Responses to Referee 2

We are thankful to the reviewer for his/her thoughtful comments and suggestions.
We have revised the manuscript accordingly. Listed below are our point-by-point responses in blue to the comments. The modified parts in the revised manuscript are highlighted in yellow.

RC2: 'Comment on acp-2021-136', Anonymous Referee #2, 31 Mar 2021 reply
The authors present measurements of aerosol mass and composition (i.e., tracer concentrations) at a tall tower in Beijing. Measurements of vertical distributions, as presented here, are valuable and generally fairly scarce, so presenting these measurements is itself of value to the community. I believe the work is useful and worth publishing in this journal, but suffers from some scientific overreach that needs to be addressed first. Specifically, as described below, the authors need to temper the strength of some of their statements to more accurately reflect the strength of their evidence, and the authors need to re-evaluate some of their interpretation of tracer ratios by either providing support from the literature or correcting their claims.
We thank the reviewer's comments and valuable suggestions. All comments and suggestions have been considered carefully and addressed below.

General comments:
1. It is a little confusing that the methods discuss Parade-based periods, but most of the paper actually is more about the pollution episodes.
Thanks for the reviewer's comments. We have discussed the properties of SOA tracers during the pollution events in Section 3.1 and the impacts of emission controls on SOC in Section 3.4 in the revised manuscript. We think that the reduction measurements during the Parade provide a unique chance to study SOA under the government interventions as mentioned in the last paragraph in Section 1. These results are conducive to the making of reduction policies.

2. There is a fair amount of English-language issues, mostly odd phrasing and the like, that should be cleaned up.
Thanks. We have revised the language of our manuscript that are highlighted in yellow. We hope the revised manuscript can meet the requirement of the journal.

3. Interpretation of tracer data is somewhat confusing and does not seem accurate to me, particularly in the case of the isoprene tracers. In particular, the authors' interpretation of the 2-MT/2-MET and the 2-MTs/C5-ATs ratio are not, to the best of my knowledge, grounded in recent literature on the sources of these tracers, and the citations provided by to the authors do not support their interpretations as far as I can tell.

Thanks for the reviewer's comments. We have rewritten Section 3.1.3 in the revised manuscript. We have deleted the discussion of 2-MT/2-MET and added some recent references about the ratio of 2-MTs/C_5-alkene triols. We hope these modifications can improve the quality of our manuscript.

4. The authors seem to draw fairly broad conclusions from somewhat limited evidence. While the vertical distributions are certainly interesting, some of the assumptions regarding regional vs. local transport, changes in partitioning, impacts of primarily particles, etc., are not very strongly supported. In some cases, these claims seem to based off of previous work, but that is not always clear. In other cases, these claims are based on tracer ratios, but as described above, it's not clear these claims are always based on a current understanding of the current tracer literature.

We have noticed the confusion of some claims in our study, and we have rewritten these claims with a clearer expression. In addition, as mentioned above, we have rewritten our description of tracer ratios and added some current literature in the revised manuscript. Please see these changes in Section 3.1 in our revised manuscript. We hope these changes can meet the high standard of the journal.

Specific comments:

P1L29: This is a very broad statement, which is fine as an opening sentence, but why are these citations specifically selected? Some of the early ones make sense, but, for instance, what is the information being conveyed by the Huang et al. reference?

We are sorry that we have cited this reference in an improper place. We have deleted it and we have checked other references in our manuscript. We hope that our manuscript has no this problem again.

P2L6: extra "a"

Thanks. We have deleted it in the revised manuscript

P2L9-10: This distinction between ASOA and BSOA is a bit out of date, in the years after these citations the idea of ABSOA being dominant became somewhat more accepted. The next sentence clarifies this a bit, but the statement that 90% of SOA is BSOA is more a historical perspective than actually informative so should be re-framed as such or removed.

