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Joint analysis of continental and regional background environments in the western Mediterranean: PM_1 and PM_{10} concentrations and composition

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Abstract. The complete chemical composition of atmospheric particulate matter (PM_1 and PM_{10}) from a continental (Montsec, MSC, 1570 m a.s.l.) and a regional (Montseny, MSY, 720 m a.s.l) background site in the western Mediterranean Basin (WMB) were jointly studied for the first time over a relatively long-term period (January 2010–March 2013).

Differences in average PM_X concentration and composition between both sites were attributed to distance to anthropogenic sources, altitude, and different influence of atmospheric episodes. All these factors result in a continental-to-regional background increase of 4.0 $\mu g\,m^{-3}$ for PM_{10} and 1.1 $\mu g\,m^{-3}$ for PM_1 in the WMB. This increase is mainly constituted by organic matter, sulfate, nitrate, and sea salt. However, higher mineral matter concentrations were measured at the continental background site owing to the higher influence of long-range transport of dust and dust resuspension.

Seasonal variations of aerosol chemical components were attributed to evolution of the planetary boundary layer (PBL) height throughout the year, variations in the air mass origin, and differences in meteorology. During warmer months, weak pressure gradients and elevated insolation generate recirculation of air masses and enhance the development of the PBL, causing the aging of aerosols and incrementing pollutant concentrations over a large area in the WMB, including the continental background. This is reflected in a more similar relative composition and absolute concentrations of continental and regional background aerosols. Nevertheless, during colder months the thermal inversions and the lower vertical development of the PBL leave MSC in the free troposphere most of the time, whereas MSY is more influenced by regional pollutants accumulated under winter anticyclonic conditions. This results in much lower concentrations of PM_X components at the continental background site with respect to those at the regional background site.

The influence of certain atmospheric episodes caused different impacts at regional and continental scales. When long-range transport from central and eastern Europe and from north Africa occurs, the continental background site is frequently more influenced, thus indicating a preferential transport of pollutants at high altitude layers. Conversely, the regional background site was more influenced by regional processes.

Continental and regional aerosol chemical composition from the WMB revealed (a) high relevance of African dust transport and regional dust resuspension; (b) low biomass burning contribution; (c) high organic matter contribution; (d) low summer nitrate concentrations; and (e) high aerosol homogenization in summer.

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1 Introduction

The influence of atmospheric particulate matter (PM) on the Earth's radiative budget generates a strong scientific interest because of its effect on climate. Atmospheric PM interacts with the Earth's climate system by scattering and absorbing solar radiation (direct climate forcing effect), and by acting as cloud condensation nuclei (indirect climate forcing effect) (IPCC, 2013). Aerosols also have adverse effects on air quality (Directive 2008/50/EC) and human health (WHO, 2013), as well as on ecosystems (e.g. Burkhardt and Pariyar, 2014). The size distribution of aerosol chemical components is a key factor in modulating these effects, and it provides valuable information on the aerosol origins and sources. Aerosol chemical composition measurements carried out at complex sites such as urban areas constrain the assessment of the origin of regional and long-range-transported aerosols, as local sources prevail. For this reason, measurements performed at a sufficient distance from large emission sources are needed to determine background conditions and to evaluate air mass transport effects. Furthermore, an improved understanding of synoptic and mesoscale meteorological effects is necessary to develop a better predictive capability of air quality and climate models.

Although there is not a well-established definition, continental background environments can be described as representative of the air quality of a wide area of hundreds of kilometers, as proposed by Laj et al. (2009), with the absence of local emissions. However, aerosols found in this type of environments are not purely natural; the presence of some pollutants in these sites indicates that they are affected by long-range transport of anthropogenic emissions, since generally they are isolated from large polluted areas (> 50 km) (Putaud et al., 2010). For this reason, these environments are also classified as remote sites. In many cases the monitoring sites chosen to represent this type of environments are located in mountaintops over 1000 m a.s.l.; therefore they are also called high-altitude sites (Nyeki et al., 1998) or free troposphere (FT) environments (Andrews et al., 2011). The areas located at sufficient distance from large anthropogenic sources but frequently within the planetary boundary (PBL) are classified as regional background environments (Putaud et al., 2010). These environments are representative of the air quality of a less extensive area, and they are more influenced by regional transport of polluted air masses than continental background environments.

