? How have the authors determined likely errors originating from the use of these compounds as surrogates? It is hard to accept them as surrogates as both of them are not homologous compounds to MBTCA and beta-caryophyllinic acid.

Responses: For MBTCA, as mentioned above, we correct its concentrations by using the response factor obtained from GC-FID. We use pinic acid as a surrogate of β -caryophyllinic acid because the latter is also a diacid whose chemical structure is similar to pinic acid (Figure 1). Such a quantification method has already been used in other studies (Jaoui et al., 2007; Lewandowski et al., 2007).

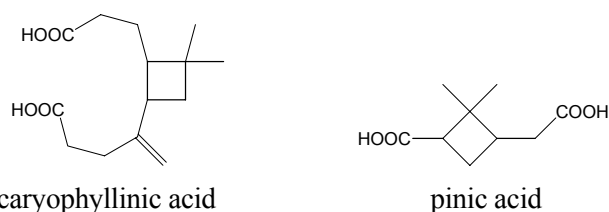


Figure 1. Chemical structures of β -caryophyllinic acid and pinic acid.

Pp. 16946, line 27: The data need to be corrected for the recoveries. Especially, the concentrations of 3-hydroxyglutaric acid and cis-pinonic acid appear to be underestimated at least 30% without the recoveries.

Response: The concentrations of 3-hydroxyglutaric acid are recalculated using an authentic standard. The recovery of 3-hydroxyglutaric acid was $91 \pm 5.5\%$ ($n=3$). Because most of the studies did not correct for the recoveries of the target SOA tracers and other organic species in ambient aerosols (Wang et al., 2006; Ding et al., 2008), we think it is reasonable to keep same technique.

Pp. 16947 onwards: How comparable are these results to the cited studies when the authors have used largely surrogate compounds?

Response: The cited studies (Claeys et al., 2004; Ion et al., 2005; Clements and Seinfeld, 2007; Ding et al., 2008; Kourtchev et al., 2008a; Kourtchev et al., 2008b; Wang et al., 2008; Kourtchev et al., 2009) used the same surrogate compounds as we used. We believe that the results are very much comparable.

Pp. 16948, lines 10-15: There are a number of other papers reporting these compounds.

Response: We will add the citation (Larsen et al., 2001; Fick et al., 2003; Ma et al., 2007) here.

Pp. 16948, lines 18-20: I suggest removing this sentence. There are a number of reasons why the concentration of pinonic acid is higher than that of pinic acid in ambient aerosols and the vapor pressure may have little to do with this observation.

Response: We will remove the sentence in the revision.

Pp. 16949, lines 23-24: Pinic acid seems to show a diel pattern in Figure 1. This is consistent with a recently published article by Kourtchev et al. (2009). In addition, both methyltetrols exhibit a clear diel cycle between May 29th and June 3rd. Some discussion of this is needed here.

Response: We fully agree with the reviewer's comments. We will add more discussions on the topic in the revised manuscript.

Pp. 16950, line 2: Do the authors have mechanisms for this reaction? As far as I know, this is still unproven. It is hard to imagine how hydroxy dicarboxylic acid can be formed from the reaction of dicarboxylic acid and OH in the presence of NO.

Response: The sentence "Malic acid is a secondary oxidation tracer of succinic acid" has been revised as "Malic acid can be produced by the photochemical oxidation of unsaturated fatty acids in the atmosphere".

Pp. 16950, line 23: I suggest changing 'lower-generation' to 'early generation'.

Response: Suggestion is taken.

Pp. 16951, line 7: It is more likely that this originated from a stress (biomass burning) induced emission of sesquiterpenes from vegetation rather than the biomass burning process itself.

Pp. 16951, line 11: This is speculative and should be removed. Why are sesquiterpenes emitted from vegetation fires? Are sesquiterpenes enriched in crop species? The authors need to substantiate this argument with references.

