D. Liu and co-workers have used filter sampling and mass spectrometry to study the composition of secondary organic aerosol (SOA) material formed from the ozonolysis of beta-pinene and limonene at different ozone concentrations. The topic is in principle highly atmospherically relevant, and the manuscript contains many interesting and publishable results. Unfortunately, the manuscript is in several places quite poorly written, and it is in places difficult for the reader to extract the genuinely novel results from the presented data. I’m happy to recommend publication of this manuscript in ACP, but only after a rather thorough revision including substantial copy-editing and scientific proofreading.

The overall problem in reading the manuscript is that a substantial fraction of its sentences sound sensible, exact and scientific at a quick glance, but on closer inspection are at best very vague, and at worst ill-defined, illogical or meaningless. Sometimes the actual intent and meaning of the writers can be deduced by clever guesswork (e.g. by correcting some rather trivial word order issues), but in other cases even a thorough and repeated reading leaves the reader confused. The following examples are not intended to be an exhaustive list, but simply illustrative examples of this overall problem (note: I mostly gave up on listing problematic sentences after the first 3 pages or so, so the examples are weighted toward the beginning of the text - while the introduction is arguably the most problematic part of the paper this should not be construed to mean that the results - section is problem-free):

- Line 17, “abundance”, please be more specific - exactly what abundance is meant

- Line 18, “prefer to stabilise”. As written, this would imply that the HOM molecules themselves would preferentially be stabilised (e.g. collisional, thermally, kinetically etc) at some certain [O3], but what is meant is that their yield saturates with respect to [O3] - not the same thing.
-Line 23, “formation of compounds with 10 carbon atoms“. As the precursors themselves contain 10 carbon atoms, no mechanism is needed for their formation. Presumably the authors mean the formation of compounds with 10 C atoms AND SOME NUMBER OF O ATOMS.

-Line 38: "wide range of volatility, which has a strong temperature-dependence“. What is meant by this - that an individual saturation vapor pressure has a strong temperature dependence (generally true), that the variation/range in volatility has a temperature dependence, or perhaps both? Whatever is meant, what is the relevance of the temperature-dependence to the rest of the discussion?

-Line 39: “ambient species are thought to consist mainly of low volatility species”, apart from the grammar issue (species…species) this is not even true as written - the ambient air certainly contains a large range of species (as indeed argued in the previous sentence!) all the way from fully volatile to ULVOC. I assume the authors do have an actual argument here, the presentation is just missing some key assumptions or concepts (e.g. do they mean the species present in ambient SOA?).

-Line 43 “ozonolysis as the most effective”, “as” should presumably be “is”, and also this sentence needs some caveats - for which compounds and in which conditions is ozonolysis the most effective, and compared to what (presumably OH and NO3 oxidation)?

-Line 45, “BVOCs forming ULVOC even in the absence of H2SO4”, it’s not the ULVOC formation in the absence of H2SO4 that is the surprising fact, but the production of aerosol particles in the absence of H2SO4. (Here I again assume the authors do actually mean the latter, they just wrote the sentence incorrectly.)

-Line 52, ”high oxygen-containing but low oxidation state determining the oxidation potential”, here I have no idea what the authors mean, this seems to make no sense.

-Line 54, “HOMs refer to the process“ - no, HOMs refer to a certain subgroup of the products of this process (and indeed that seems to be the definition the authors are using)

-Line 57, “precursors decomposing into aerosols“, certainly this is not what is going on

-Line 58, “Laboratory studies of HOMs observed by ozonolysis of monoterepenes are closely corresponded to”... this phrasing makes no sense.
-Line 60-65: the science is correct here, the phrasing is just very very poor.

-Line 66, “HOMs can provide nucleation conditions for early growth”: no, the HOMs can nucleate (or not), or participate in early growth (or not), this phrasing about providing conditions is meaningless.

-Line 70 “an exocyclic bond as the second-most-abundant VOC” - no it’s beta-pinene (not its exocyclic double bond) that’s the VOC.

-Line 80 “nucleation rate of monoterpene SOA” - SOA is by definition material that has already formed particles (e.g. nucleated), what is presumably meant (though not measured here directly) is the nucleation rate of monoterpene-derived SOA precursors, such as the HOM discussed above. These are not the same thing.

-Line 94, “obtain reaction mechanism”, this is incorrect - the mass spectra just gives the molecular compositions, these can then often be used to indirectly infer something about the reaction mechanism but claiming that the mechanism is “obtained” is wrong.

-Line 207: “high ozone concentration tends to increase oxygen reaction”, this seems to be almost trivially true - maybe the authors need to specify what is meant by “oxygen reaction” here.

-Line 208, “may indicate the importance of ozone... by ozonation”. Ozone is by definition important in ozonation, so this sentence as written is tautologically true (and thus meaningless).

-Line 215, “peak intensity of MW” - presumably what is meant is the MW (molecular weight) corresponding to the peak of highest intensity (not the same thing).

