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# Processing of biomass burning aerosol in the Eastern Mediterranean during summertime

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## Abstract

The aerosol chemical composition in air masses affected by wildfires from the Greek islands of Chios, Euboea and Andros, the Dalmatian Coast and Sicily, during late summer of 2012 was characterized at the remote background site of Finokalia, Crete. Air masses were transported several hundreds of kilometers, arriving at the measurement station after approximately half a day of transport, mostly during night-time. The chemical composition of the particulate matter was studied by different high temporal resolution instruments, including an Aerosol Chemical Speciation Monitor (ACSM) and a seven-wavelength aethalometer. Despite the large distance from emission and long atmospheric processing, a clear biomass burning organic aerosol (BBOA) profile containing characteristic markers is derived from BC measurements and Positive Matrix Factorization (PMF) analysis of the ACSM mass spectra. The ratio of fresh to aged BBOA decreases with increasing atmospheric processing time and BBOA components appear to be converted to oxygenated organic aerosol (OOA). Given that the smoke was mainly transported overnight, it appears that the processing can take place in the dark. These results show that a significant fraction of the BBOA loses its characteristic AMS signature and is transformed to OOA in less than a day. This implies that biomass burning can contribute almost half of the organic aerosol mass in the area during summertime.

## 1 Introduction

Organic species represent a significant and often major mass fraction of submicron aerosol. At continental midlatitudes, organic material contributes ~ 20–50 % of the total fine particulate mass while in tropical forested areas this contribution can be as high as 90 % (Kanakidou et al., 2000; Zhang et al., 2007). During the last few years, factor analysis of measured organic mass spectra from aerosol mass spectrometry (AMS) data has enabled the deconvolution of organic aerosol (OA) to different components

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based on composition (Zhang et al., 2011). This has provided valuable insights on the source and transformation processes of OA in the atmosphere (Lanz et al., 2008, 2010; Hildebrandt et al., 2010, 2011; Cubison et al., 2011; Aiken et al., 2011; He et al., 2011; Ulbrich et al., 2009). Different components exhibit distinct mass spectra, with the two most common ones being the hydrocarbon-like OA (HOA) and oxygenated OA (OOA) (Zhang et al., 2007). In some cases the OOA component is further resolved into two subtypes that have different degrees of volatility, namely the low-volatility OOA (LV-OOA) and semi-volatile OOA (SV-OOA) (Jimenez et al., 2009; Ng et al., 2010). Other components are sometimes identified, e.g. biomass burning OA (BBOA) (Aiken et al., 2009; Cotrell et al., 2008; He et al., 2010; Huang et al., 2011) and cooking OA (COA) (He et al., 2010; Mohr et al., 2009; Allan et al., 2010). The differentiation of the factors is based on the mass spectra of the components, mainly in terms of two mass fragments ( $\text{CO}_2^+$  at  $m/z$  44 and  $\text{C}_2\text{H}_3\text{O}^+$  at  $m/z$  43) (Ng et al., 2010) and on other characteristic fragments.

Key tracers of BBOA in AMS spectra are the enhanced signals at  $m/z$  60 and 73 from the ions  $\text{C}_2\text{H}_4\text{O}_2^+$  and  $\text{C}_3\text{H}_5\text{O}_2^+$  (Schneider et al., 2006; Alfarra et al., 2007). The enhanced signal at  $m/z$  60 is associated with levoglucosan and other similar species (e.g. mannosan and galactosan). Nevertheless, the sum of levoglucosan, mannosan and galactosan alone can account only for around 1/10 or less of the total signal of  $m/z$  60 in BBOA. This indicates that the majority of it originates from different molecules with the same fragmentation pattern as levoglucosan in the AMS (Aiken et al., 2009; Lee et al., 2010b). Given that levoglucosan is a monomer that results from the thermochemical decomposition of cellulose at elevated temperatures in the absence of oxygen, such species may include dimers and trimers of similar molecules (Cubison et al., 2011). Laboratory-based studies (Hennigan et al., 2010; Hoffmann et al., 2010; Kessler et al., 2010) have estimated an atmospheric oxidation lifetime for levoglucosan in typical conditions that ranges from 15 h to a few days. Nevertheless, the extent of degradation of the AMS tracer signals  $f_{60}$  and  $f_{73}$  after emission is not well known.  $f_{60}$  has been reported to decrease during photochemical processing of BBOA and from addition of

secondary organic aerosol (Huffman et al., 2009). It is unclear whether commonly-used tracers persist long enough in aged, atmospheric processed BBOA, to serve as effective markers;  $f_{60}$  is currently thought to be a robust tracer for BB for aging timescales of at least one day (Cubison et al., 2011).