Thanks for the comments and suggestions of the reviewer. We have re-framed this sentence to "Anthropogenic SOA (ASOA) and biogenic SOA (BSOA) are important contributors to OA and air pollution in the atmosphere (Huang et al., 2014; Volkamer et al., 2006)" in the revised manuscript (on page 2 lines 10-11).

P3L6: should say decision "makers"
Thanks. We have modified it.

P3L12: What is the basis for the claim that 120 m and 260 m are regional? There are two citations - do they measure boundary layer height? Or model it? Or measure tracer compounds in some way?
These two citations (Sun et al., 2015; Zhao et al., 2020) have measured tracer compounds and showed profiles of meteorological conditions based on the 325-m meteorological tower in the boundary layer. These results highlight that the measurements at 260 m are more representative of regional sources whereas the ground measurements are more subject to local sources. Hence, the claim here was based on the citations.

P3L27: I'm not sure "the typical urban site" really means anything. The following description of the site is more useful.
We have modified this to "an urban site" and added more descriptions about the sampling site. Please see "The sampling site is at the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences (39°58.53′N, 116°22.69′E), which is in an urban site (between 3- and 4-ring) of Beijing and surrounded by street road (~50 m), highway (~300 m), a public park (~500 m to the southwest), restaurants (~100 m), residential housing and a gas station (~200 m). The predominant vegetation types surrounding the sampling site are deciduous broadleaf vegetation (acacia and juglandaceae), shrub, and lawn. The vegetation cover of the public park is more than 50%. The predominant vegetation is also deciduous broadleaf." in Section 2.1 in the revised manuscript (on page 3 liens 23-28).

P4L6: "OC and EC in an aliquot filter" is phrased oddly and should be re-stated
We have rewritten this phrase to "OC and EC in aerosols" in the revised manuscript (on page 4 line 4).

P4L17: Are the author's sure it is a Hewlett-Packard? The last HP GC I was aware of, at least in the United States, was the 5890, and I thought subsequent GCs and MSs were all sold under the "Agilent" branding (i.e., Agilent 7890 GC and Agilent 5975 MS). But perhaps it is different in other countries?
Thanks for the comments of the reviewer. Yes, our instrument was bought from "Agilent" branding, but the mainboard is Hewlett-Packard. We have modified it to "a Hewlett-Packard model Agilent 7890A GC coupled to Hewlett-Packard model Agilent 5975C mass selective detector (MSD)." in the revised manuscript (on page 4 lines 15-16).

P4L24: Were they not corrected for recoveries because recoveries was near 100%? That should be stated if so.

Many SOA standards are not commercially available. As mentioned in the reference (Fu et al., 2009), only several SOA standards are available, other SOA tracers were obtained by comparison with those of literature data or by using the surrogates. Hence, we did not add standards in our laboratory into the samples, we only added the internal standard C_{13} n-alkane. It is why recoveries were not used for correction in our manuscript.

P5L20-23: These sentences seem to contradict, claiming both increase with height and no significant differences with height.

To avoid ambiguity, we have modified this sentence to "The concentrations of WSOC and OC were 2.73 ± 1.31 μgC m^{-3} and 5.03 ± 2.28 μgC m^{-3} at 260 m, 2.69 ± 1.55 μg m^{-3} and 5.32 ± 2.88 μg m^{-3} at 120 m, and 2.03 ± 0.99 μg m^{-3} and 4.37 ± 1.69 μg m^{-3} at 8 m, respectively" in the revised manuscript (on page 5 lines 23-24).

P527-30: Sometimes it is not clear to me when the authors are making a new claim, vs. stating a previously published result. This statement is one of those examples - is the claim that the lower WSOC:OC ratio at ground level is due to biological aerosols a claim made (and presumably supported) by Wang et al., or is that a new claim here?

We are sorry for this confusion. It is a new claim, but we used a confusing way of writing. We have changed this sentence to "In addition, primary sources from local dust and soil resuspension, such as primary biological aerosols which contain a high abundance of water-insoluble organic compounds (Wang et al., 2019), potentially caused the lower fractions of WSOC to OC at the ground surface than at the upper layers." in the revised manuscript (on page 5 line 31 to page 6 lines 1-3). We also revised the same problem in our manuscript, we hope these changes can give a clearer expression for the claims.

