



Sources and chemical
characterization of
organic aerosol

E. Kostenidou et al.

This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Sources and chemical characterization of organic aerosol during the summer in the eastern Mediterranean

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Received: 29 December 2014 – Accepted: 14 January 2015 – Published: 6 February 2015

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

The concentration and chemical composition of the non-refractory fine particulate matter (NR-PM₁) and black carbon (BC) levels were measured during the summer of 2012 in the suburbs of two Greek cities, Patras and Athens, in an effort to better understand the chemical processing of particles in the high photochemical activity environment of the Eastern Mediterranean. The composition of PM₁ was surprisingly similar in both areas demonstrating the importance of regional sources for the corresponding pollution levels. The PM₁ average mass concentration was 9–14 µg m⁻³. The contribution of sulphate was around 38 %, while organic aerosol (OA) contributed approximately 45 % in both cases. PM₁ nitrate levels were low (2 %). The oxygen to carbon (O : C) atomic ratio was 0.50 ± 0.08 in Patras and 0.47 ± 0.11 in Athens. In both cases the PM₁ was acidic.

Positive matrix factorization (PMF) was applied to the high resolution organic aerosol mass spectra obtained by an Aerodyne High Resolution Aerosol Mass Spectrometer (HR-AMS). For Patras five OA sources could be identified: 19 % very oxygenated OA (V-OOA), 38 % moderately oxygenated OA (M-OOA), 21 % biogenic oxygenated OA (b-OOA), 7 % hydrocarbon-like OA (HOA-1) associated with traffic sources and 15 % hydrocarbon-like OA (HOA-2) related to other primary emissions (including cooking OA). For Athens the corresponding source contributions were: V-OOA (35 %), M-OOA (30 %), HOA-1 (18 %) and HOA-2 (17 %). In both cities the major component was OOA, suggesting that under high photochemical conditions most of the OA in the Eastern Mediterranean is quite aged. The contribution of the primary sources (HOA-1 and HOA-2) was important (22 % in Patras and 33 % in Athens) but not dominant.

1 Introduction

Atmospheric aerosols can affect human health causing cardiovascular and respiratory problems (Davidson et al., 2005; Pope and Dockery, 2006), they reduce visibility (Wat-

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son, 2002) and influence the energy balance of our planet (IPCC, 2007) by scattering or absorbing radiation and changing cloud reflectivity and lifetime. Submicrometer atmospheric particles mainly consist of sulphates, ammonium, organic matter, nitrates, elemental carbon and metals. Organic aerosol (OA) contributes a major fraction of the submicron aerosol mass (Kanakidou et al., 2005; Zhang et al., 2007). The recent development of the Aerodyne HR-AMS (DeCarlo et al., 2006) allows high-time resolution size-resolved measurements of the fine non-refractory inorganic and organic aerosol components. In addition, several techniques have been developed for the deconvolution of the AMS organic mass spectra including custom principal component analysis (Zhang et al., 2005), multiple component analysis (Zhang et al., 2007), and positive matrix factorization (PMF) (Paatero and Tapper, 1994; Lanz et al., 2007). The most recent algorithm, the multilinear engine, (ME-2) (Lanz et al., 2008) is a hybrid of the chemical mass balance (CMB) and bilinear models (e.g., PMF). The combination of AMS measurements with other instrumentation and the use of the corresponding source apportionment techniques can provide valuable information about the ambient aerosol sources and their chemical characterization.

Zhang et al. (2005) showed that summertime OA in a major urban area of the US (Pittsburgh) consisted of oxygenated OA (OOA) and hydrocarbon-like OA (HOA). Lanz et al. (2007) further deconvolved OOA into a more oxygenated OA (OOA-1) and a less oxygenated (OOA-2) component during a summer period in Zurich. They also reported for the first time a wood burning and a charbroiling factor. Marine OA (MOA) was identified by Crippa et al. (2013a) in Paris during summertime, while Schmale et al. (2013) suggested the existence of an amino-acids/amine OA factor (AA-OA), a methanesulfonic acid OA factor (MSA-OA), a marine oxygenated OA factor (M-OOA) and a sea spray OA factor (SS-OA) on Bird Island in the South Atlantic. The major OA components found usually are OOA and HOA (Ng et al., 2010).

Most of the studies in the Eastern Mediterranean and Greece have been based on filter measurements (Manoli et al., 2002; Grivas et al., 2004; Papaefthymiou et al., 2005; Karanasiou et al., 2007; Chrysikou and Samara, 2009) or monitoring just PM

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mass concentrations (Chaloulakou et al., 2003; Vardoulakis and Kassomenos, 2008; Yannopoulos, 2008) in major Greek cities (Athens, Thessaloniki, Patras). During the past years a few field campaigns using continuous PM composition measurement techniques have been conducted in rural/remote areas and urban centres in Greece. At the remote site of Finokalia (Crete) factor analysis on filter samples revealed three sources of coarse particles (crustal, photochemical, and marine aerosols) and two additional factors in the fine mode (residual oil and secondary/combustion aerosols) (Koulouri et al., 2008). For the same site no HOA was detected by PMF analysis of AMS data neither in the summer nor in the winter (Hildebrandt et al., 2010, 2011). For the city of Patras Pikridas et al. (2013) found that the transported pollution accounted for 50 % of the PM_{2.5} during the winter and higher than 70 % during the rest of the year.

In the Western Mediterranean there have been more measurement campaigns (e.g., Viana et al., 2005; Pérez et al., 2008; Pey et al., 2010; Pandolfi et al., 2014). In Barcelona primary OA accounted for 59 % of the OA in the late winter (Mohr et al., 2012). El Haddad et al. (2013) reported that OOA contributed 80 % of the OA mass in Marseille during the summer, while only 5 % of the OA had industrial origin. Nicolas (2013) found that OOA ranged from 70 to 85 % of the total OA over a year at the Cape Corsica station, while the oxidation state of the OA during the summer reached extremely high values with f_{44} (the fraction of the AMS m/z 44, which is an indicator of the oxidation state) higher than 0.25.

Despite the previous efforts, there are still several knowledge gaps related to the characterization and OA sources in the Mediterranean Basin. For example the contribution of the primary sources is still uncertain. The fresh OA from diesel exhaust initially resembles HOA but after aging may resemble semi-volatile OOA (SV-OOA) (Jimenez et al., 2009; Chirico et al., 2010). Aged gas phase emissions from diesel engines, scooters and biomass burning may also produce secondary OA (SOA) with mass spectra similar to SV-OOA (Heringa et al., 2012). In addition, volatile organic compounds (VOCs) in chamber experiments form SOA with mass spectra close to SV-OOA (Ng et al., 2010). The conversion of SV-OOA to more oxidized OA is still very hard

to reproduce in the laboratory. Only recently Platt et al. (2013) showed that after 12 h of aging of gasoline Euro 5 car emissions, SOA with an O : C ratio of 0.7 was produced resembling LV-OOA. In the field, Hildebrandt et al. (2010) demonstrated that primary emissions are becoming highly oxygenated within 1–2 days of transport under intense photochemical conditions. Recently Bougiatioti et al. (2014) showed that a large fraction of biomass burning OA (BBOA) is transformed to OOA even in the dark in less than a day during the summer in Eastern Mediterranean. The objectives of this work are to determine the sources of OA in two Eastern Mediterranean cities and provide insights about its chemical processing.

