Comment on acp-2021-136
Anonymous Referee #2

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.

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.

2. There is a fair amount of English-language issues, mostly odd phrasing and the like, that should be cleaned up.

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 the authors do not support their interpretations as far as I can tell.

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 be 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.
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 makes sense, but, for instance, what is the information being conveyed by the Huang et al. reference?

P2L6: extra "a"

P2L9-10: This distinction between ASOA and BSOA is a bit out of date, in the years after these citations the idea of ABSOA being dominant became somewhat more accepted.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.

P3L6: should say decision "makers"

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?

P3L27: I'm not sure "the typical urban site" really means anything. The following description of the site is more useful.

P4L6: "OC and EC in an aliquot filter" is phrased oddly and should be re-stated

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?

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

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

P5L27-30: 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 WSO: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?

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 generally claims like this

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.

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, particulary for reasons related to chemical signalling and plant protection (e.g., increase SQTs with herbivory: Faiola et al, 10.1021/acsearthspacechem.9b00118)

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

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).

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.

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.

P8L23: Typo: "vitations"

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 appportionment. 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.

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

Figures:

HDCCA is not defined anywhere in the main text

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