P6L4 and P6L29: This sentence is a bit misleading - some monoterpene tracers decrease, but others (MBTCA, HDCCA) increase. Also, there is only one sesquiterpene tracer, so it is a bit tough to make general claims like this.

Thanks for the reviewer's comments. We have modified our expression to "Most of these molecular tracers showed higher abundance at high layers (≥ 120 m) than at 8 m, except for pinic acid, pinonic acid, 3-acetuldipic acid and β-caryophyllinic acid. Table S2 shows significant differences in the average concentrations of these SOA tracers with height, except for monoterpene SOA tracers." in the revised manuscript (on page 6 lines 6-9). We hope this expression can be more appropriate.

P7L4: The claim that isoprene is regional and MTs/SQTs are more local is not necessarily true. As the authors note, the vertical distribution could be due to regional transport, but conversely could be due to vertical differences in chemistry and/or partitioning.

Thanks for the valuable suggestions of the reviewer. We have changed this claim to "It suggests that regional transport potentially contributes more to isoprene SOA, while SOA from monoterpenes and sesquiterpene is likely more influenced by local sources. In addition, some other factors (such as transformation and condensation processes) can also lead to these patterns." in the revised manuscript (on page 7 lines 22-24). We hope this expression can be more appropriate.

P7L26: Is there a citation for the claim that sesquiterpenes are mainly emitted by crops and herbs? I'm not sure that is true, they are released from many plants, particularly for reasons related to chemical signaling and plant protection (e.g., increase SQTs with herbivory: Faiola et al, 10.1021/acsearthspacechem.9b00118)

Thanks for the comments of the reviewer. A previous study has mentioned that "The SQT emissions distribution is strongly influenced by the grass and crop PFT." (Sakulyanontvittaya et al., 2008). However, it was not suitable to claim that sesquiterpenes are mainly emitted by crops and herbs in our manuscript. We are very thankful for the remainder of the reviewer. Many factors can influence the emission of sesquiterpenes (Duhl et al., 2008; Faiola et al., 2019). Hence, we have modified our claim to "Sesquiterpenes are mainly emitted from plants and trees, which are controlled by many factors, such as temperature and stage of plant growth (Duhl et al., 2008; Faiola et al., 2019)." in the revised manuscript (on page 6 lines 26-28). We have deleted the explain here in the original manuscript.

P8L4: The phase" methacryloyl peroxynitrate (MPAN, e.g. methacrolein, methyl vinyl ketone and methyl butanediols)" is odd, as those latter species are not a subset of MPAN but rather separate compounds

Thanks for the reminder of the reviewer. We have added new references and modified this sentence to "The isoprene oxidation mechanisms are dependent on atmospheric conditions (Bates and Jacob, 2019; Wennberg et al., 2018)." in our revised manuscript (on page 8 lines 1-2).

P8L7: I find the use of 2-MT to mean 2-methylthreitol while 2-MTs means the sum of both isomers to be confusing. I would recommend calling the sum 2-MTs (which is fairly standard) and maybe calling the isomers 2-MT_eryth and 2-MT_threi (where "_X" denotes a subscript).

Thanks. According to the reviewer's suggestion, we have changed the abbreviation of these two isomers in our revised manuscript.

P8L16: I am not aware of work showing that the 2-MT/2-MET ratio is indicative of anything in particular. The two citations in this sentence do not seem to include such claims either. As someone who has thought a fair amount about isoprene and monoterpene tracers, it's not clear to me what this ratio is telling me, or why the authors include it.

Thanks for the reviewer's comments. We have detected the discussion of 2-MT/2-MET ratio and modified figure 5 in our revised manuscript.