The Eastern Mediterranean is considered an area of importance for atmospheric aerosol research where aerosols play a significant role in radiative cooling up to five times greater than the warming induced by greenhouse gases (Vrekoussis et al., 2005). During summertime, when enhanced anthropogenic aerosol inflow is combined with limited precipitation and persistent high relative humidity, the total light scattering may be twice of that during wintertime (Kalivitis et al., 2011). The organic fraction during summer accounts for one third of the dry submicron aerosol mass, with organics being highly oxidized (Hildebrandt et al., 2010a). Based on long-term measurements of carbonaceous aerosols in the area, Sciare et al. (2008) demonstrated that every year during two periods (March–April and July–September) there is a high biomass burning contribution to the aerosol from long-range transport from Southern Europe and European countries surrounding the Black Sea.

This study presents high temporal resolution measurements in a remote background site in the Eastern Mediterranean during late summer (August–September 2012). With the concurrent use of an Aerosol Chemical Speciation Monitor (ACSM, Aerodyne Inc.) and a 7-wavelength aethalometer we identified and studied different biomass burning-influenced air masses that arrived in the area. The measurements are analyzed to gain insights into the atmospheric processing of BBOA.

## 2 Experimental methods

### 2.1 Sampling site and period

Measurements took place at the Finokalia station (35°32' N, 25°67' E; <http://finokalia.chemistry.uoc.gr>) of the University of Crete, which is part of the

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During the whole measurement period daily PM<sub>1</sub> filter samples were collected on pre-combusted quartz fiber filters. Samples were analyzed for organic and elemental carbon using an OC/EC analyzer (SUNSET Laboratory Inc.) as well as for water-soluble ions after extraction in ultrasonic bath with nanopure water. The solutions obtained were analyzed by ion chromatography (IC) for anions (Cl<sup>-</sup>, Br<sup>-</sup>, NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, C<sub>2</sub>O<sub>4</sub><sup>2-</sup>) and cations (K<sup>+</sup>, Na<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>) using the procedure described by Bardouki et al. (2003). Results for organic carbon (OC), sulfate, ammonium and nitrate were compared to ACSM-derived concentrations. As the ACSM provides organic matter (OM) concentrations, the filter-derived OM was calculated by multiplying OC concentrations with a conversion factor (in our case 2.4; Sect. 3.3), which is the ratio of the average organic molecular mass to the carbon mass for the organic aerosol. Particle number distribution was monitored using a scanning mobility particle sizer (SMPS) in 5 min time resolution, O<sub>3</sub> concentrations and meteorological parameters (wind speed and direction, pressure, temperature, relative humidity) were also continuously monitored.

### 2.3 PMF analysis

The measured mass spectra from the ACSM were deconvolved into different components using the positive matrix factorization (PMF) analysis exploiting the Multilinear solver (ME-2) (Paatero and Tapper, 1994; Paatero, 1999). The interface for initiating and controlling the multilinear engine algorithm (ME-2, P. Paatero, University of Helsinki), as well as analyzing the results, was developed at the Paul Scherrer Institute (Canonaco et al., 2013) within the software package Igor Pro Version 6.22A (Wavemetrics Inc., Lake Oswego, OR, USA). The interface is intended to facilitate the analysis of the source apportionment of measured data (Ng et al., 2011b). Briefly, PMF uses a least squares algorithm to minimize the squared difference between modeled and the measured data, i.e. the residual weighted by the uncertainty. Here, the measurement error of the ACSM represents the uncertainty. The input organics and organics error

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matrices for the analysis are derived automatically from the ACSM data analysis software, using a simple automated procedure (Ulbrich et al., 2009). For ME-2 and PMF the entries in the model solution are fit using a least squares algorithm that minimizes iteratively the model uncertainty. For the ACSM data analysis the signals at  $m/z$  18 and 44 are equal in the fragmentation table (Allan et al., 2003, 2004).