Pp. 16951, lines 13-15: If this is the case, wouldn't isoprene and monoterpene oxidation products show the same trend? Why is beta-caryophyllinic acid the only compound influenced by biomass burning OC? As the authors stated, pinonic acid has much higher vapor pressure than other compounds detected here and its gas/particle partitioning is more likely influenced by biomass burning OC compared to beta-caryophyllinic acid.

Responses: We will delete the sentence, and will add the following sentences: "Interestingly, levoglucosan and β -caryophyllene oxidation products showed a positive correlation, indicating that β -caryophyllinic acid detected over Mt. Tai mainly originated from biomass burning process in early summer or from the same source regions. However, no correlations were found between levoglucosan and isoprene or monoterpene SOA tracers. This is reasonable because the emissions of isoprene and monoterpenes are insignificant for most of the widely planted crop species (Kesselmeier and Staudt, 1999). Although little is known about the specific sesquiterpene emissions of wheat straws, crop species such as corn and potato have been found to emit sesquiterpene species including β -caryophyllene (see Duhl et al. 2009, and references therein). Studies examined the temperature-dependency of sesquiterpene emissions have found that emissions are positively correlated with ambient temperatures (Duhl et al., 2008). Thus, the active field burning activities of wheat straws in the CEC during early summer may release levoglucosan and sesquiterpenes such as β -caryophyllene to a certain quantity through either combustion process or increasing leaf temperature, which is still poorly understood and warrants further study".

Pp. 16951, line 23: Please provide references.

Response: We will add three references (Blake et al., 1994; Andreae and Merlet, 2001; Yan et al., 2008) here.

Pp. 16951, Hierarchical cluster analysis (HCA): How have the authors performed the HCA analysis? Detailed information about the procedure must be given.

Response: We will add the following information in the revision: “Hierarchical cluster analysis (HCA) is a classification method used to divide the data into clusters. The software SPSS was used as a statistical analysis program. HCA was performed with the Ward’s method using the standardized squared Euclidean distance as a grouping criterion”.

Pp. 16952, line 5 and lines 13-14: Pinonic acid and pinic acid may also originate from ozonolysis of alpha-pinene. Pinonic acid is unlikely to be formed from the oxidation of beta-pinene.

Pp. 16952, line 15: I suggest changing higher-generation’ to ’later generation’.

Responses: These comments are taken.

Pp. 16952, Enhanced contribution of isoprene oxidation products: I am not convinced that the “ $R_{iso/mono}$ values” determined in this study can be used anything meaningfully when only a small group of rather minor alpha- or beta-pinene oxidation products are determined. The oxidation of both isoprene and monoterpenes are rather complex and their product distributions are strongly influenced by ambient conditions (i.e. T, RH, oxidants, NO/NO₂/HO₂/RO₂ etc.). The authors may compare the results obtained in this study to the results from other regions but it goes a little too far to suggest “ $R_{iso/mono}$ values” to be used for estimating the contribution of isoprene and monoterpenes to biogenic SOA formation in various ecosystems when only a limited number of compounds are determined using “surrogate” compounds.

Response: Actually we proposed the “ $R_{iso/mono}$ values” to evaluate the relative contribution of isoprene-derived and monoterpene-derived SOA tracers to atmospheric aerosols. We compare our results with the results reported by other studies because most of the studies have reported the same spectrum of SOA tracers by using the same approach (GC/MS). As also suggested by reviewer 2, we will weaken our statement and rephrase the sentence as “Thus, we propose here that the $R_{iso/mono}$ value can be used to estimate the relative contribution of isoprene and monoterpenes to biogenic SOA formation in different locations”.

Pp. 16952, line 24: It is misleading to state ’monoterpene oxidation tracers’ as the authors have determined only a small group of alpha- or beta-pinene originating oxidation compounds.

Response: In the revision, we will use “ α/β -pinene oxidation tracers” instead of “monoterpene oxidation tracers” accordingly.

Pp. 16953, lines 15-16: Do the authors mean isoprene is the ’missing’ large source of organic aerosol in the FT?

Response: Yes, our results provide the direct evidence that isoprene oxidation products are one of the dominant species of SOA in the FT.