Aerosol chemical characterization has been performed at many locations across Europe, providing information on PM_{10} and $PM_{2.5}$ chemical composition from different types of environments (e.g. Putaud et al., 2010), improving the knowledge on the variation and trends of PM composition (e.g. Cusack et al., 2012) and increasing the understanding of PM sources (Belis et al., 2013). Nevertheless, the PM_1 fraction remains relatively understudied, especially outside urban areas. Most studies focusing on PM_1 have been carried out within the PBL, whereas measurements at continental background sites in Europe are scarce and were mostly taken in short-term measurement campaigns (e.g. Carbone et al., 2010; Marenco et al., 2006). The study of PM_1 chemical composition at continental background sites may be necessary to assess the contribution of regional and long-range transport, since it is in the PM_1 fraction where most of the anthropogenic constituents are concentrated (Minguillón et al., 2012; Pérez et al., 2008b).

Among the few long-term European studies at continental background environments, Cozic et al. (2008) investigated the chemical composition of coarse and PM_1 aerosols for 7 years at the high Alpine site of Jungfraujoch (Switzerland). Bourcier et al. (2012) studied PM_{10} and PM_1 water-soluble inorganic components over 1 year at the high-altitude site of Puy de Dôme (France). Recently, Carbone et al. (2014) performed a study on long-term measurements of chemical composition in the continental background environment of southern Europe/northern Mediterranean. This study was focused on nocturnal PM_1 chemical composition for 3 years at the high mountain station of Mt. Cimone (Italy).

The Mediterranean region is characterized by particular atmospheric dynamics strongly influenced by its topography (Jorba et al., 2013; Millan et al., 1997). Over this region, elevated emissions of anthropogenic pollutants occur, arrival of natural and anthropogenic aerosols as a result of long-range transport from Africa and Europe is frequent (Pey et al., 2010, 2013b; Ripoll et al., 2014), and accumulation and recirculation processes are recurrently observed (Rodríguez et al., 2002).

For these reasons, results of PM_{10} and PM_1 chemical characterization from Montsec (MSC) and Montseny (MSY) Global Atmosphere Watch (GAW) stations for the period of January 2010–March 2013 are presented in this study. MSC is representative of the continental background conditions of the western Mediterranean Basin (WMB) (Ripoll et al. 2014). This station is located in the FT most of the time due to its elevation (1570 m a.s.l.); although during the warmer months that isolation is broken as a result of vertical mixing and mountain breeze regimes (Ripoll et al. 2014). MSY is a regional background observatory located in the WMB (720 m a.s.l.) in operation since 2002 (Cusack et al., 2012; Pérez et al., 2008a), and it is influenced by regional anthropogenic emissions in specific scenarios (Pérez et al., 2008a; Pey et al., 2010). The record of a relatively long series of PM_{10} and PM_1 concentrations and complete chemical composition at two different WMB environments has allowed for the investigation of temporal and spatial aerosol variations in the WMB with a focus on regional and long-range transport processes. Daily and seasonal patterns of PM_{10} and PM_1 concentrations, as well as their major components and trace elements at MSC and MSY, were investigated. Greater emphasis was placed on the evaluation of the influence of different meteorological scenarios, with a focus on the partitioning of the chemical components into different size fractions in

order to discriminate natural and anthropogenic impacts affecting PM_{10} and PM_1. To the authors' knowledge, no similar studies exist in the literature that compare continental and regional background environments and their seasonal variation.

2 Methodology

2.1 Monitoring sites and sampling schedule

The continental background site was set up in the Montsec (MSC) mountain range, located in the NE of the Iberian Peninsula (42°3′ N, 0°44′ E, 1570 m a.s.l.). This station is situated 50 km to the S of the Axial Pyrenees and 140 km to the NW of Barcelona (Fig. S1). A detailed description of this site can be found in Ripoll et al. (2014).

Results from MSC were jointly studied with those simultaneously obtained at the Montseny (MSY) station, a regional background observatory located in the Montseny Natural Park (41°19′ N, 2°21′ E, 720 m a.s.l.), 40 km to the N-NE of the Barcelona urban area, and 25 km from the Mediterranean coast (Fig. S1) (Pérez et al., 2008a).