### 3 Results and discussion

#### 3.1 PM<sub>1</sub> Composition

ACSM sulfate and ammonium concentrations were on average 19 and 15% lower than the corresponding PM<sub>1</sub> filter measurements ( $R^2 = 0.8$  and  $0.68$  respectively) (Fig. SI-2). These discrepancies can be attributed to errors and fluctuations in the collection efficiency of these species. Using the parameterization proposed by Middlebrook et al. (2012) for the effect of high aerosol acidity the  $\text{NH}_4/\text{NH}_{4,\text{neutr}}$  is equal to  $0.803$  ( $R^2 = 0.67$ ), where  $\text{NH}_{4,\text{neutr}}$  represents the theoretical ammonium concentration needed to neutralize the inorganic anion mass concentrations. Similar acidic behavior of the aerosol has been observed for the site in previous measurements (Hildebrandt et al., 2010). Organic matter concentrations calculated from OC analysis ( $\text{POM} = 2.4\text{OC}$ , as derived from our dataset, using the estimate of Aiken et al. (2008), Sect. 3.3) and organic concentrations from the ACSM are in good agreement with an average slope of  $0.91$  and  $R^2 = 0.57$ . Slightly higher OC concentrations from filter analysis may be due to the adsorption of gaseous organic compounds on the quartz-fiber filters. The time series of the high temporal resolution measurements is shown in Fig. 2a.

We compared the PM<sub>1</sub> mass concentrations by adding the ACSM and BC masses with the mass estimated by the SMPS volume, using an average organic aerosol density equal to  $1.35\text{ g cm}^{-3}$  as determined by Lee et al. (2010a) for the same site during late spring 2008 (Fig. 2b). The ACSM concentrations for ammonium and sulfate are corrected based on the filter concentrations. The mass derived from gravimetric mea-



measurements of the concurrent daily PM<sub>1</sub> filters is also given. The ACSM + BC mass concentrations have an average value of  $9.2 \pm 4.8 \mu\text{g m}^{-3}$  which is in good agreement with the  $10.9 \pm 4.2 \mu\text{g m}^{-3}$  calculated from the SMPS. The difference may be attributed to components that cannot be measured by the ACSM.

The ACSM measurements were then divided to those corresponding to fire events and the rest, based on black carbon (BC) measurements from the aethalometer, the HYSPLIT analysis and available satellite pictures. Figure 2c and d represent the relative contribution of each constituent to the PM<sub>1</sub> mass (sum of the derived constituents) during non-fire and fire events, respectively. During fire events, the contribution of organics and BC increase substantially at the expense of sulfate.

### 3.2 Identification of biomass burning events

Biomass burning events were identified by an enhanced BC concentration (Fig. 3a). We assume that  $250 \text{ ng m}^{-3}$  corresponds to the background concentration for Finokalia during periods where the site is not affected by biomass burning. This limit is consistent with the  $213 \pm 34 \text{ ng m}^{-3}$  found by Cristofanelli et al. (2013) in Mt. Cimone, Italy and comparable to average background levels reported for European continental background sites (Van Dingenen et al., 2005; Putaud et al., 2004). The average BC concentration during the measurement period was of  $387 \pm 149 \text{ ng m}^{-3}$ , with lower values being close the background value ( $250 \text{ ng m}^{-3}$ ). Possible biomass burning incidents are identified by having BC concentrations at least 2.5 times higher than the Finokalia background (higher than  $625 \text{ ng m}^{-3}$ ). This is approximately equivalent to two standard deviations above the average background value. As a second step, locations of possible fire events were identified by the Fire Information for Resource Management System (FIRMS). The exact location and plume age was calculated by the HYSPLIT backtrajectory analysis.

Five distinct wildfire events affecting the site were identified using the approach described above during the sampling period: a large fire that burned most of the island of Chios (19–21 August); the extensive wildfire at the Dalmatian Coast in Croatia and

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southern Bosnia-Herzegovina which resulted in smoke plumes which spread all across the Balkans during 28–30 August; the smaller fires on the islands of Euboea on 5 September and Andros on 13 September (backtrajectories and MODIS images for each can be found in the Supplement); and a fire that took place in the island of Sicily during 18–21 September and influenced the site on 21 September.

The above periods were characterized by strong biomass burning ACSM tracers at  $m/z$  60 and 73 (Fig. 3). The signal was considerably weaker for the Sicily fire emissions that travelled the longest before reaching Finokalia. BC concentrations were also relatively high towards the end of September, with an average concentration of  $535 \pm 109 \text{ ng m}^{-3}$ , which is close to 2.5 higher than the background level for Finokalia. The difference is that during this period BC concentrations build up gradually and the ACSM tracers do not reveal any noticeable variability. In addition, FIRMS does not identify any significant events in the area during 25–30 September, therefore this is not regarded as a fire event. Using the approach of Sandradewi et al. (2008) we also calculated the contribution of the biomass burning to the total BC concentrations (see Supplement).