2 Experimental

2.1 Measurement campaign

The results presented here were part of a larger study, which involved measurements in several areas in Greece (Patra, Athens, Thessaloniki, and Finokalia) both in summer and winter. In this work we will focus on the summer AMS measurements in Patras and Athens.

2.2 Sampling sites in Patras

Patras has approximately 300 000 inhabitants and is located at the Gulf of Patras, at the foothills of a 2 km high mountain. The major activities include a small industrial zone about 16 km southwest of the city center and a harbor around 2.5 km southwest of the city. The nearest major city is Athens, around 220 km to the east. Measurements in Patras were performed simultaneously at two locations: in the center of the city (38°14'46" N, 21°44'08" E) and at the Institute of Chemical Engineering Sciences (38°17'52" N, 21°48'31" E), which is 8 km (north-east) away from the city center and 1 km (south) from the Patras-Athens highway. The site is surrounded by olive tree fields. A few small settlements are located in a distance of 1 km (southwest and north-east).

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2.3 Instrumentation in Patras

A HR-ToF-AMS from Aerodyne Research Inc. (DeCarlo et al., 2006) was measuring the size-resolved chemical composition of the NR-PM₁ aerosol species. The tungsten filament for electron ionization was run at an accelerating voltage of 70 eV, while the vaporizer temperature was set at 600 °C. Alternative runs between V-mode (single stage reflectron) and W-mode (double stage reflectron) were performed with 3 min of measuring time for each mode. In this paper the V-mode data are presented.

A Proton Transfer Reaction Mass Spectrometer (PTR-MS, Ionicon Analytik) was used for the characterization of the volatile organic compounds (VOCs). More details about the VOC measurements are presented in Kaltsonoudis et al. (2015). A Scanning Mobility Particle Sizer, (SMPS, classifier model 3080, DMA model 3081, CPC model 3787, TSI) was operated at a sheath flow rate of 5 L min⁻¹ and a sample flow rate of 1 L min⁻¹. The SMPS measured the number size distribution in the 10–500 nm range. A Multiple-Angle Absorption Photometer (MAAP, Thermo Scientific Inc.) (Petzold and Schönlinner, 2004) was used for the BC measurement. NO_x, SO₂, O₃ and CO concentrations were measured by the corresponding monitors (Teledyne, models T201, 100E, 400E and 300E respectively).

A filter sampler (MetOne SAASS) was used to collect PM_{2.5} samples for inorganic and organic chemical composition analysis. The sampling resolution was 24 h and the flowrate of each filter was 6.7 L min⁻¹. Teflon filters (Whatman 7582 004, 0.2 μm pore size) were used for the measurement of the inorganic anion and cation mass concentrations, using two ion chromatography systems (Metrohm 761 Compact IC). Pre-baked Quartz filters were used for the EC/OC measurement using a laboratory EC/OC analyzer (Sunset Laboratory Inc). More details about the filter extraction procedure are given by Pikridas et al. (2013) and Tsiflikiotou et al. (2015).

The HR-AMS and the PTR-MS measurements covered the period from 8 to 27 June 2012, while the rest of the instrumentation provided measurements from 8 June to 26 July 2012.

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2.4 Sampling site in Athens

Athens is the most populated city in Greece with around 4 million inhabitants. The sampling site was at Demokritos National Center for Scientific Research (37°59'43" N, 23°48'57" E), at the municipality of Agia Paraskevi. This is a suburban background site 10 km from the city center located at the foothills of Ymittos mountain and is surrounded by pine trees. This site is close (0.5 km) to the ring highway of Ymittos and approximately 1.5 km away from the Mesogion highway to the north-west. The closest residences are 0.5 km away.

2.5 Instrumentation in the Athens site

For the measurements in Athens we used our mobile laboratory as a fixed station, in which the HR-AMS, PTR-MS and SMPS (same models as described in session 2.3) were placed. An aethalometer (Magee Scientific, AE31) measured the BC concentrations at 880 nm. An SMPS, (classifier model 3080 TSI, custom DMA, CPC model 3022, TSI) was operated at a sheath flow rate of 3 L min⁻¹ and a sample flow rate of 0.3 L min⁻¹ and measured the number size distribution in the 10–660 nm range. PM_{2.5} was collected on Teflon filters every 24 h for the inorganic composition characterization, while a semi-continuous OC/EC analyzer (Field Instrument, Model 4F, Sunset Laboratory Inc) equipped with a PM_{2.5} inlet and an activated carbon denuder was used for the PM_{2.5} EC/OC measurements with a 3 h resolution.

The sampling period was from 8 to 26 July 2012. Due to technical problems the AMS started measuring on the 12 July 2012.

2.6 Data analysis

For HR-AMS data analysis SQUIRREL v1.51C and PIKA v1.10C (Sueper, 2014) with Igor Pro 6.22 A (Wavemetrics) were used. For the HR-AMS organic mass spectra, we used the fragmentation table of Aiken et al. (2009).

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PMF and ME-2 analysis (Paatero and Tapper, 1994; Lanz et al., 2007, 2008; Ulbrich et al., 2009; Canonaco et al., 2013) were performed using the HR-AMS organic mass spectra in order to investigate the different organic aerosol sources. The m/z 12–200 were used as inputs following the procedure of Ulbrich et al. (2009).

The Patras AMS data were corrected for the collection efficiency CE using the algorithm of Kostenidou et al. (2007) with a 2 h resolution through out the campaign. A shape factor (χ) of 1 was found to be the most appropriate for this data set (Supplement Sect. 1.A, Figs. S1 and S2). The average CE was 0.91 ± 0.10 and it was quite higher compared to other studies (e.g. Crippa et al., 2013a; CE = 0.38). One reason could be that the particles entering the AMS were not dried. Drying the particles usually decreases the CE because the particles bounce on the vaporizer (Matthew et al., 2008). Comparing the PM_{10} AMS sulphate (after CE corrections) to the $PM_{2.5}$ filter sulphate measurements a slope of 1.05 and a high correlation ($R^2 = 0.98$) were found as shown in Fig. S3. The average OA density estimated from the above algorithm was $1.34 \pm 0.21 \text{ g cm}^{-3}$, which is very close to the organic density calculated for Finokalia during the summer of 2008 ($1.35 \pm 0.22 \text{ g cm}^{-3}$, Lee et al., 2010).