At the MSC site, 24 h samples of PM_{10} and PM_1 were collected every 4 days on 150 mm quartz micro-fiber filters (Pallflex QAT) using high-volume samplers ($30\,m^3\,h^{-1}$, MCV CAV-A/MSb) equipped with MCV PM_{10} and PM_1 cut-off inlets. PM_{10} and PM_1 sampling began in November 2009 and in March 2011, respectively. In this work we study the results from January 2010 (March 2011 for PM_1) to March 2013. In addition to the routine measurements, five intensive campaigns (daily sampling) were performed during March–April 2011, July–August 2011, January–February 2012, June–July 2012, and January–February 2013. Overall, 391 and 235 samples of PM_{10} and PM_1, respectively, were collected throughout the study period, and PM_{1-10} concentrations were calculated by the difference of simultaneous PM_1 and PM_{10} daily samples (190 days).

At the MSY site, 24 h samples of PM_{10} and PM_1 were also collected from January 2010 to March 2013 using high-volume samplers ($30\,m^3\,h^{-1}$, DIGITEL-DH80) equipped with PM_{10} and PM_1 cut-off inlet (also DIGITEL). A total of 351 and 335 samples of PM_{10} and PM_1, respectively, were collected during the study period, and PM_{1-10} samples were calculated for 147 days. In most cases, sampling days were coincident at MSC and at MSY.

2.2 Chemical characterization

PM mass concentrations were determined by standard gravimetric procedures, and complete chemical analysis for all filters was performed following the procedures proposed by Querol et al. (2001).

Acid digestion ($HF:HNO_3:HClO_4$) of one-fourth of each filter was carried out to determine and quantify major and trace elements by inductively coupled plasma mass spectrometry (ICP-MS, X Series II, THERMO) and atomic emission spectroscopy (ICP-AES, IRIS Advantage TJA Solutions, THERMO). A few milligrams of the reference material NIST 1633b were added to one-fourth of laboratory blank filters to check the accuracy of the analysis of the acidic digestions. One 1/4 of each filter was leached with ultrapure water (miliQ) to determine the content of Cl^-, SO_4^{2-}, and NO_3^- by ion high-performance liquid chromatography (HPLC) using a WATERS ICpakTM anion column with a WATERS 432 conductivity detector, and NH_4^+ concentrations with a selective electrode (MODEL 710 A+, THERMO Orion). A rectangular portion ($1.5\,cm^{-2}$) of the remaining filter was used for the analysis of organic carbon (OC) and elemental carbon (EC) by a Sunset OCEC analyzer using the EUSAAR 2 protocol (European Supersites for Atmospheric Aerosol Research; Cavalli et al., 2010). Moreover, one blank filter was kept for each set of 10 filters. Blank concentrations were subtracted from the total concentration measured for each sample, thus giving ambient concentrations. To complete mass balances, the following indirect determinations were obtained: (a) CO_3^{2-}, calculated from Ca as $CO_3^{2-} = 1.5 \times Ca$; (b) Al_2O_3, calculated from Al as $Al_2O_3 = 1.889 \times Al$; (c) SiO_2, calculated as $SiO_2 = 2.5 \times Al_2O_3$; and (d) organic matter (OM) obtained by applying a factor of 2.2 to the OC concentrations for MSC samples and a factor of 2.1 for MSY samples, following the suggestion from Takahama et al. (2011). By following these procedures we were able to determine and quantify the concentrations of major components (OC, EC, SiO_2, CO_3^{2-}, Al_2O_3, Ca, Al, Na, Mg, Fe, K, NO_3^-, SO_4^{2-}, NH_4^+, and Cl^-) and trace elements (Li, P, Ti, V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Cd, Sn, Sb, Ba, La, Pb, among others). Overall, the aforementioned components accounted for 60–90 % of the total PM mass. Most of the undetermined mass was attributed to water not eliminated during filter conditioning in the presence of hygroscopic species, but a contribution from sampling artifacts and from the use of factors to determine CO_3^{2-}, SiO_2, and OM cannot be discarded.

At MSC the mineral matter (MM) determination was calculated as

$$MM = CO_3^{2-} + SiO_2 + Al_2O_3 + Ca + Fe + K + \text{nss-Na} + Mg + Mn + Ti + P, \quad (1)$$

where nss-Na is the non-sea-salt sodium; it was calculated as $\text{nss-Na} = Al_2O_3 \times 0.067$ according to the composition of the mineral particles from the Sahara given by Moreno et al. (2006), and hence the remaining sodium was sea-salt sodium ($\text{ss-Na} = Na - \text{nss-Na}$).