### 3.3 Elemental composition of organic aerosol

The ACSM dataset was used to calculate the elemental composition and ratio of organic mass to organic carbon (OM/OC) based on the estimates proposed by Aiken et al. (2008) and Ng et al. (2011c), as follows:

$$\text{O/C} = (3.82 \pm 0.005)f_{44} + (0.0794 \pm 0.007) \quad (1)$$

$$\text{OM/OC} = (1.26 \pm 0.02)\text{O/C} + (1.18 \pm 0.001) \quad (2)$$

$$\text{H/C} = (1.01 \pm 0.08) + (6.07 \pm 1.39)f_{43} + (-16.01 \pm 5.34)\frac{f_{43}^2}{25978} \quad (3)$$

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where  $f_{43}$  and  $f_{44}$  are calculated as  $f_{43} = [m/z 43] (\mu\text{g m}^{-3}) / C_{\text{OA}}$  and  $f_{44} = [m/z 44] (\mu\text{g m}^{-3}) / C_{\text{OA}}$  with  $C_{\text{OA}}$  being the total OA mass concentration in  $\mu\text{g m}^{-3}$ . Figure 4 shows the variation of the atomic ratios of O/C and H/C and the mass ratio of OM/OC of the OA during the measurement period, averaging every 2 h, as well as  $f_{43}$  vs.  $f_{44}$  compared to other studies. According to Herringa et al. (2011) the presence of wood-burning POA and its aging SOA are found at the left edge of the triangle. The O/C ratio varies between 0.52 and 1.34, with a mean value of  $0.97 \pm 0.13$ , values which are within the observed values of low-volatility and semi-volatile OOA (0.3–1.1; Ng et al., 2011c) and dicarboxylic acids (0.44–2.00; Kuwata et al., 2012). For the same region, Hildebrand et al. (2010a) estimated an O/C of 0.8 for a sunny May period, corresponding to an average  $f_{44}$  of 18.2%. The higher O/C value found in this study can be attributed to the higher contribution of  $f_{44}$  ( $23.3 \pm 3.7\%$ ). The H/C ratio varies in a range of 1.12–1.34, with a mean value of  $1.23 \pm 0.03$ . The O/C ratio is a proxy for the oxidation state and the photochemical age of organic aerosols (Ng et al., 2011c; Huang et al., 2011) and thus its variability during the measurement period reflects variations in OA age at Finokalia. Combining information from both ratios, for most of the cases, lower O/C coincides with higher H/C and all fire events can be clearly identified in the time series. During the identified fire events both O/C and H/C have average values of  $0.77 \pm 0.09$  and  $1.27 \pm 0.02$ , respectively. These values are consistent with the presence of less oxidized aerosol than the study average. There are lower-than-average O/C ratios around the 21st of September, when biomass-burning-influenced air masses from Sicily arrived at the station after a travel time of more than 30 h.

The average OM/OC was  $2.4 \pm 0.14$  which is characteristic of a remote background site such as Finokalia and which is comparable to the value of 2.2 found by Hildebrandt et al. (2010b) and the value of 2.1 used by Sciare et al. (2005) to convert OC to POM for mass reconstruction during the MINOS summer campaign. These very high values of OM/OC reflect the highly oxidized nature of organic Eastern Mediterranean aerosol, which during the middle of the summer is in fact even more oxidized than found by Hildebrandt et al. (2010a) for the same site during late spring and early summer.



arrival times of smoke-influenced air masses during early morning hours. The identified factors were also examined for their mass spectra signatures, their correlation with reference mass spectra and also with other characteristic time series which justify their behavior.

5 The BBOA component had the characteristic markings of biomass burning aerosol, which are enhanced signals at  $m/z$  60 and 73 probably from levoglucosan and related compounds (Schneider et al., 2006; Cottrell et al., 2008). It also had prominent peaks at  $m/z$  27, 29, 41, 43, 55, 57, 77 and 91 and significant contributions at high  $m/z$ 's which are indicative of freshly-emitted organic aerosol. Apart from levoglucosan fragments,  $m/z$  57 can also be a good marker for fresh aerosol, as it is a typical fragment of saturated hydrocarbon compounds or of long alkyl chains ( $C_4H_9^+$ ) (Alfarra et al., 2004; Capes et al., 2008) or can also originate from levoglucosan (most probably  $C_3H_5O^+$ ), fragment which is pronounced in our spectrum. The fresh BBOA factor in our study had an O/C of 0.19 and an H/C of 1.35 consistent with other observed BBOA (Fig. 4c). 10 The BBOA factor time series showed good correlation with both BC ( $R^2 = 0.62$ ) as well as with nitrate ( $R^2 = 0.75$ ) and no correlation with sulfate or ammonium.