For Athens the aerosol was not dried before it entered the AMS, but the particles entering the SMPS were dried to maintain compatibility with long term measurements performed in the site. Thus we modified the algorithm of Kostenidou et al. (2007) converting the ambient AMS mass distributions to dry mass distributions using the relative humidity (RH) inside the sampling line and calculating the inorganic aerosol water content using the Extended Aerosol Inorganic Model II (E-AIM, Carslaw et al., 1995; Clegg et al., 1998; Massucci et al., 1999). As inputs to the E-AIM model we used the inorganic concentrations of sulphate, ammonium and nitrate and the temperature and RH at the entrance of the AMS line. The algorithm was run for shape factors 1–1.4 and the optimum solution was selected (i.e. the minimum of the minimum error scores) which corresponded to the optimum CE, organic density, and shape factor (Supplement Sect. 1.B, Figs. S4 and S5). The average CE was 0.63 ± 0.13 . After applying CE corrections the PM_{10} AMS sulphate correlated well with the $PM_{2.5}$ filter sulphate (Fig. S6)

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The PM₁ aerosol as measured by the AMS was always acidic with an average inorganic cations / anions equivalent ratio of 0.75 ± 0.07 . This indicates that there was no available ammonia to fully neutralize the sulphate and thus the formation of PM₁ ammonium nitrate (from ammonia and nitric acid) in the PM₁ was not thermodynamically favorable.

In order to estimate the organic nitrate mass (ONit) contribution to the total nitrate mass (TotNit) we followed the procedure of Farmer et al. (2010):

$$\frac{\text{ONit}}{\text{TotNit}} = \frac{(1 + R_{\text{ONit}}) \times (R_{[\text{NO}_2^+/\text{NO}^+]_{\text{meas}}} - R_{[\text{NO}_2^+/\text{NO}^+]_{\text{cal}}})}{(1 + R_{[\text{NO}_2^+/\text{NO}^+]_{\text{meas}}}) \times (R_{\text{ONit}} - R_{[\text{NO}_2^+/\text{NO}^+]_{\text{cal}}})} \quad (1)$$

where $R_{[\text{NO}_2^+/\text{NO}^+]_{\text{meas}}}$ is the measured ratio of $\text{NO}_2^+/\text{NO}^+$ ions as a function of time, $R_{[\text{NO}_2^+/\text{NO}^+]_{\text{cal}}}$ is the ratio of $\text{NO}_2^+/\text{NO}^+$ ions obtained during NH_4NO_3 calibrations (0.58 on average) and R_{ONit} is a fixed value set to 0.05, as the minimum ratio of $\text{NO}_2^+/\text{NO}^+$ observed in this campaign was 0.05. The average organic nitrate fraction was 0.91 ± 0.05 , which suggests that most of the nitrate was in the form of organic nitrate (Fig. S10a). The correlation between OA and nitrate was moderate ($R^2 = 0.38$).

3.2 Athens (Demokritos station)

On average the PM₁ mass concentration in Athens (not including dust) was $14.2 \mu\text{g m}^{-3}$. This is a different period than the one in Patras so direct comparisons are not that useful. Figure 1b shows the time series of the concentration of the NR-PM₁ components measured by the AMS in Athens and the BC measured by the aethalometer. The average organic concentration was $6.6 \mu\text{g m}^{-3}$, sulphate $5.3 \mu\text{g m}^{-3}$, ammonium $1.4 \mu\text{g m}^{-3}$ and BC $0.7 \mu\text{g m}^{-3}$. Similarly to Patras nitrate levels were low, $0.2 \mu\text{g m}^{-3}$ on average.

The average O : C mass ratio was 0.47 ± 0.11 , lower than in Patras (0.50), because of the higher contribution of primary emissions (Sects. 3.3.1 and 3.3.2). The average

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OM:OC ratio was 1.76 ± 0.14 . The diurnal cycle of O:C is shown in Fig. 2 and is similar to the one in Patras. The O:C exhibited two peaks one in the early morning around 03:00–05:00 LT and a second in the late afternoon 17:00–20:00 LT. The OA profile is characterized by three peaks (Fig. S7b). Two of them, around 07:00–12:00 LT and 20:00–24:00 LT are related to fresh emissions since the BC follows the same trend. A smaller increase around 14:00–17:00 LT is probably due to photochemistry. On average f_{44} and f_{57} were 0.13 and 0.02 respectively (Fig. S8b).

The OM based on the $PM_{2.5}$ semi-continuous OC/EC measurements was calculated using the OM:OC ratio provided by the AMS. Figure S9b illustrates the correlation for the organics between the AMS and the semi-continuous OC/EC samples. The measurements are in reasonable agreement ($R^2 = 0.47$), with the AMS providing slightly higher concentrations.

The equivalent ratio of PM_1 anions to cations (measured by the AMS) was on average 0.70 ± 0.09 and thus that the aerosol was acidic. Again free ammonia was not enough to neutralize the sulphate and thus there was no ammonium nitrate observed in the PM_1 fraction.

Applying Eq. (1) to the Athens data set (with an average $R_{[NO_2^+ / NO^+]}_{cal} = 0.68$, and $R_{ONit} = 0.12$), the organic nitrate fraction was 0.89 ± 0.08 , indicating that most of the nitrate was actually organic nitrate (Fig. S10b). The correlation coefficient between OA and nitrate was $R^2 = 0.62$.

3.3 OA sources

3.3.1 Patras

For the PMF analysis both PMF Evaluation Tool, PET, (Lanz et al., 2008; Ulbrich et al., 2009) and ME-2 (Lanz et al., 2008; Canonaco et al., 2013) solutions were examined and evaluated using the HR organic mass spectra. A five factor solution was chosen as the best to describe the major sources of OA in Patras. More information about the choice of the solution is provided in the Supplement (Sect. 6.A, Figs. S11–S17). The

factors corresponded to very oxygenated OA (V-OOA, 19%), moderately oxygenated OA (M-OOA, 38%), biogenic oxygenated OA (b-OOA, 21%), HOA-1 (7%) and HOA-2 (15%). The assignment of the factors to specific types was based on their mass spectra and diurnal profile characteristics: V-OOA had a high contribution of m/z 44, M-OOA had a moderate m/z 44 contribution, b-OOA was characterized by both biogenic and oxidized OA signatures, HOA-1 was similar to the literature HOA factor related to traffic emissions and HOA-2 included primary cooking and aged traffic emissions. Overall, the oxygenated OA was the dominant component (78%) and the primary OA accounted for only 22%. The HR mass spectrum of each factor is illustrated in Fig. 3a, while their time series are shown in Fig. 4a and their average diurnal profiles in Fig. 5a.

V-OOA was characterized by the m/z 44 ($\sim 22\%$) with an O : C = 0.81 and is related to aged aerosol (Fig. 3a). The V-OOA factor correlated well with sulphate ($R^2 = 0.48$) and ammonium ($R^2 = 0.51$), which is typical for highly oxygenated OA. Its correlation with nitrate was low ($R^2 = 0.09$). The V-OOA diurnal profile was almost flat (Fig. 5a). R^2 between V-OOA and individual VOCs measured by the PTR-MS were all less than 0.13.

