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Atmospheric Chemistry and Physics Discussions

ACPD

13, C11432–C11440, 2014

Interactive Comment

Interactive comment on "Processing of biomass burning aerosol in the Eastern Mediterranean during summertime" *by* A. Bougiatioti et al.

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The paper of Bougiatioti et al. describes the effect of wildfires on the chemical composition of aerosol at the remote background site of Crete. In the paper authors characterized five fire events in late summer 2012 when the fires originated from the Greek islands of Chios, Euboea, Andros, the Dalmatian coast and Sicily. Aerosol chemistry was measured by the Aerosol Chemical Speciation Monitor for organics, nitrate, sulfate, ammonium and chloride and with the aethalometer for black carbon. Organic aerosol was studied in more detail by positive matrix factorization analysis. The focus of the paper was in the atmospheric processing of biomass burning aerosol. That is a topical issue and therefore this paper discusses relevant subject. The paper is well

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written and clear, however, there are several important details missing in the paper that are listed in the comments later. I think this paper merits publication after addressing all the issues listed below.

Response: We would like to thank the reviewer for the careful reading and do the best to integrate his suggestions to improve the scientific content of our manuscript.

Page 25971, line 9; why was it mentioned that the aethalometer had seven wavelengths as the data from different wavelengths was not really discussed (only in supplements)

Response: We merely stated the characteristics of the instrumentation. In the revised version we have removed the “seven-wavelength” from the abstract. The source apportionment of black carbon based on the aethalometer absorption measurements supports our identification of the fire events.

Page 25974, lines 21-22; How was the CE determined for sulfate (0.25), was it RIE instead of CE?

Response: The issue of the use of a different collection efficiency for sulfate was also raised by Referee #1. We use the nominal 1.15 proposed by Ng et al. (2011) as the RIE of sulfate.The CE of 0.25 for sulfate was determined by comparison of the ACSM values with concurrent PM1 filter analysis using ion chromatography for a time-period of 4 months. The resulting ACSM values were one fourth of those determined by IC (ACSM sulfate=0.248 Filter sulfate with R2=0.984).

Page 25974, lines 24-25; How were absorption measurements converted to mass concentrations in aethalometer data?

Response: We converted the absorption measurements of the aethalometer to mass concentrations of BC using the approach described by Weingartner et al. (2003) and Sandradewi et al. (2008). This is based on the relationship between BC mass concentration and the corresponding absorption and attenuation coefficients at 370 and 880 nm. This information is included in the supplementary information.

Page 25975, lines 8-11; Why average OM:OC was used if the ratio was determined for each 30 min periods (Fig. 4)? Would that have changed Fig. SI-2.1?

Response: We explored this issue calculating POM by multiplying the OC concentrations derived from the PM1 filter analysis with the respective average OM/OC for the specific filter sampling period. This did not change Fig. SI-2.1 as far as the slope is concerned (y=0.907x instead of y=0.913x) but the correlation improved significantly (R2=0.703 instead of 0.578) (see attached Fig.1) The error bars represent the standard deviation of the 30-min ACSM values that correspond to the daily filter measurement. In the revised version of the manuscript we are including the revised calculation based on the time-dependent OM/OC values and the updated figure.

Page 25975, lines 11-15; Any details about the SMPS measurements are not provided. What was the size range that compared with the ACSM (Fig. 2b)?

Response: The size range of the used SMPS system was 9-850 nm. This information has been added to the revised manuscript. The ACSM also had a 1micron cut-off size.

Page 25976, lines 8-10; Did you take into account the contribution of sea-salt to sulfate concentrations? Could that cause a part of the difference between ACSM and filter sulfate as they had different cut-off sizes (ACSM lenses vs. filter PM1)?

Response: The contribution of sea-salt sulfate to the PM1 sulfate concentrations is very small. Based on Sciare et al. (2005) and using Na and Mg as seawater tracers we calculated an average contribution of nss-SO42- to the total sulfate of >99%. Both the ACSM and PM1 filter sampler have a cut-off of 1 micron but part of the difference between the ACSM and IC sulfate may be caused by the water content of the particles.

Page 25976, lines 19-20; Was there back-up filter for OC in order to subtract the gas phase contribution of OC?

Response: The PM1 filter sampler has been operating in the Finokalia station since the summer of 200.During FAME 2007 field campaign in the summer of 2007 we used

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a back-up filter for the OC in order to measure the positive artifact of the gas phase OC. In the approximately 40 samples analyzed, the OC concentrations of the back-up filter were on average 8.4±3.3% of the front filter. Based on these results that are consistent with the presence of highly aged OA in the site we have not used a back-up filter for OC. Note also that during a year a denuder was used in front of the filter and comparison between denuded and undenuded filters showed also no significant difference. Despite these previous results a fraction of the difference between the PM1 OC (and hence the POM) and the ACSM may be due to the gas phase contribution of the OC. This potential explanation is included in the revised version of the manuscript.