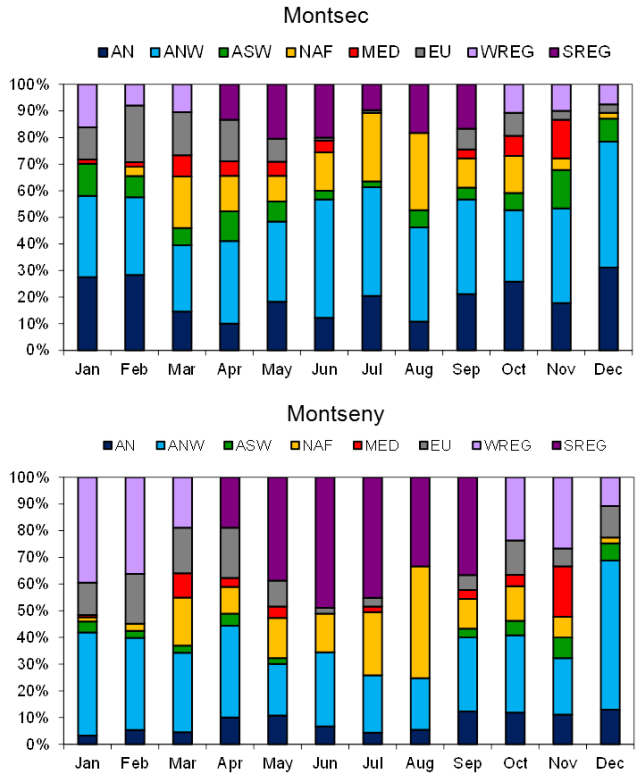

Figure 1. Average frequency of air mass origin at Montsec and Montseny for the different months based on daily calculations between January 2010 and March 2013.

Consequently, the sea salt (SS) determination at MSC was given by

$$\mathrm{SS} = \mathrm{Cl}^- + \text{ss-Na}. \quad (2)$$

At MSY, Na concentrations were entirely attributed to SS, given that it is located closer to the sea and in agreement with Pey et al. (2009).

2.3 Principal component analysis

Principal component analysis (PCA) was performed using the software STATISTICA v10.0. The orthogonal transformation method with varimax rotation (Thurston and Spengler, 1985) was employed, retaining principal components with eigenvalues greater than 1. The data set used for PCA was comprised of the following PM_{10} constituents: Cl^-, NO_3^-, NH_4^+, SO_4^{2-}, Al_2O_3, Ca, K, Na, Mg, Fe, Li, Ti, V, Cr, Mn, Ni, Cu, Zn, As, Se, Sr, Cd, Sb, Pb, OC, and EC. All days with measurements of PM_{10} chemical analysis were included for PCA analysis, which totalled 390 cases from MSC and 351 cases from MSY. A typical robust PCA analysis requires at least a data set with 100 cases. This technique allowed for the identification of main common groups of trace elements in PM_{10} at the continental and regional background sites.

2.4 Classification of atmospheric episodes

The classification of the atmospheric episodes affecting the MSC and MSY sites on each day of the sampling period was performed following the procedure described by Ripoll et al. (2014), and the different air mass transport pathways determined were (1) Atlantic north (AN), (2) Atlantic northwest (ANW), (3) Atlantic southwest (ASW), (4) north Africa (NAF), (5) Mediterranean (MED), (6) Europe (EU), (7) winter regional (WREG, from November to April), and (8) summer regional (SREG, from May to October) (Fig. 1).

Additionally, the boundary layer height was calculated at the MSC and MSY sites using the READY (Real-time Environmental Applications and Display sYstem) model from the NOAA Air Resources Laboratory (http://www.ready.noaa.gov/READYamet.php), which is based in meteorological conditions defining Pasquill stability classes, and uses a resolution grid of 50 km. This was calculated every 3 hours during the whole period (Fig. S2). Despite the limited suitability of this type of model for mountainous terrains, the differences found throughout the year and among different atmospheric scenarios can be considered as a good approximation of the actual PBL variations.

