

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

### **Anonymous Referee #3**

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Authors present the results obtained during an airborne measurement campaigns over 2 Mediterranean forested areas (South of France). More precisely, they report results combining c-ToF-AMS, HS-PTR-MS, SP2, SMPS/OPC, Aerolaser (for formaldehyde measurements) as well as offline TEM analysis, from 4 flights (2 for each forest). Simulations performed with the Polyphemus model and comparisons with the measurements are also presented. While the paper is well written with a clear structuration, significant work is still needed to make it suitable for publication in ACP. Important details are missing and some major issues can also be found here and there throughout the manuscript (despite 21 co-authors).

Main comments:

1- There are major inconsistencies between VOCs concentrations reported in the text

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(p2 line 6, for instance), in figure 1 and in table 1. Not only a question of units. Must be checked very carefully and corrected.

2- Regarding PMF analysis, much more details are needed (at least in the SI)

3- It's not clear if the average values reported in the text and tables refers to the whole flight or only to the low altitude legs. This should be clarified and homogenized throughout the text. Considering the vertical profiles presented in figure S4, whole flight averages make no real sense. These vertical profiles and their implications are not discussed in the text except for boundary layer heights (p6, line 20).

4- Either the definition of externally mixed aerosol has recently changed either I don't understand the difference between fig 3a and b. This leads to important confusions in the discussion and conclusions.

Other comments

P1 line 30-31. Direct comparisons between AMS and TEM analysis are not relevant. AMS refers to a mass concentration without distinction between internally and externally mixed aerosol particles and TEM analysis refers, at best, to a number concentration. Authors state that at least 50 particles per gird have been analysis. What is the representativeness of the analyzed particles with respect to the total particles population? (SMPS/OPC measurement can help to assess this representativeness)

P 1 line 32. "Externally mixed organic aerosols, were equally identified with S signals, which may suggest the presence of organo-sulphates" I really don't understand this sentence. (cf main comment 4)

P2 line 6 : "high mixing ratio of isoprene (2-4 ppbV) and oxidation products (0.6 and 1.2 ppbV)" These concentration ranges cannot be found in table 1 and figure 1. Please check carefully.

P3 line 5 : I suppose that the authors refer to airborne measurements. There is a significant amount of ground based studies in the literature which already identified

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and quantified SOA in the Mediterranean region.

P3 line 30. Specify the lower cut off diameter of the aircraft aerosol inlet.

P4 line 2-4. Please specify the time resolution of both SMPS and OPC

P5 line 14-22 : The PMF analysis requires much more details and explanation (Q/Q<sub>exp</sub> vs number of factors, residuals, bootstrap/f<sub>peak</sub> etc..). This could be added in the SI. Also explain why adding inorganic ions (SO<sub>4</sub> and NO<sub>3</sub>) allows a better separation of the factors. Why NH<sub>4</sub> ions are not considered?

I also suppose that the relative contributions of MOOA and LOOA reported in section 3.4.1 refer to the OA+SO<sub>4</sub>+NO<sub>3</sub> mass concentration. Please clarify and discuss. Relative contributions to OA are from my point of view more relevant.

P5 line 24-31. Aerosol were apparently collected on TEM grids by means of a 2 stages impactor, but this information totally disappears in the discussion of the results. Is there any difference between the 2 particle size ranges? Also, what is the representativeness of the analyzed particles (at least 50 / grid) with respect to the whole particles population.

P6 section 3.1. Please add a map (with the flight tracks) in this section instead of in the SI. The authors state that one of the forest is a high isoprene emitter while the second is apparently more dominated by monoterpenes emissions. Considering the results presented in Tab1 and fig 1, isoprene and monoterpenes concentrations do not support this affirmation. Do you have any explanation?

P7 Section line 9-12. Even if the concentrations of aromatic VOC are low with respect to the total VOC concentrations measured here, they cannot be considered as negligible compared to isoprene and monoterpenes concentrations. They deserve more attention from the authors. For instance, what is B/T ratio. Does this ratio make sense with aged air masses? Are the concentrations of aromatics homogeneous all along the flight?

P7 line 13-20. I'm puzzled by the OH reactivity section. Either provide more details

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(Atkinson and Arey 2003 is a 40 pages review, and no reference are provided for Waked et al), either this section can be removed from the paper.

P7 line 23-24. I suppose that the uncertainties provided here (and throughout the whole manuscript/tables/figures) correspond to the standard deviation associated to the average values. This must be specified clearly as well as which part of the flights have been averaged.

P7 line 29-31 : see main comment 4

P7 line 31-32 : “The high contribution of externally mixed particles indicates that most of aerosol particles were recently emitted from their source” sounds contradictory with the sentence p7 line 27 “The organic aerosol measured during all flights was well oxidised, . . . , with little evidence of fresh primary organic aerosol.”

P8 line 3 : direct comparison between TEM analysis and AMS results are not relevant without a thoughtful analysis of representativeness of the particles analyzed by TEM with respect to the whole particles population.

P8 line 15. In order to support the assumption of the presence of organo-sulfates, did the authors check the ionic mass balance ( $\text{SO}_4$ ,  $\text{NO}_3$ ,  $\text{NH}_4$ ) from the AMS results? In other words, did they observe a deficit of  $\text{NH}_4$ ?

P8 line 30 : The authors should also mention and discuss the potential influence of industrial activities (Fos-Berre area) on the ultrafine mode observed during the easterly flights.

P9 line 6-7: ‘with lowest values corresponding to highest fractions of fine particle concentrations (Fig. 5). These observations are in agreement with previous field studies’ Considering fig 5 and the error bars, it’s impossible to conclude.

P9 line 24-26. I don’t understand why fig S8 is in the supporting information. From my point of view one of the main results from this study.

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P9 line 30 : “MOOA, contributing 55%” of OA or OA+SO<sub>4</sub>+NO<sub>3</sub>? What are the O:C ratio for MOOA and LOOA?

P10 line 16 : “This m/z 91 was present in all OA mass spectra and was significantly higher for the LOOA than for the MOOA.” Be more quantitative

P11 section 3.4.2. Not really convinced by the relevance of this section within the scope of this paper. Anyway. Why modelling results are only compared with measurements during the vertical profiles? How model and measurements compare during the low altitude legs?

P12 line 11-12. “The model estimates a significant contribution of isoprene SOA (approximately 15 to 35% to the total SOA). This cannot be confirmed by measurements due to the lack in a significant contribution of m/z 82 in the AMS spectra” This sentence makes no real sense. A very low contribution of m/z 82 means 1/ that there is a very little influence of isoprene SOA ; in that case the model is wrong or 2/ the isoprene SOA formation is formed through non-IEPOX pathways and in that case the question is how isoprene SOA is modelled?

Table 1 : please check the values and unit. Concentrations averaged during the whole flight or only low altitude legs? How the uncertainties are calculated (same for table 2)? Why C8 and C9 aromatics are not reported in the table?

Table 3 : What do you mean by “Pr”?

Figure 1 : Check values, add error bars, add % in the pie charts and add the flight codes (RF15 3006 etc..) in each panel in addition to a/, b/, .. (same for all figures)

Figure 5 : why no x error bars?

Figure 6 b : Why only 7 points represented here while ~100 are represented in fig S8?

Figure S6 : 11 RF represented, only 4 discuss in the manuscript. C8 aromatics are not only m-xylene.

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