Page 25978, lines 14-16; Because the biomass burning cases were identified based on elevated BC concentrations, it would be good to present the contribution of biomass burning to BC concentration (Fig. SI-1.3.) in the manuscript, not in supplement.

Response: This is a good point. The BC source apportionment in the supplementary information reinforces the identification of the biomass burning events. Part of this material has been moved to the main part of the revised paper.

Page 25980, line 28; To me the diurnal profile of OOA-BB in Fig. 5c looks pretty flat

Response: We agree that the diurnal profile of OOA-BB is not very pronounced, nevertheless there is an increase in the OOA-BB concentrations between 5 and 10 am. The arrival of the biomass burning laden air masses is also the reason for the variability of the OOA-BB during this early morning period.

Page 25981, lines 9-13; I didn't really understand the end of the sentence; how do you know that fragment $C_3H_5O^+$ was pronounced in your mass spectrum at m/z 57 when you do not have high resolution mass spectrometer? Page 25983, lines14-5; Did OOA increase during fire events? If not (especially in Sicily fire), how can you say that BBOA was eventually transformed to OOA? OOA can have several different sources other than biomass burning.

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Response: There were clear OOA peaks during the time that the site was affected by the Sicily fire plume (Figure 5a. Obviously OOA can have several different sources other than biomass burning but its behavior does support our conclusion. Moreover, there is also an increase both in BC but also in the wood burning contribution of BC from the source apportionment.

Technical corrections

Page 25996, Fig 3; It is difficult to see the episodes when the data is presented as dots. I suggest to use lines instead of dots

Response: We chose to use dots instead of lines because when using lines, all lines fall on each other and it is difficult to differentiate between the different time series presented.

Page 25997, Fig 4c; BBOA (this study), OOA-BB (this study) and OOA (this study) are difficult to find, I suggest to change colors

Response: We use different colors for the 3 different factors in the revised version.

Page 25998, Fig 5a; see comment for Fig. 3

Response: We believe that it is easier to follow the different time series the way the data are currently presented. With the use of lines for both time series compared, the time series overlap and it is not clear that they coincide.

Page 25999, Fig 6; it would be nice if x-axis could be divided between good, moderate and poor correlation with vertical lines (see earlier comment for Page 25982, lines1-2)

Response: We have followed this suggestion in the revised manuscript.

Supplementary information

Fig. SI-1.3; use same y-axis for both the BC concentrations

Response: Done

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Fig. SI-4.1; this figure must be wrong. Both plots are the same and there is no seed or fpeak run shown.

Response: We would like to thank the reviewer for the careful reading and pointing out this problem. It has been corrected in the revised version of the manuscript.

Response: This is a valid point that can lead to misunderstandings. We have rephrased this sentence stating that the m/z 57 was pronounced in our mass spectrum and we also mention the possible origins of this fragment.

Page 25982, lines1-2; many readers are probably not familiar with theta angle. It would be good to define range of theta angles that represent good, moderate and poor correlations and add the ranges in all the figures with theta angle

Response: Based on Kostenidou et al. (2009) mass spectra that exhibit theta angles less than 15 degrees are similar to each other. Spectra with theta angles between 15 and 30 degrees indicate some similarity but also some important differences, while spectra with theta angles larger than 30 degrees do not compare well. We have included this information in the revised version of the manuscript and also included indicative vertical dashed lines in Fig.6.

Page 25982, lines18-20; ..."separate PMF analysis for each event"... It wasn't explained clearly in the text why you decided to do PMF separately for each fire case. Would the results in Table 1 be different if you had used the 3-factor solution shown in Fig. 5? I'm just a bit worried if you had enough data points in each fire event for a valid PMF analysis? Any comment on that?

Response: One of the novel aspects of this study is the analysis of individual fire plumes from different areas (therefore potentially different fuel compositions) and different ages. We decided to perform a separate PMF analysis for each event to avoid the a-priori constraint that all BBOA spectra were the same. This way we were able to obtain different BBOA spectra and quantify their similarities and differences (summa-

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rized in Table 1). The data points for each PMF analysis were 74, 152, 62, 80 and 74 respectively. As far as Table 1 is concerned the averaged values using the respective periods from the 3-factor solution shown in Fig.5 is shown in the attached Table, which, when compared to Table 1 does not show any significant difference.