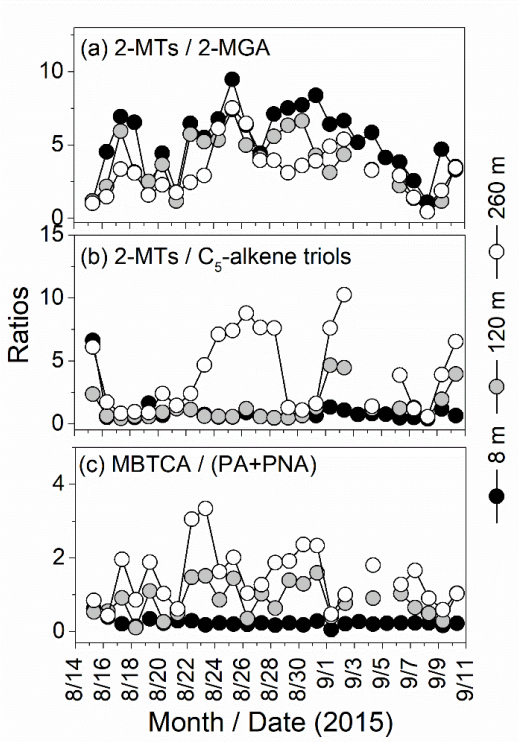

Figure 5. Temporal variations in the mass concentration ratios among different biogenic SOA tracers in $PM_{2.5}$: (a) 2-MTs / 2-MGA; (b) 2-MTs / C_5-alkene triols and (c) MBTCA / (PA+PNA).

P8L20-23: The interpretation of C5-alkene triols as precursors in the oxidation of 2-MTs is confusing to me, to the point of making me feel the authors are interpreting their tracer data through an outdated lens. Since the Wang et al., 2005 paper, lots of work has been done on IEPOX oxidation pathways, and I'm not aware that any of it has made the claim the authors are making here. Even in the Wang et al., 2005 paper, Scheme 1 shows both C5-ATs and 2-MTs to be products of IEPOX (one through addition and one through rearrangement). Since then, there has been a fair amount of work to understand what C5-alkene triols are actually "telling us", in particular from the Surratt group and Goldstein group, and I think both groups would agree it's still not quite clear. See for example: Cui et al. doi.org/10.1039/C8EM00308D and Yee et al. 10.1021/acs.est.0c00805.

Thanks for the reviewer's comments. We have searched the references mentioned by the reviewer. The formations of C_5-alkene and 2-MTs are not quite clear, and many factors (such as aerosol acidity and humidity) can influence the ratios of 2-MTs/C_5-alkene (Cui et al., 2018; Surratt et al., 2010; Yee et al., 2020). Hence, we have rewritten the paragraph about 2-MTs and C_5-alkenes. Please see "The average ratios of 2-MTs to C_5-alkene triols were 0.97±1.17, 1.33±1.24, and 3.97±3.08 at 8 m, 120 m, and 260 m, respectively (Figure 5b). C_5-alkene triols have been suggested to convert into 2-MTs (Wang et al., 2005). Some studies also suggested that the loading of 2-MTs increased with the enhancement of aerosol acidity (Surratt et al.,

2007), and the relative humidity can affect the ratio of 2-MTs to C_5-alkene triols (Surratt et al., 2010). Recent studies suggested the ratio of 2-MTs / C_5-alkene triols decreased with aerosol acidity (Yee et al., 2020), and C_5-alkene triols were likely formed from thermal degradation of 2-methyltetrol sulfates for GC/MS artifacts (Cui et al., 2018). Hence, it is still not clear the meaning of the ratio 2-MTs to C_5-alkene triols. However, the large differences of 2-MTs / C_5-alkene triols values at three heights highlight the significance of studying vertical profiles of SOA, and more field investigations are needed." in the revised manuscript (on page 8 lines 15-22).

P8L23: Typo: "vitations"

Corrected.