3 Results and discussion

3.1 Continental vs. regional background PM concentrations in the western Mediterranean

PM_{10} and PM_1 average concentrations ($\pm$SD) measured at MSC continental background site reached $11.5 \pm 9.3\ \mu g\ m^{-3}$ and $7.1 \pm 3.9\ \mu g\ m^{-3}$, respectively, whereas at MSY regional background site these concentrations were $15.5 \pm 7.9\ \mu g\ m^{-3}$ and $8.2 \pm 4.1\ \mu g\ m^{-3}$ (Table S1). Thus, the continental-to-regional background increase is estimated to be $4.0\ \mu g\ m^{-3}$ for PM_{10} and $1.1\ \mu g\ m^{-3}$ for PM_1 in the WMB. This increase is caused by differences of (a) altitude, (b) distance to anthropogenic sources, and (c) impact of atmospheric episodes. The contribution of different aerosol chemical components to this increment will be discussed in Sect. 3.2.

A significant seasonal variation of PM_{10} and PM_1 mass concentrations was observed at both sites, with maximum values in summer and minimum values in winter (Fig. 2). Comparable seasonal patterns for PM_X concentrations were described for MSC (Ripoll et al., 2014) and for other regional and continental background sites in southern Europe (e.g. Cozic et al., 2008; Querol et al., 1998; Rodríguez et al., 2003; Tositti et al., 2013). In those studies the seasonal pattern was attributed to changes in the air mass origin from summer to winter, and to the different PBL height between seasons. Moreover, at both MSC and MSY a secondary maximum of PM_{10} and PM_1 concentrations was observed in early spring (Fig. 2).

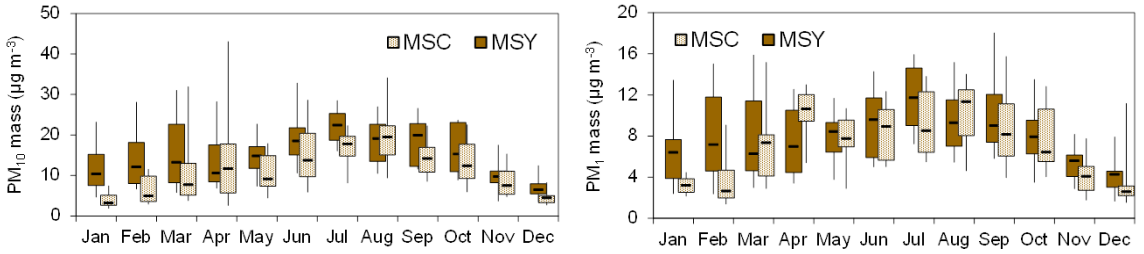

Figure 2. Monthly median (black line within the boxes) and percentiles (5–25–75–95, boxes and whiskers) of daily PM_{10} and PM_1 mass concentrations at Montsec (MSC) and Montseny (MSY) based on daily measurements between January 2010 and March 2013.

PM_{10} concentrations showed a stronger seasonal pattern than PM_1 concentrations at both sites (Fig. 2). This is attributed to the higher impact of resuspended and long-range-transported dust on the PM_{10} fraction, both enhanced in summer (see Sect. 3.3), and to the prevalence of the anthropogenic constituents in PM_1, whose emissions occur throughout the year.

Comparison of these results with those from other continental background sites in central Europe, such as Puy de Dôme at 1465 m a.s.l. in France (Bourcier et al., 2012) and Jungfraujoch at 3454 m a.s.l. in Switzerland (Cozic et al., 2008), shows that PM_{10} and PM_1 concentrations were higher at the continental background site in the WMB (Fig. S3 and Table S1). Such higher PM_{10} and PM_1 concentrations at MSC are related to the increasing role of Saharan dust particles over this area, as discussed in Sect. 3.3 and in agreement with Ripoll et al. (2014), and to the more polluted atmosphere in summer as a result of the air mass recirculation over the WMB (Millan et al., 1997). By contrast, PM_{10} concentrations at the regional background site in the WMB were lower than those at the rural sites in Switzerland (Gianini et al., 2012) (Fig. S3 and Table S1), probably because of the specific Alpine location of these sites which hinders pollution dispersion.