M-OOA had a pronounced but of lower intensity m/z 44 ($\sim 16\%$) and an O : C = 0.54 implying aged but less oxidized particles (Fig. 3a). Nitrate has been suggested as a tracer for the less oxygenated species (e.g. Lanz et al., 2007; Mohr et al., 2012), however there was very little nitrate during our measurements and most of it was organic nitrate resulting in a weak correlation between the M-OOA and nitrate ($R^2 = 0.04$). M-OOA had a very similar pattern with the solar radiation peaking at around 14:00 LT. M-OOA had low correlation with methyl vinyl ketone (MVK) and methacrolein (MACR) (m/z 71, $R^2 = 0.31$), hydroxyacetone (m/z 75, $R^2 = 0.29$), C₅ carbonyls/2-methylbutene-2-ol (MBO)/methacrylic acid (m/z 87, $R^2 = 0.29$), terpene oxidation products (m/z 113, $R^2 = 0.30$) and nopinone (m/z 139, $R^2 = 0.28$), (Table S1). Comparing the M-OOA spectrum to the α -pinene SOA spectrum of Heringa et al. (2012) the angle theta was quite high ($\theta = 35^\circ$), while the similarity between the M-OOA and the toluene photooxidation SOA spectrum (Kostenidou et al., not published data) was greater

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($\theta = 16^\circ$). Thus this factor may include contributions by both anthropogenic and biogenic sources.

The biogenic OOA factor had an $f_{44} = 0.13$ and O : C = 0.48 (Fig. 3a) which indicates a relatively moderate degree of oxygenation. It was characterized by a distinct m/z 82 (mainly composed of $C_5H_6O^+$), an elevated m/z 53 (mostly $C_4H_5^+$) and a significant contribution at m/z 39 (5 %, mainly CH_3O^+). These characteristics were similar to those found at a rural area in Ontario, Canada (Slowik et al., 2011), downtown Atlanta, Georgia, USA (Budisulistiorini et al., 2013), in tropical rainforests in the central Amazon Basin (Chen et al., 2014) and in Borneo, Malaysia (Robinson et al., 2011) and in Centreville in rural Alabama (Xu et al., 2014) which were associated to secondary OA produced by isoprene photooxidation. Using the HYSPLIT back trajectory model (Draxler and Rolph, 2013), the mass concentration of b-OOA was almost zero when the air masses were coming from the west (e.g. Ionian Sea, 11–15 June, Fig. S18), while M-OOA and V-OOA were still high. However, the b-OOA increased in periods (e.g. 10–11 June, 16–17 and 24–26 June) when the air masses passed over the forested mountains of Central Greece (Fig. S18), which is an area characterized by high terpene and isoprene emissions (Karl et al., 2009). This supports the biogenic character of this factor. The highest concentration of b-OOA was at 06:00 in the morning on 16 June ($5.4 \mu\text{g m}^{-3}$). b-OOA exhibited low correlation with isoprene (m/z 69, $R^2 = 0.13$), isoprene peroxides (m/z 101, $R^2 = 0.28$) and the first generation isoprene products such as MVK and MACR (m/z 71, $R^2 = 0.21$) (Table S1). However, it correlated better with acetone (m/z 59, $R^2 = 0.35$), hydroxyl-acetone (m/z 75, $R^2 = 0.41$), PAN (m/z 77, $R^2 = 0.37$), nopinope (m/z 139, $R^2 = 0.39$) and pinonaldehyde (m/z 151, $R^2 = 0.30$) which are products of terpenes ozonolysis (Matsunaga et al., 2003; Holzinger et al., 2005; Lee et al., 2006).

HOA-1 was characterized mainly by the m/z 39, 41, 43, 55, 57, 67, 69 and 81 (Fig. 3a) which are typical hydrocarbon fragments of fresh traffic emissions (Zhang et al., 2005; Aiken et al., 2009). Its O : C was 0.1 which is in the range found in literature (the HOA factor of Mohr et al. (2012) had an O : C = 0.03, while the HOA factor of

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Ulbrich et al. (2009) had an O : C = 0.18). HOA-1 had a medium correlation with BC ($R^2 = 0.38$) and nitrate ($R^2 = 0.27$). The HOA-1 diurnal profile was characterized by two peaks during the morning (08:00) and evening rush hours (21:00) (Fig. 5a). HOA-1 correlated moderately with benzene (m/z 79, $R^2 = 0.36$), toluene (m/z 93, $R^2 = 0.35$), xylenes (m/z 107, $R^2 = 0.37$), C₉ aromatic compounds (m/z 121, $R^2 = 0.45$). The correlation with NO_x was relatively low ($R^2 = 0.26$).

The HOA-2 O : C was 0.21. HOA-2 had a significant contribution at the m/z 39, 41, 43, 44, 55, 57 and 67 (Fig. 3a) which are features of cooking organic aerosol, COA (Ge et al., 2011a, b; Crippa et al., 2013a, b). However, the reported COA spectra (e.g. Ge et al., 2011a, b; Crippa et al., 2013a, b) have lower contributions at m/z 44 (0.011–0.022), indicating less oxygenation. The f_{44} in the HOA-2 mass spectrum was 0.034 which implies that this factor may also contain species that have been oxidized to some degree. The HOA-2 diurnal profile had a small peak around 14:00 and a higher one around 22:00, which are consistent with the Greek lunch and dinner periods (Fig. 5a). Thus, the HOA-2 in Patras is mainly due to charbroiling of meat cooking OA. The correlation between HOA-2 and BC, nitrate, benzene, toluene, xylenes and C₉ aromatic compounds was moderate ($R^2 = 0.38, 0.38, 0.33, 0.30, 0.33$ and 0.43 correspondingly).

During 16–23 June the wind speed was relatively high (average 5.3 ms^{-1}) compared to the rest of the sampling days (average 2.5 ms^{-1}). The contribution of the local sources (HOA-1 and HOA-2) was less than 9% of the OA during that windy period compared to 28% during the remaining days (Fig. 4a).

3.3.2 Athens

Four OA factors could be identified in the Athens AMS data set: V-OOA (35%), M-OOA (30%), HOA-1 (18%) and HOA-2 (17%). A detailed description of the reasons for this selection can be found in the Supplement (Sect. 6.B, Figs. S19–S25). The corresponding mass spectra are provided in Fig. 3b. The time series of the four PMF

factors are shown in Fig. 4b, while their diurnal cycles in Fig. 5b. Similarly to Patras the contribution of the oxygenated OA in Athens was high (65%), while the primary sources contributed 35%.

The very oxidized OA (V-OOA) had an $f_{44} = 0.18$ and $O:C = 0.68$ and showed a good correlation with sulphate ($R^2 = 0.53$) and ammonium ($R^2 = 0.50$) consistent with the aged character of this factor. The diurnal profile of the V-OOA (Fig. 5b) was characterized by a peak around 15:00–18:00, which is probably associated with production of this component in the afternoon over the region. At the same time M-OOA decreased, which indicates that the V-OOA is a product of photochemical processing. V-OOA correlated with formic acid (m/z 47, $R^2 = 0.47$), hexenal (m/z 99, $R^2 = 0.42$), isoprene peroxides (m/z 101, $R^2 = 0.35$), terpene oxidation products (m/z 113, $R^2 = 0.40$), and heptanal (m/z 115, $R^2 = 0.42$) (Table S1).