P10L28: It would be helpful in the figures and pie charts about source apportionment if they also included what fraction of OC and/or WSOC was not captured by the source apportionment. I think this sentence here is telling me that only 8-13%% of SOC is accounted for in their source apportionment, but it's not totally clear to me.

Thanks for the reviewer's comments. This sentence tells the fractions of estimated SOC in OC. We have added the uncaptured fraction of OC in figure 7 in our revised manuscript according to the suggestion of the reviewer.

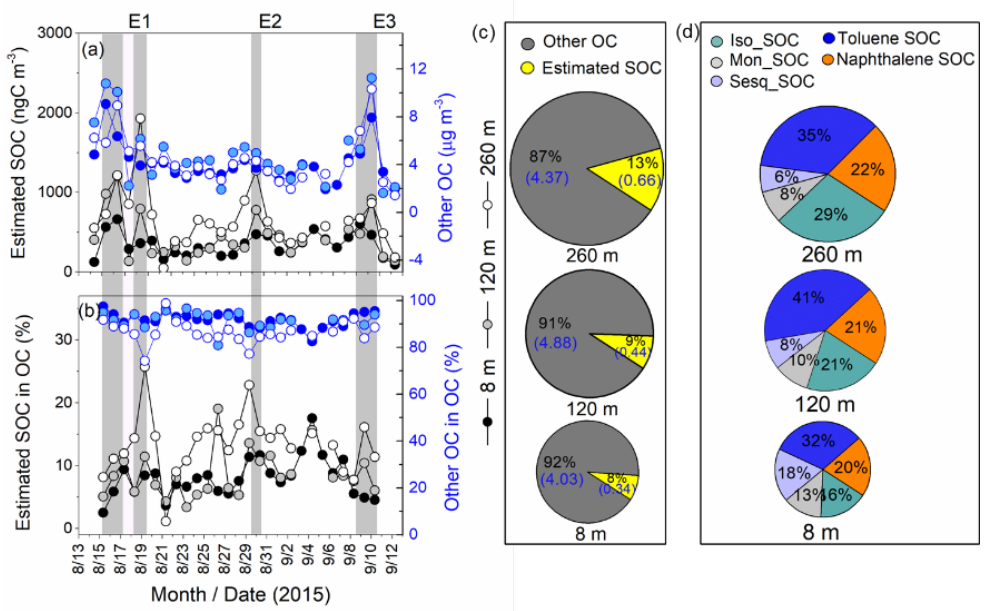

Figure 7. Temporal variations in the estimated SOC and other OC at three heights: (a) the concentrations of estimated SOC (right axis) and other OC (left axis), (b) the fraction of estimated SOC and other OC in OC. Relative mass fractions of OC and estimated SOC is shown in (c) and (d). Other OC is not captured by the source apportionment. Iso_SOC, Mon_SOC, and Sesq_SOC represent BSOC estimated from isoprene, monoterpenes, and sesquiterpene, respectively. Toluene SOC and naphthalene SOC represent ASOC that were estimated from DHOPA and phthalic acid, respectively.

P11, Sect. 3.4: Are these reductions in total WSOC, or just the fraction of SOC that is captured in the source apportionment?

Thanks. These reductions are the fraction of SOC that is captured in the source apportionment.

Figures:

HDCCA is not defined anywhere in the main text

We defined HDCCA in table S1 in the support information. HDCCA is the abbreviation of 3-(2-hydroxyethyl)-2,2-dimethyl-cyclobutane carboxylic acid. We have added this sentence in the caption of Figure 4 in the revised manuscript.

Figure 5. The caption is in the wrong order, and the description of panel (d) is tough to understand.

Thanks. We have changed the order and the description of panel (d). The revised caption is "Figure 5. Temporal variations in the mass concentration ratios among different biogenic SOA tracers in $PM_{2.5}$: (a) 2-MTs / 2-MGA; (b) 2-MTs / C_5-alkene triols and (c) MBTCA / (PA+PNA)." In the revised manuscript.

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