

## Response to Anonymous Referee #1 comments

Overall, the manuscript presents a thorough analysis of four biomass burning events monitored in Finokalia station. Plumes are measured by their particle numbers, sizes, chemistry, CCN activity and hygroscopicity, making it possible to assess the climatic impacts of these biomass burning (BB) aerosols. However, the manuscript does not give any quantitative information about these BB events impact for climate, rather it satisfies with reassuring the previous observations on the organics aerosols impacts on aerosol hygroscopicity. As such, its originality is not very high, yet in Introduction the authors promise “The originality of this study relies on the fact that... very view studies focus on hygroscopicity of ambient biomass burning aerosol for a range of atmospheric aging, which is addressed here”. This indeed is very interesting and I find the manuscript well written and important, yet I would hope to see slightly deeper analysis of the background conditions for each of the events, to give the manuscript the originality it deserves. Below are my detailed suggestions which I hope the authors would address before manuscript publication in ACP.

*Response: We thank the reviewer for the well-articulated and thoughtful arguments. Our analysis was approached in view of these concerns to include more information about the fire events, which will help with the aspects of aging and atmospheric processing of the plumes. We have further elaborated on these points in the revision for clarity. 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. Below is our response to the comments raised in italics.*

General comments:

1) Aging of the BB plumes is mentioned many times, and terminology such as “aged” or “freshly-emitted” BB aerosol are used. Yet, it’s not very clear what are the criteria for more or less aged, or fresh, plumes? Also no information on transport conditions (how many hours air masses traveled, on what time of day and over which route) is given, nor any information on the type of fires (grass, forest, soil type). As current, it seems the age of each plume is rather deduced based on measured aerosol quantities, even if vice versa, the BB plume age should be predetermined. Could the authors clearly state how the age of each plume and BB aerosol was determined, and analyse the impact of this aging on each of the remaining measured quantities (as promised in introduction)?

*Response: All the information concerning the fire events are given in detail in the publication of Bougiatioti et al. (2014) (ACP). In this publication we wanted to focus on the hygroscopicity and the CCN during these events. Nevertheless, we will try including some more information on the issues raised by the anonymous referee in the revised version of the manuscript.*

2) In many occasions the manuscript analysis the “change” of observed aerosol quantities caused by BB emissions (e.g. 21551 lines 1-2 and 12-15 and 21-23; 21552 lines 1-2; 21555 lines 5-6; etc.). However, it’s not always very clear what is the reference point? Measured values prior to BB events, in the beginning of the events, or something else? Could this be more clearly stated? Also, is the reference point relevant for the current location, or representing e.g typical conditions at Finokalia? Do the air masses remain unchanged over the course of each of the events or may these play a role in observed changes?

**Response:** Good point. As a reference point, the averaged data from at least 6 hours of data prior to the arrival of the plume time are taken. This will be clarified in the revised text. Prior to the events, conditions are regarded as characteristic for a background site as Finokalia.

Minor comments:

p. 21552 lines 1-2: Authors state that CCN concentrations increase during the majority of the BB events. I find it very surprising if during a BB event, CCN concentrations are not increasing? Was the site affected by some other aerosol sources on those times when concentrations did not increase (compared to reference)?

**Response:** The increase of the CCN concentrations during the BB events was more pronounced, depending on the proximity of the fire and therefore, the travel time of the air masses. The only case when concentrations did not increase considerably was the event from Croatia, when air masses had a travel time of around 16h before reaching the station. Probably air masses were more diluted therefore the effect the biomass burning event had on CCN concentrations was less intense. This will be clarified in the revised text.

p. 21553 line 24-25; 21557 lines 1-2 and lines 18-19: Slightly confusing and mixed information is given on observed particle internal/external mixing states. Could the authors check that all this information is consistent?

**Response:** We would like to thank the reviewer for pointing out this inconsistency. Indeed, for the HTDMA the majority of the data exhibited unimodal distributions, apart from the data during the arrival of the smoke plumes. The confusing sentence that bimodal distributions were not taken into account for this specific study is referring to the comparison between kappa values derived from both the CFSTGC and the HTDMA, which will be clarified in the text. Also the statement that “all selected particle fractions were internally mixed” is modified according to the rest of the findings in the manuscript.

p. 21555 line 25: Authors say that aging of smaller particles takes longer than aging of larger particles. Maybe so, but in this case, how they rule out e.g. a possibility that these smallest particles were not just born later by a secondary route, but are actually from the same source?

**Response:** Good point. We do not rule out the possibility that the smallest particles were not formed later by a secondary route, it is a suggestion that the aging of smaller particles takes longer than aging of larger particles. Indeed, because of the difference in mass and interface with the gas phase, larger particles would age later than smaller ones. The text has been modified accordingly.

p. 21563 lines 8-9: Importance of coagulation vs. condensation could also be calculated by a dynamical model, having all this information the authors have. Would this support the statement that for 60 nm particles coagulation is dominant over condensation?

**Response:** We do agree that the discussion on the importance of coagulation vs. condensation needs strengthening and this is why the variance of the chemical composition of each particle size is now included in the revised version of the manuscript. According to Jacobson (2002) and numerical simulations, coagulation internally mixes a greater fraction of larger particles than smaller particles, and condensation increases the fractional coating of small particles more than it does large particles. The route of secondary formation of the smaller particles during the transport of the smoke is now also added in the text.

p. 21561 lines 1-2: Is kappa(BBOA) factor from ACSM seen to coincide with the occurrence of less-hygroscopic mode seen in HTDMA?

**Response:** *In general, the occurrence of two modes of different hygroscopicity seen by the HTDMA was not very frequent, but all of them occurred during the arrival of the smoke and indeed the derived  $\kappa_{\text{BBOA}}$  is very close to the  $\kappa$  of the less hygroscopic mode seen by the HTDMA.*

Typo: 2nd sentence of summary has repetition.

**Response:** *Done, the repeated sentence is deleted.*

*Jacobson, M.Z.: Analysis of aerosol interactions with numerical techniques for solving coagulation, nucleation, condensation, dissolution, and reversible chemistry among multiple size distributions, J. Geophys. Res.: Atmospheres, 107, D19, 4366, doi:10.1029/2001JD002044, 2002.*