The OOA-BB mass spectrum displayed characteristics of oxidized OA with enhanced signals at  $m/z$  18, 29 and 44. It also had a small signal at  $m/z$  43, implying its potential origin from the fresh BBOA. Its O/C was 0.96 and its H/C 1.3, indicating its processed nature. OOA-BB correlated with nitrate ( $R^2 = 0.63$ ) and BC ( $R^2 = 0.48$ ) and displayed lower correlation with sulfate and ammonium ( $R^2 = 0.29$  and 0.38 respectively). OOA-BB peaked during the fire periods and exhibited similar diurnal variability with the BBOA factor (Fig. 5) supporting its classification as processed biomass-burning OA. 20

The OOA component had the characteristics of oxidized organic aerosol, like OOA-BB, but the relative intensity of  $m/z$  18 and 44 is even more pronounced, indicating more oxidized aerosol, which is also supported by its elevated O/C (1.23). The intensity of other fragments in its spectrum is low. This factor correlates well with both sulfate and ammonium ( $R^2 = 0.59$  and 0.62, respectively). 25

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Figure 6 shows the similarity (expressed as the angle  $\theta$  between the spectra; Kostenidou et al., 2009) between the identified factors in this study and other relative organic aerosol spectra found in the AMS mass spectral database (<http://cires.colorado.edu/jimenez-group/AMSsd/>). Our BBOA spectrum correlates well with the average BBOA of Ng et al. (2011), obtained by averaging together BBOA mass spectra from two sites (Mexico City and Houston data sets) and has some similarity with the spectra of OA from laboratory burns of pine, oak and spruce (Schneider et al., 2006). The derived BBOA had comparable features with ambient BBOA spectra from Mexico (Aiken et al., 2009), Po Valley (Saarikoski et al., 2012) and Barcelona (Mohr et al., 2012) ( $\theta$  between 25 and 30°). The OOA-BB factor correlates well with organic aerosol from the Po Valley and Barcelona, as well as with less oxidized OOA reported for the same sampling site (Finokalia) in past studies during spring and fall (Hildebrandt et al., 2010a, 2011). It does not correlate well with OOA associated with wood burning in fireplaces in Paris during the winter (Crippa et al., 2013) nor with semi-volatile OA from Pasadena (Hersey et al., 2011). Finally, OOA correlates well with the more oxidized OOA found for the same site in previous campaigns and resembles to the OOA found in Barcelona, Zurich during winter and OOA Po Valley.

The contribution of each factor to the OA and the characteristic fragments to the OA mass spectrum of the different fire events is summarized in Table 1. These results are based on separate PMF analysis for each event by selecting the corresponding 3-factor solution. OOA-BB and OOA mass spectra during each event correlated well with the respective OOA-BB and OOA spectra for the whole measurement period (shown in Fig. 5b) but also with the corresponding spectra of the rest of the events ( $\theta < 15$  and 9 degrees respectively).  $f_{60}$  and  $f_{73}$  diminish and  $f_{44}$  increases as the transport time of the plume increases. According to Cubison et al. (2011),  $f_{60}$  has consistent background level values around 0.3% in OA with negligible BB influence, with plumes exhibiting a trend toward lower  $f_{60}$  with time. This is also the case in our study where the Sicily and Croatia fire events exhibit the lowest values, close to these background levels. We estimated an e-folding time of 12 h for  $m/z$  73 and 28 h for  $m/z$  60 for these fire events.



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September 2012. During the measurement period  $PM_1$  concentrations ranged from 3.2 to  $34.2 \mu\text{g m}^{-3}$  with an average value of  $12.2 \pm 4.7 \mu\text{g m}^{-3}$  with organics and sulfate being the most abundant species, each accounting for  $\sim 40\%$  of the  $PM_1$ . Five discrete biomass burning events were identified from characteristic tracers in the organics mass spectrum as well as in the BC time series, and origin of the air masses was verified by backward trajectory analysis. For all the cases the smoke was transported for several hundreds of kilometers. In three events the smoke arrived at the site early in the morning, after around 8 h of night-time atmospheric processing. One additional fire event involved air masses with an approximate travel time of 30 h, coming from Sicily. During the fire events the contribution of organics to the total mass increased to almost 50 %, with BC also showing an increase in contribution to almost 10 % of the  $PM_1$ .

