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Response to Anonymous Referee #3 comments

The paper presents case studies of aerosol hygoscopicity and CCN-activity as recorded at the Finokalia measurement station in the presence of biomass burning influence from Greece or Croatia. While not revolutionary, the paper presents useful data on the hygroscopicity of ambient aerosol particles, and the analysis and discussion presented are generally reasonable. I therefore recommend publication in ACP after the following, mostly technical, comments have been addressed satisfactorily.

Response: *We thank the anonymous referee for the thoughtful review. Most of the issues raised were also concerns of the anonymous referee #2 therefore we have further elaborated on these points in the revision manuscript. Finally, in order to strengthen the aspect of the direct impact of biomass burning, we have included a section on calculations of potential droplet number in marine boundary layer clouds formed over Finokalia. The focus of the analysis is on the relative impact of BBOA CCN on CDNC, supersaturation and the contributions of aerosol number and hygroscopicity on the resulting CDNC.*

General / major comments:

1. The authors present the plume from the Balkans as a representative case of more aged biomass burning aerosol. This is certainly true, but it is also possible that the type of biomass burned at the Balkans can be different from the Greek plumes. It is well known that the properties of biomass burning aerosol from different types of fuels can differ significantly, so I would expect it to be difficult to separate the effect of ageing vs. the type of fuel being burned. The authors should comment on this. In general, the generalizability of the reported results to areas outside the eastern Mediterranean should be discussed in the manuscript.

Response*: We never said that the type of biomass burning at different locations is the same; indeed, a detailed analysis of the different kind of each fire event spectra can be found in the study of Bougiatioti et al. (2014) and the respective supplementary material. Nevertheless, in that study the organic aerosol derived mainly from the aging of the biomass burning aerosol (OOA-BB) had a similar profile, regardless of the BBOA it was derived from. What we wanted to point out was the effect that aging and atmospheric processing may have on the hygroscopicity. Nevertheless, as pointed out by the other anonymous referees as well, more details and information concerning the fire events will be given in the revised version of the manuscript.*

2. I am not convinced, at least based on the presented discussion, that the differences in condensation and coagulation are the primary factors explaining the different mixing states of the smaller and larger particles. Wouldn't it be a more plausible explanation that the smaller particles that make their way to Finokalia have originated primarily from secondary sources while the larger particles have a more important primary particle component? This smaller variability in the sources of the smaller as compared with larger particles could also potentially explain the observed smaller variability in the hygroscopicity distribution as well. In general, the discussion of the coagulation and the condensation is unquantitative and thus sloppy, for instance I suspect the authors in refer only to self-coagulation when they talk about "coagulation" in general in the text. It is well known that coagulation is more efficient for particles with different sizes, acting therefore primarily as a loss mechanism for the smaller particles, while being similar to condensation from the perspective of the large particles. This section of the discussion of the

results on p. 21555-21556 needs to be revised, along with the corresponding discussion in the conclusions section.

***Response**: The issue about the differences in condensation/coagulation explaining the differences in the mixing state of the different particle sizes is raised by all referees. We completely agree that the path of secondary formation of the smaller particles during the transport of the air masses is the most plausible explanation of the different mixing state. This will be included in the discussion, along with the estimate of the composition of the different particle sizes.*

Minor / technical comments:
3. When giving the kappa values throughout the manuscript (particularly in Tables 1- 3), please pay attention to the number of significant digits given in light of experimental variability and error. Is it really possible to constrain kappa within 0.001-0.01? If yes, please provide justification why you think so.

***Response**: Good point. The significant digits were derived from the statistical analysis of the data. All kappa values are now provided with 2 significant digits.*

4. The quality of the figures is in many cases insufficient and the fonts and linewidths chosen are too small. Please revise all figures keeping in mind the readability of the figures in typical ACP print versions.

***Response**: We would link to thank the reviewer for pointing out this issue. Depending on the amount of information on each figure, fonts and linewidths will be the largest possible to ensure good readability of the figures.*

5. P. 21541, line 12: Please quantify what you mean by “smaller” particles.

***Response**: Good point. Done. The text now reads “particle sizes smaller than 80 nm”.*

6. P. 21541, line 23: “BBOA” not defined before used. Please revise.

***Response**: Done.*

7. P. 21541, line 23: “enhancements” compared to what? Please clarify.

***Response**: Done. The text now reads “..with enhanced CCN concentrations than the ones before the arrival of the smoke plume, ranging between…”*

8. P. 21542, line 20: “impact” -> impacting

***Response**: Done.*

9. P. 21542, line 25: What do you mean by bb aerosol being “half of ammonium sulphate”? I presume you mean the hygroscopicity parameter, but please clarify.

***Response**: Done.*

10. P. 21550, line 1: “CALPSO” -> CALIPSO

***Response**: Done.*

11. P. 21550, line 8: “bellow” -> below

***Response**: Done.*

12. P. 21550, lines 24-26: The sum of the average concentrations of the individual components is quite a lot less than the average of the total aerosol. Could you elaborate on why? Perhaps using the medians instead of the arithmetic averages would be warranted?

***Response**: We would like to thank the reviewer for pointing out this inconsistency which arises from the daily averaging of the ACSM concentrations. Based on the suggestion, median values from all the data points are now 3.56, 1.31, 3.03 and 0.47 μg m^{-3} for sulfate, ammonium, organics and black carbon, respectively.*

13. P. 21553, line 25: “It appears, thus” -> It thus appears

***Response**: Done.*

14. P. 21555, line 14: “role … to” -> “impact …on the composition of the sampled…”

***Response**: Done*

15. P. 21555, line 15: I assume the sigma denotes arithmetic standard deviation. Please clarify.

***Response**: As noted in the text, σ(κ) is the chemical dispersion of the hygroscopicity parameter kappa, expressed indeed by the standard deviation of kappa around the most probable hygroscopicity κ*. This will be clarified in the text.*

16. P. 21559, line 17: Please add a reference to the “prior studies” mentioned.

***Response**: Good point. Done.*