-Line 215, “maximum proportion”, what is meant by proportion here?

-Line 235, “limonene is preferred to proceed oxygenate and accretion reaction”, what is meant by this?

-Line 279, “front” should presumably be “former”
-Line 323-326, The first sentence here lists possible gas-phase formation pathways for (HOM) monomers and dimers. The second sentence then mentions rapid particle-phase reactions of some of the monomer and dimer types. The third sentence says “Due to the high activity of these pathways, the dimers...are expected to be distributed directly onto particles after gas-phase production”. While I agree that the dimers will be “distributed directly onto particles”, surely this is due to their low volatility, and not the rapidity of the subsequent reactions? How could a dimer still in the gas phase be affected by the reactions going on in the particle phase?

-Line 339, “monomers with CH2”, what is meant by this?

-Line 347, “The presence of...” this whole sentence does not seem to make sense - please rephrase, rewrite and give more background as this is potentially a quite important point! The next sentence is also very long and hard to parse - please split it up into two or more sentences.

-Line 375, “almost hardly”, just “hardly” is enough

-Line 378, “related to the way of broken bonds” please rephrase

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Other technical or notation-related questions and suggestions:

-Please note in the abstract that measurements were made from filter samples (as many readers may initially assume the gas phase is being probed).

-Line 17 in the abstract, “5-13% higher than” - should there be a comma here? Without
the comma this literally means that the abundance (see above for a note on this term) of HOMs in limonene SOA was 5 to 13 percentage points higher than the abundance of HOMs in beta-pinene SOA (a claim not actually validated by the numbers in the text). With a comma this just more vaguely implies that the former is in general higher than the latter (which would seem to be true).

-Line 90, what is meant by “gradient concentrations”? Just that there were three different concentrations? OR something else?

-Do the authors expect ROOR or ROOH - type compounds to fragment in their ionisation setup, as recently predicted even for milder chemical ionisation (https://doi.org/10.5194/amt-15-1811-2022)? If not, why not?

-Line 145, please redefine BDE here (it’s defined in the intro but that is easily missed)

-What are the Int_i weights used in equation 4? I couldn’t find the actual numbers anywhere. Also on line 205 the weighed average is mentioned with reference to Table 1, but I don’t see the weighted average numbers in that table.

-What are the likely error margins of the (useful but crude) volatility estimate of equation 5? At least a couple of orders of magnitude I assume, since functional group identities are completely ignored?

-Line 204, “plausible different partition and agglomeration kinetics”, can you be a little more specific, and e.g. suggest which mechanisms could lead to the observed difference?

-Line 212, “number of organic molecules” - first I thought this was a mistake, but it seems the authors actually do mean the number of distinct molecules, i.e. the number of different elemental compositions measured (above some noise threshold). This might be good to specify explicitly here, to avoid misunderstandings (e.g. that “number” would mean “number concentration” or similar).

-Line 231, “crack” has a very definite meaning in hydrocarbon chemistry, “fragment” is probably the word needed here.

-Line 249, “It seems” - could the authors speculate on the reason/mechanism of this inhibition? Could for example the OH produced by ozonolysis begin to cause more fragmentative oxidation at high O3 levels?
-Line 259, “overoxidation” - not a well-defined concept - I think I understand what is meant but the authors should still spell it out

-Lines 270-280, “dimer” often misspelled as “dimmer”, please correct

-Line 273, “This may be due”: this is almost certainly the explanation (already included in standard chemical mechanisms such as MCM) so the sentence could be a bit stronger here.

-Section 3.4, accretion reactions are probably going on BOTH in the gas phase and the particle phase (and potentially some gas-phase accretion products are, in parallel, broken up in the particle phase). So the measured particle-phase “dimers” may be quite different from the original gas-phase ones. (This is certainly implied already by the present discussion, but could be explicitly stated.)

-RA_HOM in the conclusions is not defined, please spell out what is meant.

-The last paragraph of the conclusions seems to be very general, and unrelated to the specific study performed here. While the presented arguments are correct, would it fit better in the discussion or even the introduction?

-Figure 8, WHAT are the markers colour-coded to? Please specify.

-Figure 10, all the proposed channels are oxygen-increasing, and ALL of them proceed through the initial (well-established) Criegee intermediate => vinyl hydroperoxide => vinoxy radical + OH => peroxy radical sequence. I thus fail to understand why some of the channels are denoted “Criegee channels” and “OIR” while others are not. (I would call the right-hand-side for example alkoxy channels - “hydroperoxy channel” is an acceptable name for the left-hand-side.) Also, wouldn’t the alkoxy radical form equally well in reactions with HO2 or NO - or are these concentrations known to be low by the authors? Finally, the postulated H-shift from a C4 carbon in the (incorrectly labelled, see above) “Criegee channel” of b-pinene does not seem very plausible due to the associated ring strain - would not the H-shift from the alcohol carbon be the most likely one here also (as in the “hydroperoxide channel“)?