By performing PMF analysis three factors were derived: one having characteristics of biomass burning, one with oxygenated BBOA, and one with highly oxygenated organic aerosol. The BBOA component showed reasonable correlation with reference average BBOA mass spectra while more oxidized OOA correlated well with reference OOA mass spectra. BBOA correlated well with BC and nitrate, processed BBOA (OOA-BB) also had high correlation with nitrate and more oxidized OOA correlated well with sulfate and ammonium. More than 80 % of the total organic mass is accounted for by the two oxygenated OOA factors. The OA as a result was highly oxygenated with an overall O/C ratio that had a mean value of  $0.97 \pm 0.13$ .

Even within 8–9 h of travel, and mostly during nighttime, most of the freshly-emitted BBOA had been processed and transformed to more oxidized OOA, with processed-BBOA accounting for 30 % on average of the total organic mass. This suggests that the biomass-burning contribution to OA can be misidentified as OOA contribution, therefore underestimating the importance of BBOA.

The biomass burning-influenced air masses arriving from Sicily after 30 h of travel, lacked any characteristic BBOA marker. This is an indication that these markers have a finite lifetime, especially during summertime Mediterranean conditions, which combine



high temperatures and high insolation periods.  $f_{60}$  seems to be a robust BB-tracer only for timescales up to one day from emission.

**Supplementary material related to this article is available online at**  
[http://www.atmos-chem-phys-discuss.net/13/25969/2013/  
acpd-13-25969-2013-supplement.pdf](http://www.atmos-chem-phys-discuss.net/13/25969/2013/acpd-13-25969-2013-supplement.pdf).

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**Table 1.** Relative contribution of each one of the three identified PMF factors to the OA and relative intensity of characteristic markers in each fire event.

	Chios	Croatia	Euboea	Andros	Sicily
BBOA	0.4	0.16	0.29	0.29	0.18
OOA <sub>1</sub> -BB	0.32	0.28	0.37	0.27	0.25
OOA <sub>2</sub>	0.28	0.46	0.34	0.44	0.57
$f_{43}$	0.08	0.069	0.06	0.034	0.028
$f_{44}$	0.05	0.037	0.142	0.122	0.193
$f_{57}$	0.021	0.017	0.018	0.02	0.014
$f_{60}$	0.011	0.006	0.01	0.008	0.004
$f_{73}$	0.009	0.004	0.008	0.006	0.001
Transport time	7 h	16 h	9 h	8 h	33 h

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**Fig. 1.** (a) Occurrence of wildfires as spotted by the Fire Information for Resource Management System (FIRMS) (<https://firms.modaps.eosdis.nasa.gov/firemap/>) during the whole measurement period (1 August–30 September) with specific locations highlighted in red and (b) MODIS image of Chios fire (18 August 2012, [http://eoimages.gsfc.nasa.gov/images/imagerecords/78000/78918/chios\\_amo\\_2012231\\_lrg.jpg](http://eoimages.gsfc.nasa.gov/images/imagerecords/78000/78918/chios_amo_2012231_lrg.jpg)).

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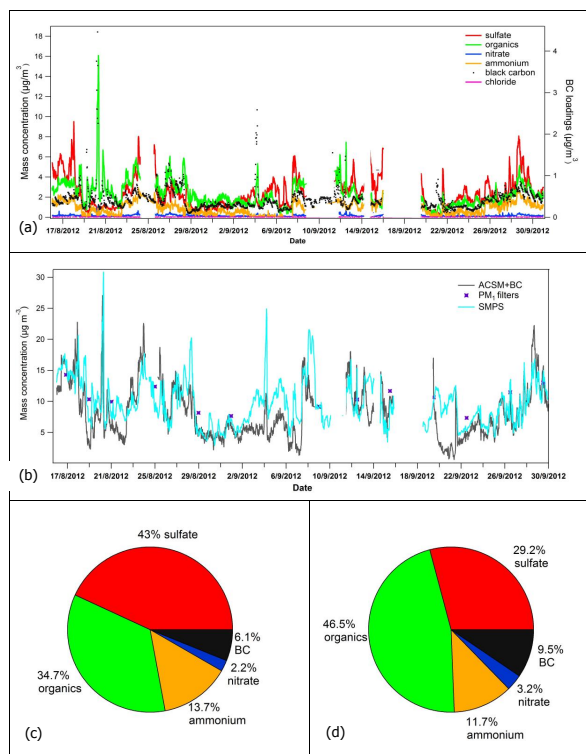
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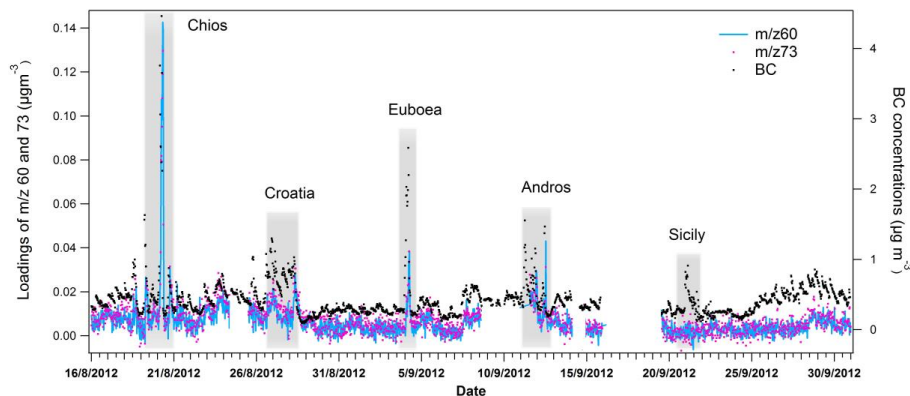
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**Fig. 2.** (a) Time series of concentrations of major PM<sub>1</sub> species; (b) comparison of ACSM-derived, SMPS-derived and filter-derived mass time series; PM<sub>1</sub> composition during (c) non-fire periods and (d) and fire events.

