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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol composition and the contribution of SOA formation over Mediterranean forests" *by* Evelyn Freney et al.

Anonymous Referee #1

Received and published: 16 September 2017

The authors present airborne measurements of particles and gases above Mediterranean forests during the ChArMEx campaign. Offline TEM analysis is also presented. These measurements are used to investigate the sources of SOA in this forest. PMF and the Polyphemus model was also used to estimate the contribution of various sources to SOA. This manuscript employs a nice combination of measurements and modeling to investigate SOA. However, I find numerous major and technical corrections that need to be addressed prior to publication in ACP, as listed below:

Major Comments:

P2 L7: You have defined "isoprene epoxydiols SOA" as "(IEPOX)", which is incorrect. Isoprene epoxydiols are compounds typically found in the gas phase and can be called IEPOX for short. They can go on to form isoprene epoxydiols-derived SOA, which is

often called IEPOX-SOA.

P3 L15-18: I recommend that some of this information about what took place during this campaign be moved to the methods section, and replaced with a general description of what you investigated in this particular manuscript.

P3 L22: Is ATR an acronym? Please define if possible. Also, for readers who are unfamiliar with this aircraft, please provide more general information in this paragraph. E.g., what is the general size of the aircraft, how large is the payload, did it sample anything other than meteorological parameters and aerosols?

P4 L5: Provide a reference for the cToF-AMS instrument. Also, assuming it was the Aerodyne instrument, it should be referred to as the "Aerodyne compact time-of-flight aerosol mass spectrometer (cToF-AMS)" and called cToF-AMS instead of C-ToF-AMS to be consistent with previous literature.

P4 L6: I think you mean "spatial" instead of "temporal"? What was your spatial resolution, i.e., how far does the plane travel in 40 s?

P4 L26: Was it a high resolution (HR) or unit mass resolution (UMR) PTR-MS?

P6 L13: Section 3.1 belongs as part of your Methodology section 2 above. This section describes where and when the measurements were taken (along with some information about general conditions of the atmosphere), but doesn't provide any science results.

P6 L13: When you introduce the four RF's in this section (and then use them throughout the paper), please change it to use a uniform style. In this version, you have everything from, e.g., "RF20" to "RF20 03/07" to "RF 03/07 RF20" to "RF0307" (in Fig. S1) all referring to the same flight. I recommend providing the date of each flight in the methodology and then only use "RF20" in the results section.

P6 L20: Wherever you decide to specify the dates of the flights, I strongly recommend that you use a date format such as "3 July" instead of "03/07" throughout the manuscript and figures. 03/07 could mean 7 March or 3 July depending on where in the world the

reader is.

P7 L11: When you say ". . .and aromatics)", do you mean C8- and C9-aromatics? This sentence doesn't make sense as is.

Table 1: All of the VOC concentrations are in pptV, not ppbV, correct? Please check the units. Also what does the +- value represent? Range? Standard deviation?

P7 L27: ". . .was well oxidized" is a subjective statement that doesn't add scientific value or understanding, please change.

P7 L28: "with little evidence of fresh primary organic aerosol": Please provide or cite your evidence for this statement. The O:C value of the bulk OA by itself does not provide information about what types of OA (primary vs secondary) constitute the bulk OA.

P8 L15: With the evidence that you've shown, I'm not really convinced that you can conclude that you're seeing organosulphates. You state that you don't have any visual evidence that there were other compounds e.g. ammonium sulphate present, but can you show evidence that NH4 (or N) was NOT present? That's what I would want to see to really back up your conclusion of organosulphates, particularly because you've included this conclusion in your abstract.

Figure 3: Please explain in the figure caption what each panel is showing. Also, in 3a), are all four panels showing the same particle? It looks to me like the substrate in the top left panel is at a different angle than it is in the other three panels (the substrate overlaps with the bottom left corner in all panels except the top left panel).

P8 L30: I do not see sufficient evidence to say that it is "likely" that the fine mode particles were linked to new particle formation. You haven't shown measurements of new particle formation or even particles <20nm in size (and it seems those measurements were not taken during these flights), nor have you discussed possible primary sources of particles such as cities, vehicular traffic, etc. Please change this text so that

all of your conclusions are backed up with evidence, or change the strength of your conclusions to reflect the uncertainties.