The moderately oxidized OA (M-OOA) was characterized by an $f_{44} = 0.14$ and $O:C = 0.56$ and exhibited a weak correlation with sulphate, ammonium and nitrate ($R^2 = 0.17, 0.17$ and 0.13 respectively). M-OOA increased during the day with a maximum at 12:00–14:00, following the diurnal profile of solar radiation, which implies relatively fast photochemical reactions. M-OOA had a second peak during the night around 24:00, which could be an indication of nighttime production. M-OOA did not show any correlation (R^2 less than 0.07) with the measured VOCs.

The HOA-1 $O:C$ was 0.07. Surprisingly it had a rather weak correlation with BC ($R^2 = 0.05$) and it also showed a low correlation with nitrate ($R^2 = 0.13$). The HOA-1 diurnal profile had 2 peaks, a small increase during the morning (07:00) and a larger peak in the evening hours (20:00), consistent with the contribution of traffic emissions. HOA-1 did not correlate with the VOCs characteristic of traffic such as benzene, toluene and xylenes (the R^2 were correspondingly 0.15, 0.13 and 0.16) and inorganic gases as NO_x ($R^2 = 0.06$). The HOA-1 mass spectrum and time series were very stable for 2 to 5 factorial solutions and for f_{peaks} values from -2 to 2 . The R^2 between the HOA-1 of the selected solution and the HOA-1 of the solutions of 2, 3 and 5 factors was always greater than 0.973 (both for the time series and the mass spectra). This weak

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correlation between the HOA-1 factor and other primary organic pollutants indicates that their concentrations were not dominated by the same sources. For example HOA-1 possibly originated mostly from passenger cars while BC from diesel vehicles. The location of the sampling site and the inhomogeneity of the surrounding areas, in combination with the wind direction change have confounded these effects. The rose plots of HOA-1, BC, NO_x and benzene for wind speeds greater than 1 m s⁻¹ indicate that BC, NO_x and benzene had the same origin, while HOA-1 did not. For example at 08:00 the HOA-1 was on average coming from southwest (Fig. S26–S29) likely from the ring highway of Ymittos, while BC, NO_x and benzene from the north probably influenced by the Mesogion highway.

HOA-2 had an O : C = 0.24 and exhibited a good correlation with BC ($R^2 = 0.57$). HOA-2 also correlated well with nitrate ($R^2 = 0.75$) implying that organic nitrate compounds were possibly emitted or produced along with this OA type. The HOA-2 profile had 2 peaks at around 11:00 and 22:00. The second peak is characteristic of Greek dinner period, thus part of HOA-2 could be attributed to meat cooking OA. However, the first peak can not be explained by the Greek lunch period. The correlation with the BC implies that HOA-2 and BC had the same origin. Comparing the HOA-2 mass spectrum with aged POA or SOA emissions from other sources such as α -pinene, wood burning, scooter and diesel (Heringa et al., 2012) the angles θ were 19, 19, 28, 17° correspondingly. This indicates that HOA-2 could include aged diesel emissions since α -pinene is not associated with BC and there were no observed biomass burning events during the sampling period. The HOA-2 factor correlated with the m/z 43 ($R^2 = 0.43$), acetone (m/z 59, $R^2 = 0.44$), methyl ethyl ketone (MEK, m/z 73, $R^2 = 0.49$), benzene (m/z 79, $R^2 = 0.62$), toluene (m/z 93, $R^2 = 0.54$), xylenes (m/z 107, $R^2 = 0.56$), C₉ aromatic compounds (m/z 121, $R^2 = 0.58$) and C₁₀ aromatic compounds (m/z 135, $R^2 = 0.55$). It also had a good correlation with NO_x ($R^2 = 0.58$).

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3.3.3 Comparison of the PMF factors in the two cities

The spectra of the V-OOA factors in the two cities were almost the same ($\theta = 6.7^\circ$). However, the Athens V-OOA exhibited lower f_{44} and O : C ratio (0.18 and 0.68 correspondingly) compared to Patras V-OOA (0.22 and 0.81 respectively). This difference could be partially due to the different periods of the two measurements.

The two M-OOA mass spectra were even more similar to each other ($\theta = 5.4^\circ$). A high correlation was also observed between the two HOA-2 mass spectra ($\theta = 5.9^\circ$). The Athens HOA-2 was slightly more oxygenated (O : C = 0.24), as the oxygenated part of m/z 43 was more elevated, 65 %, compared to 51 % observed in Patras HOA-2 and the f_{44} was just slightly higher (0.07), while in Patras HOA-2 was 0.06.

The two HOA-1 factors exhibited a lower correlation with each other ($\theta = 22.8^\circ$). The main differences were at m/z 41, 43, 55, 57, 69, 71, 81, 83 and 85 which were more abundant in the Athens HOA-1 spectrum, while in the Patras spectrum the f_{44} was higher (0.04) compared to Athens (0.02). In the Patras HOA-1 spectrum the m/z 57 was lower in comparison with m/z 55. The O : C in Athens HOA-1 was somehow lower (0.07) than in Patras HOA-1 (0.10).

3.3.4 Comparing the PMF factors with other studies

Table 1 shows comparisons of the mass spectra of the factors found in the two cities with selected PMF factors from the literature (Aiken et al., 2009; Docherty et al., 2011; Sun et al., 2011; Robinson et al., 2011; Heringa et al., 2012; Mohr et al., 2012; Ge et al., 2012a, b; Crippa et al., 2013a, b; Budisulistiorini et al., 2013; Xu et al., 2014) that have all been extracted using the fragmentation table of Aiken et al. (2009).

The two V-OOA factors were quite similar with LV-OOA at Riverside (Docherty et al., 2011), $\theta = 8-10^\circ$, and with LV-OOA at New York City (Sun et al., 2011), $\theta = 11-14^\circ$. They also showed good correlation with LV-OOA measured in Barcelona (Mohr et al., 2012) and Paris (Crippa et al., 2013a, b) $\theta = 14-19^\circ$.

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M-OOA had moderate to low agreement with the majority of the literature profiles. The lowest angle θ (around 22–24°) corresponded to the comparison with the SV-OOA found in Mexico City (Aiken et al., 2009). It did however resemble ($\theta = 13$ –16°) the toluene SOA spectrum (Kostenidou et al., not published data).

The b-OOA factor in Patras correlated moderately with α -pinene SOA, $\theta = 29^\circ$, (Heringa et al., 2012) and the isoprene factor found in Alabama, $\theta = 28^\circ$, (Xu et al., 2015). However, it had low correlation with the Factor 82, $\theta = 47^\circ$, found in Malaysia (Robinson et al., 2011) and the IEPOX OA factor extracted in Atlanta, $\theta = 76^\circ$, (Budisulistiorini et al., 2013).