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**Fig. 3.** The time series of the biomass tracer loadings of  $m/z$  60 and  $m/z$  73 and black carbon. The shaded areas represent the identified fire events and locations.

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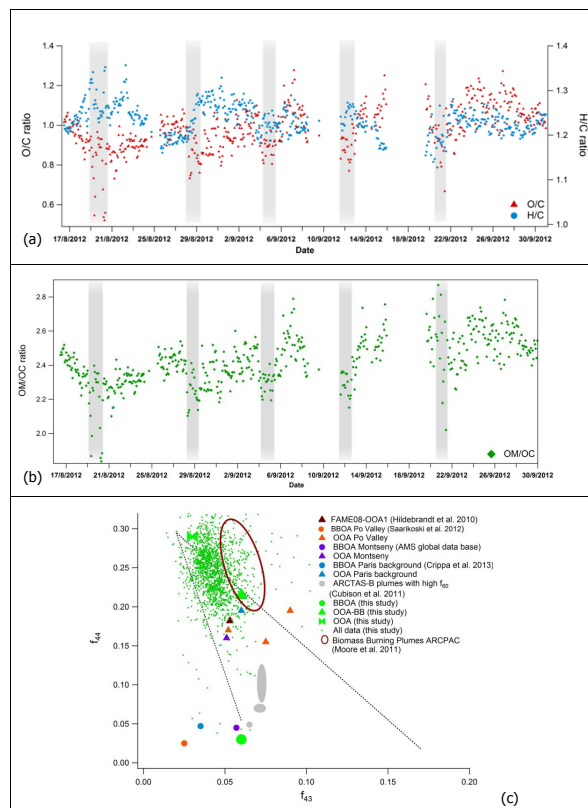
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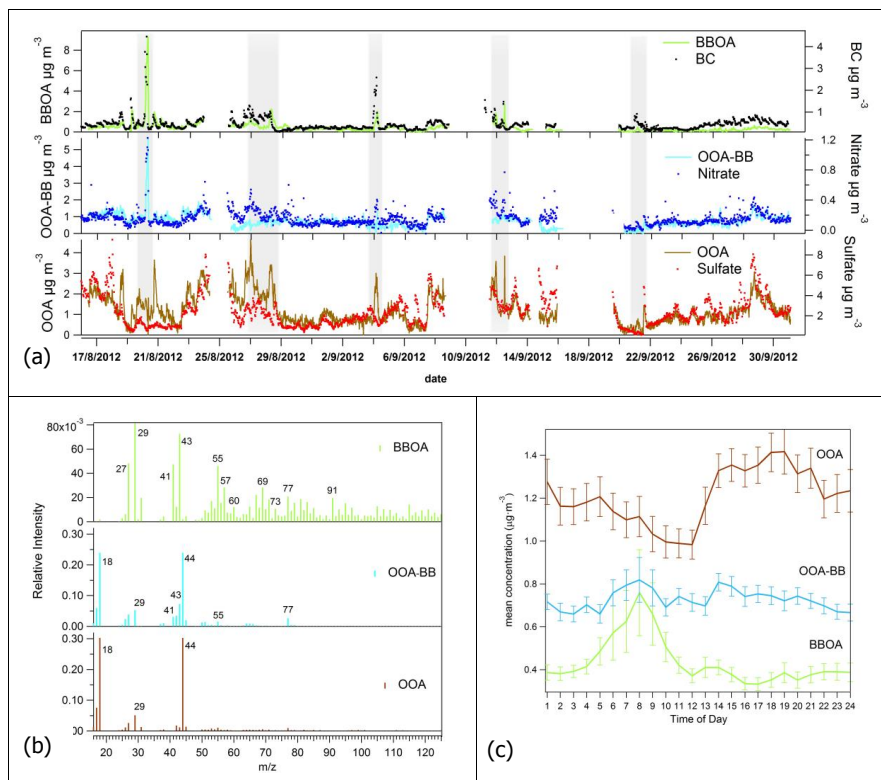
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**Fig. 4.** Time series of atomic ratios of O/C and H/C (a), the variation of the mass ratio of OM/OC (b), and the fractional signals  $f_{43}$  and  $f_{44}$  of biomass burning and oxygenated organic aerosol in our study and other studies (c).