Pp. 16953, lines 17-19: More measurements are needed at different locations to substantiate the authors’ claim.

Response: We will modify the sentence as “This also suggests that the isoprene oxidation products are more abundant than other biogenic SOA in the atmosphere at high altitudes in Central East Asia”.

Pp. 16953, line 20: What do the authors mean by 'enhanced SOA formation'? This is simply 'higher concentrations of isoprene oxidation products'.

Response: We will modify the sentence “The enhanced SOA formation from isoprene may be...” to “The higher concentrations of isoprene oxidation products may be” in the revised manuscript.

Pp. 16953, lines 24-26: This sentence is unclear. Is it a mass concentration or a number concentration? Is it the highest in China or the world? Are there any connections to the accumulation mode particles and the high concentration of isoprene oxidation products?

Response: It is a number concentration, not a mass concentration. We would like to keep this sentence in the revision because it provides useful information on the studied region, although little is known about the connections between accumulation mode particles and the high isoprene oxidation products. We will modify the sentence “Aerosol concentrations in accumulation mode are suggested to be the highest in CEC” into “Aerosol accumulation mode number concentrations in CEC are suggested to be among the highest regions in the world”.

Pp. 16954, line 4: Are they concentrations or mixing ratios? The mixing ratios (or concentrations) are not a good indicator for the completeness of the reactions under atmospheric conditions. How have the authors determined the conversions of isoprene and monoterpenes without initial mixing ratios for them?

Response: The mixing ratios of non-methane volatile organic compounds including isoprene and monoterpenes over Mt. Tai during MTX2006 have been reported by (Suthawaree et al., 2009). Unfortunately, we didn't have any information on the initial mixing ratios for BVOCs. We will delete “especially isoprene” in the revised manuscript.

Pp. 16954, line 6: This part contradicts the authors' earlier statement (Pp. 16949, lines 23-24). Indeed, the temperature can be important for the gas/particle partitioning but the argument put forward by the authors to rationalize higher isoprene tracer concentrations in the nighttime samples than the daytime samples is unsubstantiated. If this is the case, why should monoterpene oxidation products behave differently? 2-Methyltetrols are known to be present predominantly in the particle phase and are unlikely to be influenced by temperature yet they also show higher concentrations in the nighttime samples according to Table 2. If you pay close attention to Figure 1, the opposite trend (i.e. higher isoprene oxidation products during the daytime) can be seen between May 29th and June 3rd. It is evident that the higher nighttime average concentrations of isoprene SOA tracers summarized in Table 1 are likely caused by the extremely high concentrations observed on June 20th. Furthermore, are the differences statistically significant, especially for monoterpenes?

Pp. 16967 Fig. 1: It helps the readers to differentiate the day and night samples if the authors could use e.g. a black filled circle for the night time samples.

Responses: We have carefully checked both Figure 1 and our original datasets. From May 29th to June 3rd, higher concentrations of isoprene oxidation products during the nighttime can be observed, especially for 2-methyltetrols. The extremely high concentration was also observed during the nighttime of June 20th. On the contrary, α -pinene oxidation products such as pinic acid showed relatively lower concentrations during the nighttime.

As pointed out by the reviewer, it really helps if we could use a black filled circle for the night time samples in Figure 1, Page 16967. We will make such a change in the revision.

Pp. 16954, lines 12-26 and Figure 5: It is hard to see any relationship between the mixing ratios of O_3 and NO_x , and $R_{iso/mono}$ from Figure 5. It is much more informative to show scatter plots when there is a relationship between two variables. This part is largely a collection of random information and it leaves readers to interpret the data presented in this study. Both OH and O_3 were high during the sampling period. How do they relate to high $R_{iso/mono}$. Does the ratio of OH to O_3 relate to the compound distribution? How does the ratio of NO to NO_2 change? Does high NO relate to the presence of some compounds? Does high O_3 relate to high temperature?