The HOA-1 factors correlated well with most of the literature HOA profiles, $\theta = 10$ –22°, except Paris during the winter (Crippa et al., 2013b).

The HOA-2 mass spectra resembled the COA factor extracted in New York City, $\theta = 11$ –14°, (Sun et al., 2011) and the SOA from diesel VOCs emissions, $\theta = 17^\circ$, (Heringa et al., 2012).

Figure 6 summarizes Patras and Athens OA measurements and PMF factors in the Ng triangle (Ng et al., 2010). All the data fall within the triangle.

3.4 Discussion

In both cities the composition of NR-PM₁ was surprisingly similar. Organics' contribution in Patras and Athens was around 45 %, which is similar to other areas in Europe: London (UK) 46 %, mountain Taunus (Germany) 59 %, Melpitz (Germany) 59 %, Mace Head (Ireland) 39 %, Po Valley (Italy) 33 %, and Paris (France) 50 % (Cubison et al., 2006; Hings et al., 2007; Poulain et al., 2011; Dall'Osto et al., 2010; Saarikoski et al., 2012; Crippa et al., 2013a). With 38 %, sulphate made a larger contribution compared to other studies in Europe during the summer: London 31 %, mountain Taunus 24 %, Mace Head 32 %, Melpitz 22 %, Po Valley 9 %, and Paris 25 % (Cubison et al., 2006; Hings et al., 2007; Dall'Osto et al., 2010; Poulain et al., 2011; Saarikoski et al., 2012; Crippa et al., 2013a). Nitrate contributed very little (less than 2 %) and was mostly attributed to organonitrate compounds, in contrast with other European studies where

nitrate ranged from 6 % (Melpitz) to 39 % (Po Valley) (Poulain et al., 2011; Saarikoski et al., 2012) and mainly was ammonium nitrate. The absence of particulate nitrate in PM₁ was also observed at Finokalia (Hildebrandt et al., 2010) and is characteristic of the Eastern Mediterranean. Ammonia levels in this region are quite low (Wichink Kruit et al., 2012) to fully neutralize the existing relatively high sulphate.

The O : C ratios (0.50 for Patras and 0.47 for Athens) were moderately high. The O : C ratio at Finokalia was 0.8 (Hildebrandt et al., 2010), at Cape Corsica 0.9 (Nicolas, 2013), while in Paris 0.38 (Crippa et al., 2013a).

In both cities the OOA was the dominant OA component (78 % in Patras and 65 % in Athens). This fraction is within the range that has been measured in previous summer studies in the Mediterranean. For example, the OOA at Finokalia was 100 % of the OA (Hildebrandt et al., 2010), in Marseille accounted for 80 % (El Haddad et al., 2013), at Cape Corsica 80–85 % (Nicolas, 2013), while in Po Valley 61 % (Saarikoski et al., 2012).

In Athens 35 % of the OA was V-OOA, while in Patras only 19 % of the OA was attributed to V-OOA. In addition the V-OOA in Athens was increasing in the afternoon, suggesting its production either locally or regionally. In Patras V-OOA had an almost flat diurnal profile. This difference could be attributed to the different air masses that arrive to each site. According to back trajectory analysis, based on FLEXPART (Stohl et al., 2005) (Fig. S30) and HYSPLIT (Draxler and Rolph, 2013) during most of the sampling days in Athens the air masses had spent considerable time over the source-poor Aegean Sea, while the majority of the air masses that arrived in Patras were continental and passed over the mountains of Central Greece.

4 Conclusions

During the summer of 2012 the air pollution in Patras (June) and Athens (July) was monitored continuously. The sum of the NR-PM₁ and BC concentration was on average 8.6 µg m⁻³ in Patras and 14.2 µg m⁻³ in Athens. However, the aerosol composition

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was quite similar in both areas: 45 % OA, 38 % sulphate, 11 % ammonium, 1 % nitrate (mostly organic) and 5 % BC indicating the importance of regional sources. In both cities the fine aerosol was acidic.

For Patras the average O : C ratio was 0.50 ± 0.08 , while in Athens 0.47 ± 0.11 . An HOA-2 factor was found in both cities attributed to primary emissions such a meat cooking. This factor may also contain oxygenated primary emissions (e.g., aged diesel emissions). In Patras a biogenic oxidized OA factor could be identified, which was related to air masses passing over the forests of Central Greece. In both cities oxygenated OA was the major source of organic aerosol (78% in Patras and 65% in Athens), indicating the impact of the transported pollution in Mediterranean. The contribution of the primary sources was significant (22 % in Patras and 35 % in Athens) but not dominant.

The Supplement related to this article is available online at doi:10.5194/acpd-15-3455-2015-supplement.

Acknowledgements. The authors are grateful to Evangelos Louvaris and Magda Psichoudaki for their assistance with the measurements in Patras. This research was supported by the European Research Council Project ATMOPACS (Atmospheric Organic Particulate Matter, Air Quality and Climate Change Studies) (Grant Agreement 267099) and the European FP7 project PEGASOS. This research has been co-financed by the European Union (European Social Fund – ESF) and Greek national funds through the Operational Program “Education and Lifelong Learning” of the National Strategic Reference Framework (NSRF) – Research Funding Program: THALES.

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**Table 1.** Correlations between PMF factors from Patras and Athens and PMF factors from selected studies.

	Angle (°) with V-OOA Patras	Angle (°) with V-OOA Athens
LV-OOA Mexico City ¹	21	20
LV-OOA Riverside ²	10	8
LV-OOA Barcelona ³	18	16
LV-OOA Paris (SIRTA) summer ⁴	19	15
OOA Paris (SIRTA), winter ⁵	14	16
LV-OOA New York City ⁶	14	11
LV-OOA Fresno ^{7,8}	36	30
	M-OOA Patras	M-OOA Athens
SV-OOA Mexico City ¹	24	22
SV-OOA Riverside ²	42	39
SV-OOA Barcelona ³	33	31
SV-OOA Paris (SIRTA) summer ⁴	42	39
SV-OOA New York City ⁶	30	27
α -pinene ozonolysis SOA aged ⁹	35	32
Toluene photooxidation (HONO) SOA ¹⁰	16	13

Table 1. Continued.