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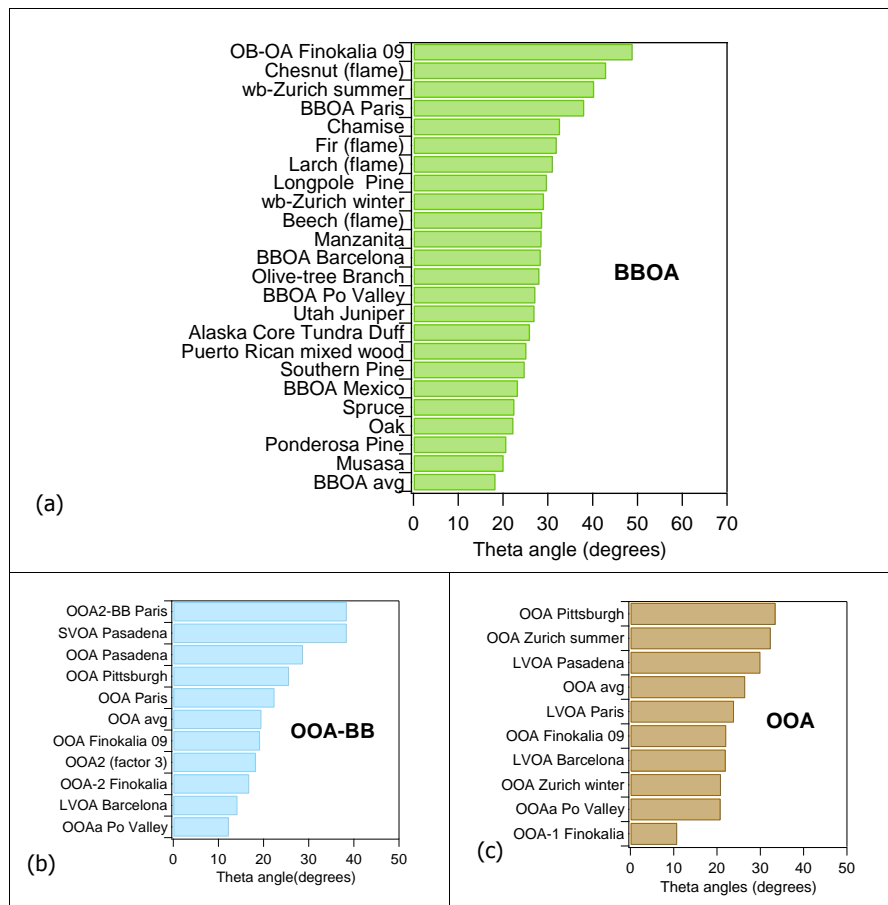


**Fig. 5.** (a) Time series for the components contributing to OA mass loadings at Finokalia during the measurement period as identified by PMF analysis. For comparison, time trace of BC, nitrate and sulfate are shown. The extracted mass spectra for each factor are shown below the time series (b), along with the diurnal profile of each factor (c). Error bars in the diurnal profiles represent the standard deviation of diurnal variability.

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**Fig. 6.** Comparison (theta angles) between the identified factors of this study with relative organic aerosol spectra (color-coded by PMF solution): **(a)** fresh biomass burning factor, **(b)** aged biomass burning factor, and **(c)** oxygenated organic aerosol factor.

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