Response: We appreciate the reviewer's comments. We have modified the hour-based mixing ratios of O_3 to day- and night-based values. Although there is no linear correlation found between O_3 and $R_{iso/mono}$, we still believe it is valuable to keep the temporal trends of O_3 and NO_x in the figure. The temporal variations of the mixing ratios of OH and the ratio of NO to NO_2 showed clear diurnal patterns with higher values in daytime (see the following Figure 2). There are no correlations observed between these parameters and SOA tracer compounds. The diurnal variations of ambient temperature showed higher daytime values around 13:00. while the diurnal pattern of O_3 showed higher As suggested earlier by the reviewer, the information on the temporal variations of NMHCs/ NO_x will be added in the revised manuscript.

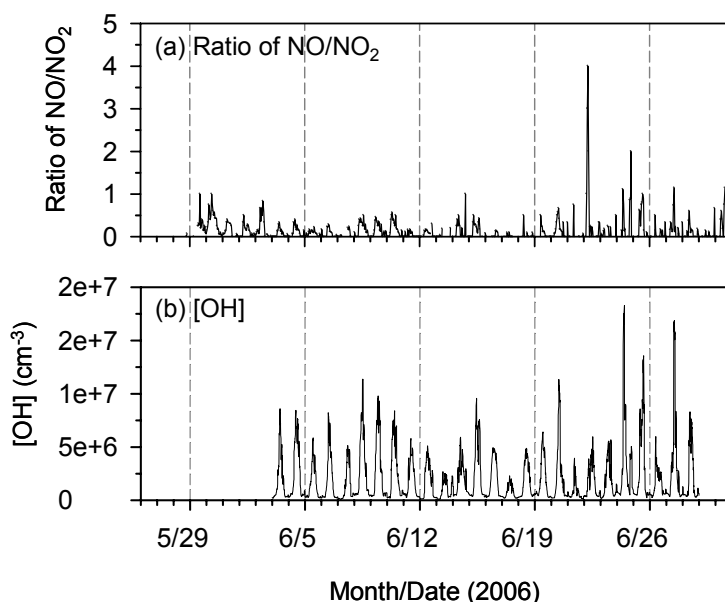


Figure 2. Temporal variations of (a) Ratio of NO/NO₂, and (b) OH during the campaign.

Pp. 16954, lines 29: Were air masses passing through the sampling site significantly influenced by cloud chemistry? Alternatively, have the authors determined higher isoprene oxidation products when the sampling site was covered in polluted cloud? The authors need to clarify this point.

Response: During sampling, brown clouds (polluted clouds) were occasionally observed uplifting from the mountain waist to the mountaintop from late afternoon to early evening. This point will be added in the revised manuscript.

Pp. 16956 Estimated contributions of BVOCs to secondary organic carbon: The authors need to clearly state that the numbers presented here are rough estimation and contain large uncertainties.

Response: We will add the following information in the revision. "The uncertainties of this approach range

from 20 to 48%, based on the standard deviation of f_{soc} mentioned above (Kleindienst et al., 2007a).”

Pp. 16957 line 4: Why should the Mt. Tai data be consistent with the data obtained in North Carolina?

Response: We will modify the sentence as “This is similar to the results reported by...”. The organic molecular compositions of secondary organic aerosol and their concentrations in the atmosphere largely depend on the vegetation area, plant species, and meteorological conditions or trace gases, and so on. Thus, we will add more discussions here in the revised manuscript.

Pp. 16957 line 18-Pp. 16958 line 7: This section is largely a descriptive summary of the manuscript rather than conclusions.

Pp. 16957 line 24-Pp. 16958 line 3: It is hard to draw a conclusion from the authors’ data if the high OC or pollutants had any influence on the gas/particle partitioning of SOA. Conclusions need to be drawn solely from what the data actually shows.

Response: We removed these sentences in the revised manuscript. Instead, we will briefly mention the limitations (such as a lack of information on organosulfates) and future needs in the section Conclusions.

Pp. 16971 Fig. 5: “Concentration” should be “mixing ratio”

Response: “Concentration” will be reworded as “mixing ratio”.

References

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