	Angle (°) with	Angle (°) with
	HOA-1 Patras	HOA-1 Athens
HOA Mexico City ¹	13	14
HOA Riverside ²	20	10
HOA Barcelona ³	22	11
HOA Paris (SIRTA) summer ⁴	16	18
HOA Paris (SIRTA), winter ⁵	33	31
HOA New York City ⁶	12	10
HOA Fresno ^{7,8}	11	11
	HOA-2 Patras	HOA-2 Athens
COA Barcelona ³	77	77
COA Paris (SIRTA), summer ⁴	30	34
COA Paris (SIRTA), winter ⁵	41	42
COA Paris (LHVP), winter ⁵	40	42
COA New York City ⁶	11	14
COA Fresno ^{7,8}	28	33
Aged VOCs diesel emissions ⁹	17	17
α -pinene ozonolysis SOA aged ⁹	20	19
Toluene photooxidation (HONO) SOA ¹⁰	26	23
	b-OOA Patras	
α -pinene ozonolysis SOA aged ⁹	29	
Factor 82 ¹¹	47	
IEPOX OA ¹²	76	
Isoprene-OA ¹³	28	

¹ Aiken et al. (2009), ² Docherty et al. (2011), ³ Mohr et al. (2012), ⁴ Crippa et al. (2013a),

⁵ Crippa et al. (2013b), ⁶ Sun et al. (2011), ^{7,8} Ge et al. (2012a, b), ⁹ Heringa et al. (2012),

¹⁰ Kostenidou et al., (not published data), ¹¹ Robinson et al. (2011), ¹³ Budisulistiorini

et al. (2013), ¹³ Xu et al. (2014).

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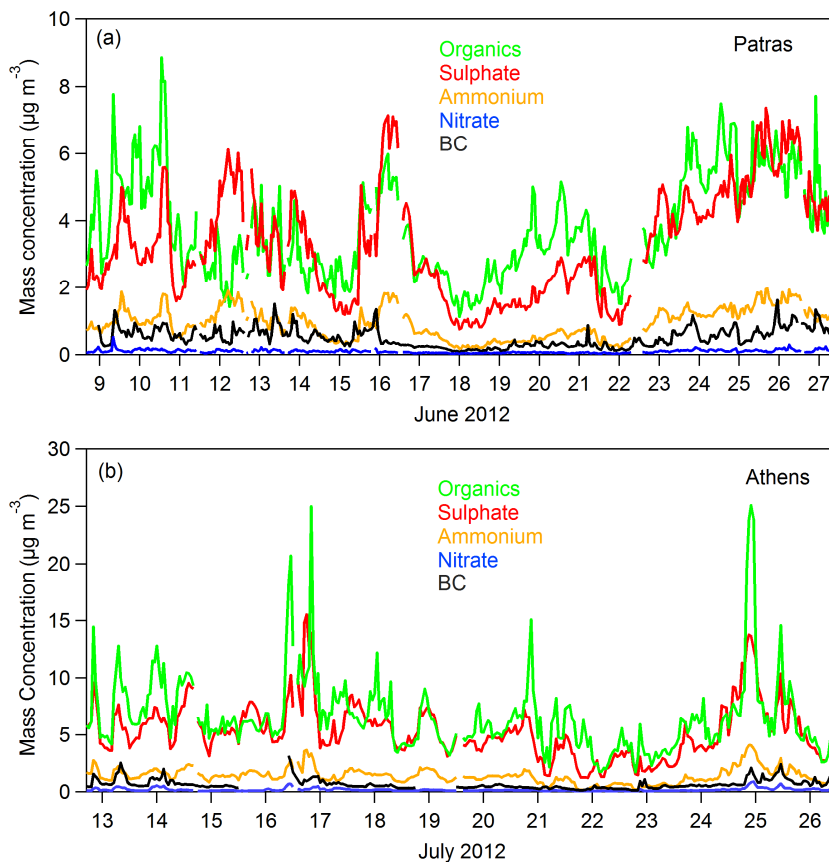


Figure 1. Time series of organics, sulphate, ammonium and nitrate mass concentration measured by the AMS (corrected for the CE) and BC (a) for Patras and (b) for Athens. The BC was provided by MAAP for Patras measurements and by an aethalometer for Athens.

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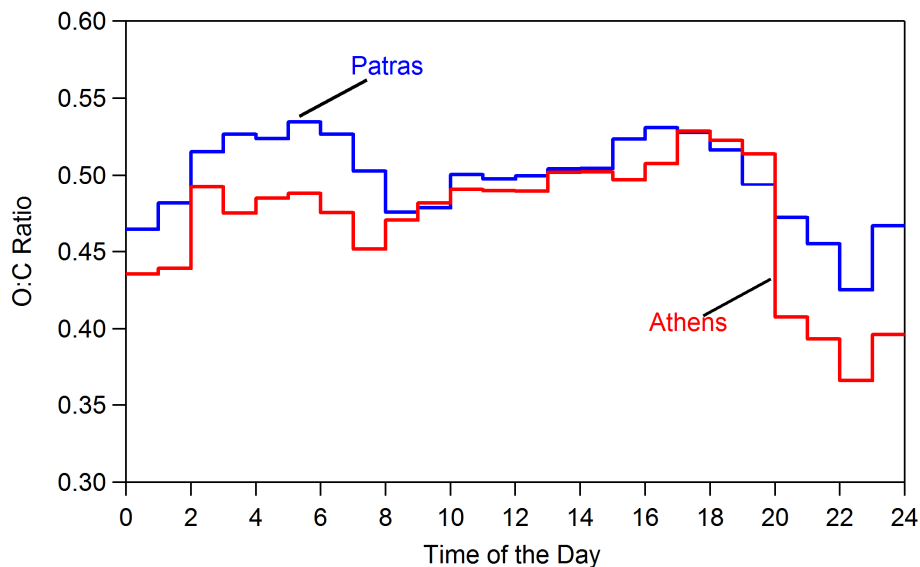


Figure 2. Diurnal profile of O : C ratio for Patras (blue line) and Athens (red line) data set. The O : C was calculated with the Aiken et al. (2009) fragmentation table.

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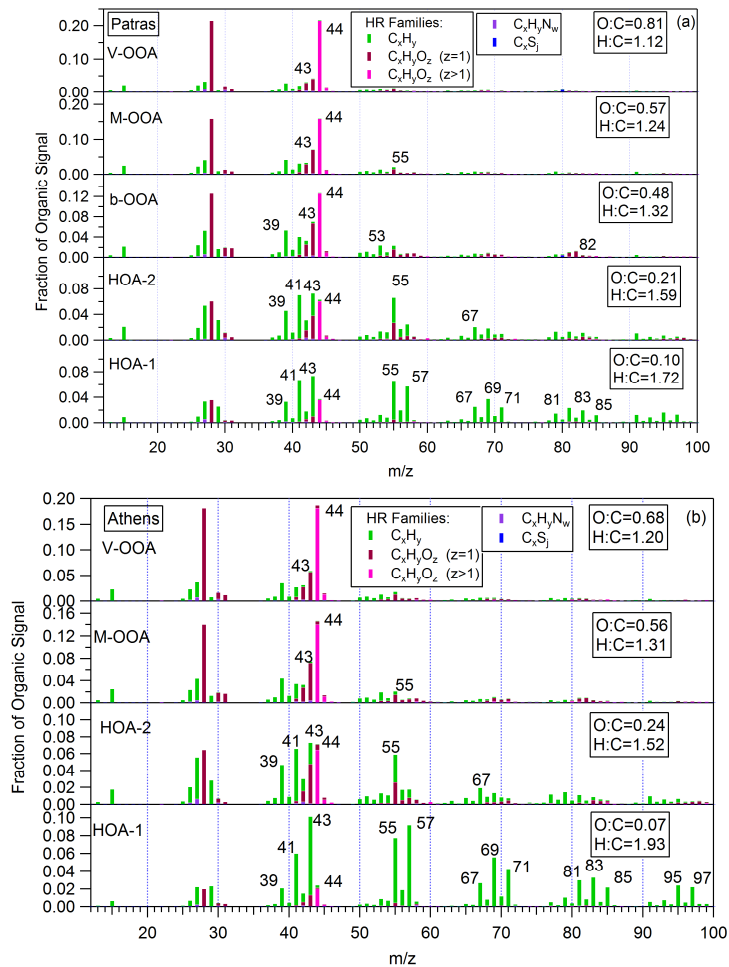


Figure 3. HR mass spectra profiles of the sources found **(a)** in Patras and **(b)** in Athens.