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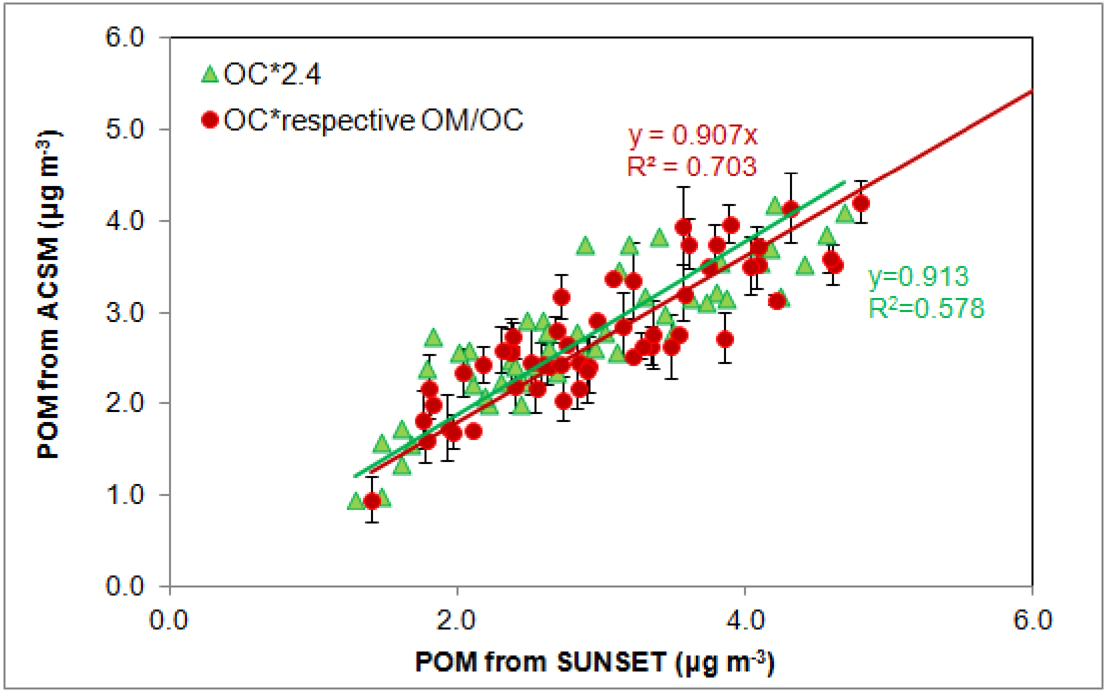
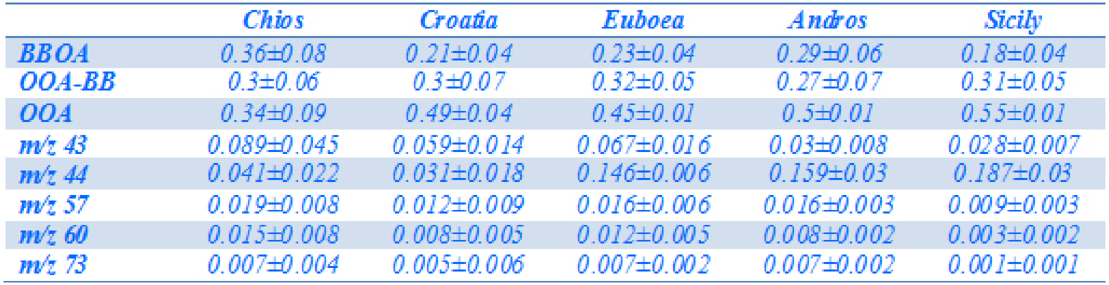

6.0
5.0
4.0
3.0
2.0
1.0
0.0
0.0
2.0
4.0
6.0
POM from ACSM (µg m-3)
POM from SUNSET (µg m-3)
▲OC*2.4
●OC*respective OM/OC
y = 0.907x
R² = 0.703
y=0.913
R²=0.578

Fig. 1.



	Chios	Croatia	Euboea	Andros	Sicily
BBOA	0.36±0.08	0.21±0.04	0.23±0.04	0.29±0.06	0.18±0.04
OOA-BB	0.3±0.06	0.3±0.07	0.32±0.05	0.27±0.07	0.31±0.05
OOA	0.34±0.09	0.49±0.04	0.45±0.01	0.5±0.01	0.55±0.01
m/z 43	0.089±0.045	0.059±0.014	0.067±0.016	0.03±0.008	0.028±0.007
m/z 44	0.041±0.022	0.031±0.018	0.146±0.006	0.159±0.03	0.187±0.03
m/z 57	0.019±0.008	0.012±0.009	0.016±0.006	0.016±0.003	0.009±0.003
m/z 60	0.015±0.008	0.008±0.005	0.012±0.005	0.008±0.002	0.003±0.002
m/z 73	0.007±0.004	0.005±0.006	0.007±0.002	0.007±0.002	0.001±0.001

Fig. 2.

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