P9 L14: This section 3.4.1 and also 3.4.2 are really part of aerosol chemical properties. I think they belong much better as subsections to 3.3 (aerosol chemical properties) rather than 3.4 (aerosol physical properties). Alternatively, they can be standalone sections.

P9 L26: Yes, isoprene oxidation is likely leading to some SOA formation, but what about other precursors? Is there any correlation with, e.g., monoterpenes? I would like to see a more thorough analysis here that considers all measured SOA precursor gases.

P9 L31: In Fig. 6a, m/z 80 is shown as an organic peak (green). It should be red (sulphate), correct?

P10 L3: You state here and in other places that you are plotting as a function of photochemical age, which seems incorrect. Photochemical age would have units of time (hours or days), but you're plotting against the ratio of isoprene oxidation products to isoprene. It should be possible to convert this ratio to a unit of time, depending on whether the rate constants and all of the relevant reactions involving these compounds are known enough to do so. If not, I suggest that you say instead that you plot as a function of relative age, or simply as a function of the ratio which represents the relative age of the airmass.

P10 L15: What is the value of f91 in your two spectra, and how does this compare with the other cited studies that investigated the contribution of monoterpenes or other pathways to m/z 91? The f91 in your spectra appear to be very small, suggesting these pathways aren't important.

Table 3: Please fix the formatting, m/z 71 in the first column should not be bold. Also, what does Pr mean? Are these Pearson r correlation values? Or R^2? Please clarify.

P10 L34: "...we can conclude that the observed OA can probably be related to a non-IEPOX isoprene SOA.": This conclusion as written is not backed up by the evidence you are presenting. First, a given compound's contribution to photochemical activity (by which I think you mean OH reactivity, which is different) has little to do with it's ability to form SOA. Sure, isoprene may be the main contributor to OH reactivity (among the measured compounds), but what matters for SOA formation is the SOA yield, and you haven't discussed that in this work. Second, you are presenting some evidence that suggests that IEPOX-SOA was not present in substantial amounts, but then your only conclusion in this section of the manuscript should be that IEPOX-SOA was not an important contributor. You have presented no measure of non-IEPOX isoprene SOA, so you have no basis on which to speculate about the magnitude of the non-IEPOX isoprene SOA at this site, whether it is dominant, negligible, or somewhere between. I suggest you clarify that you find that IEPOX-SOA appears negligible at this site, and that non-IEPOX isoprene SOA may play a role but it's unclear how much.

P11 L4: It will be very difficult for the editor, reviewers, and future readers to properly interpret this modeling work if the full model details are not yet published in this work or elsewhere! It would seem improper for this manuscript to be published before the Chrit et al. manuscript (in which you say the details can be found) at least appeared in ACPD.

P12 L11: This final paragraph should be removed, it is repeated information and doesn't add to the manuscript. In its place, you could consider strengthening this section by adding a paragraph to compare the model results with your measurements that were presented earlier in the paper. Are your measurements consistent with the model? What new information have we learned? What are the next steps, e.g., what other model or measurement results would we need to learn more?

Technical Corrections:

P3 L23: Please change "preformed" to "performed".

P3 L29: Please change "chaneling" to "channeling".

P4 L2: Please change to "scanning mobility particle sizer".

P4 L17: Please specify explicitly which standard temperature and pressure, to eliminate any possible confusion.

P4 L26: Update the Waked et al. citation if possible.

P4 L26: Add "(VOC)" after the word "compounds" to define this acronym.

P4 L27: Add references for the PTR-MS instrument.

P5 L16: I believe the proper name to use here in the "PMF Evaluation Tool (PET)".

P5 L20: SQUIRREL is an acronym and should be capitalized.

P7 L18: Please change from Fig. A6 to Fig. S6.

Table 2: Specify the units, as well as what the +- values represent.

P8 L15: Change from A7 to S7. There are other locations in the manuscript with similar A instead of S, please correct all instances.

P9 L5: Please define what isopreneC and monoterpeneC are.

P9 L11: Change "rations" to "ratios".

P10 L7: m/z should be italicized in all places in the paper.

P11 L6: Your supplemental figures are not referenced in order. Your Fig. S3 was just referenced here for the first time. I'm not sure if ACP has strict guidelines about this, but it's common to number them in the order they appear in the manuscript.

Many figures: The text and labels in a lot of the figures are too small to read. I suggest you make the font larger for clarity.

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