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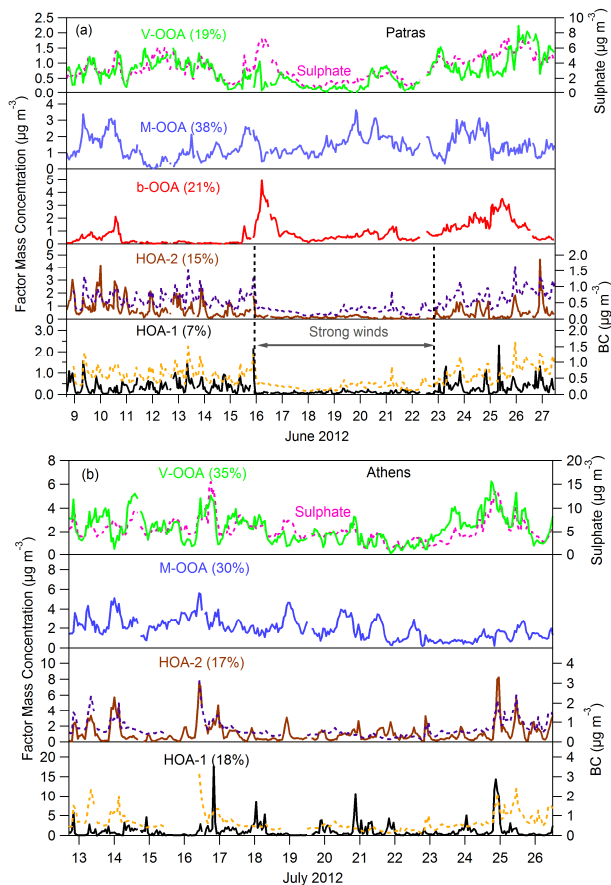


Figure 4. Time series **(a)** of the five PMF factors using HR organic mass spectra for Patras and **(b)** of the four PMF factors found in Athens. For the Patras measurements the HOA-1 and HOA-2 contribution was very low between the 16 and 23 June 2012 due to the high wind speed during that period.

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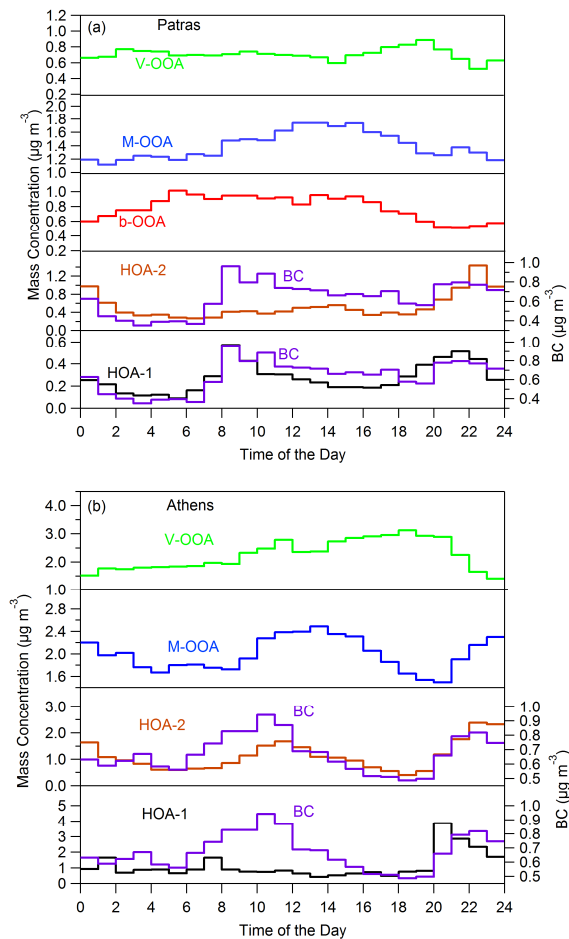


Figure 5. Diurnal cycles of the PMF factors (a) in Patras and (b) in Athens.

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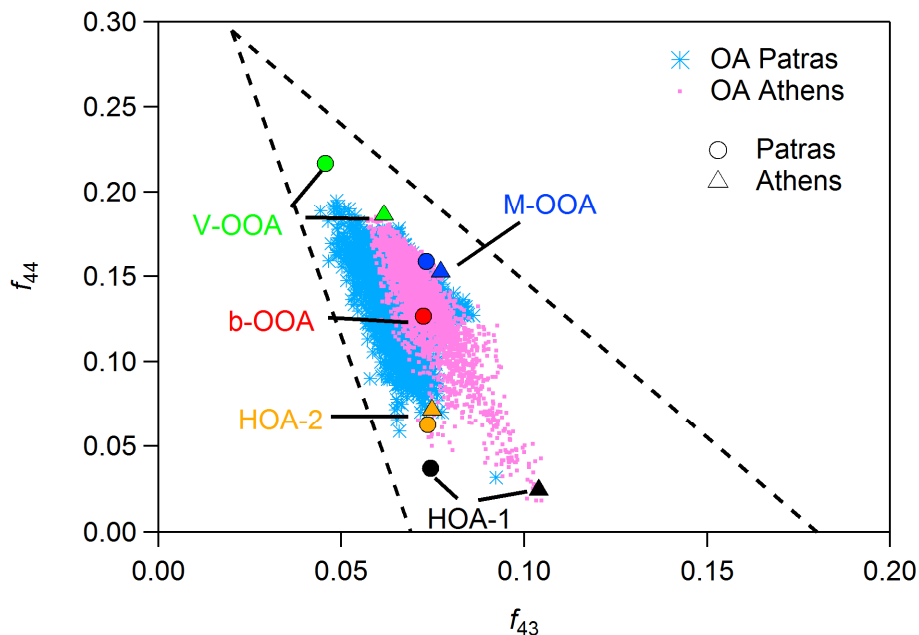


Figure 6. Patras (circles) and Athens (triangles) factors in the Ng et al. (2010) triangle. In both cases OA fall within the triangle. All the measurements in Patras (blue symbols) and Athens (pink symbols) are also shown.

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