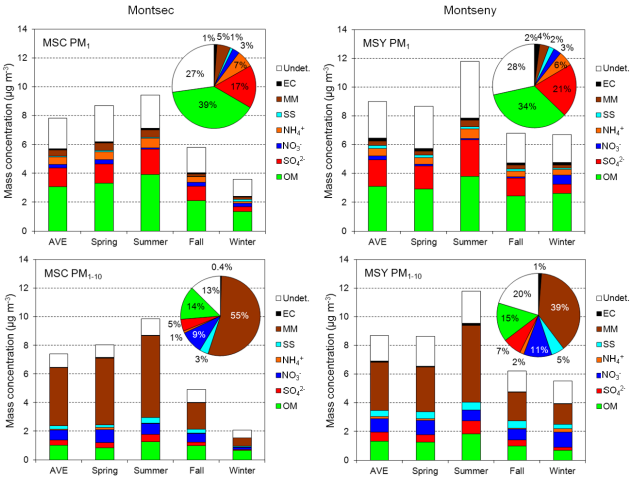

Figure 3. Average concentrations of aerosol major components in PM_1 and PM_{1-10} fractions at Montsec and Montseny for the whole period (AVE) and for different seasons based on daily measurements between January 2010 and March 2013. Pie charts represent the average relative contribution of aerosol major components to total mass for each fraction.

3.2 Continental vs. regional background aerosol chemical composition in the western Mediterranean

3.2.1 Average aerosol chemical composition

PM_1 was mainly composed of OM at both sites (39 % at MSC and 34 % at MSY), followed by sulfate (17 and 21 %), ammonium (7 and 6 %), MM (5 and 4 %), nitrate (3 %), SS (1 and 2 %), and EC (1 and 2 %) (Fig. 3 and Table S1). The undetermined mass accounted for 27 and 28 %. The PM_{1-0} fraction mainly differed in the contribution of MM (55 % at MSC and 39 % at MSY), whereas the other components contributed similarly: OM (14 and 15 %), nitrate (9 and 11 %), sulfate (5 and 7 %), SS (3 and 5 %), ammonium (1 and 2 %), and EC (0.4 and 1 %). The undetermined mass was 20 % at MSY and 13 % at MSC. The closer compositional similarities for PM_1 fraction point to the suitability of using PM_1 as an indicator of regional anthropogenic pollution in Europe, and reflect the wider spatial representativeness of the fine PM.

Overall, average absolute concentrations of chemical components were more similar between MSC and MSY for PM_1 than for the PM_{10} (Fig. 3 and Table S1). The PM_1 continental-to-regional background increase of 1.1 µg m^{-3} is attributed to the higher concentrations of PM_1 sulfate, EC, OM, and some anthropogenic trace elements (V, Ni, Cu, Zn, and Pb) at the regional background site. The increase of 4.0 µg m^{-3} of PM_{10} at MSY with respect to MSC is attributed to higher concentrations of OM, sulfate, nitrate, and SS. These differences confirm that MSC is located at a sufficient altitude and distance from large urban/industrial agglomerations to avoid direct anthropogenic influence and to be considered a continental background site of the WMB. This also confirms that MSY is more affected by the marine aerosols as it is located closer to the coast. Nevertheless, MM and MM-related elements' (Ti, Mn, Li, and Sr) PM_{10} concen-

trations were higher at the continental background site than at the regional background site, probably due to the prevalence of long-range dust transport at higher altitude layers (Sicard et al., 2011), and to the higher dust resuspension at MSC.

In order to provide a global picture of the origin and variability of trace elements in the study region, a principal component analysis (PCA) was performed. This exploration permitted the identification of three main common groups of trace elements in PM_{10} at the continental and regional background sites. Ordered by their contribution to the total mass of trace elements in PM_{10}, these groups were mineral, industrial and road traffic, and fuel-oil-combustion-related elements (Tables S2 and S3).

In the mineral group typical crustal elements (Ti, Mn, Li, and Sr) were included; furthermore, V, Cr, Co, Ni, and As were also partially associated with this factor since these elements, usually attributed to anthropogenic sources, are also found in clay mineral assemblages. The group for which high loading factors were obtained for Cu, Zn, As, Cd, Pb, Sb, and Sn was associated with industrial and road traffic sources, based on previous studies which identified (a) Pb, Zn, Mn, and Cd as tracers of the influence of industrial activities located in the surroundings of Barcelona, such as smelters and cement kilns, and (b) Cu, Sn, and Sb as tracers of non-exhaust vehicle emissions (Amato et al., 2009). These sources could not be split by the PCA probably because these emissions are mixed during their transportation from industrial and urban areas to MSY and MSC. The fuel oil combustion group was better identified at MSY than at MSC, and it was traced by V and Ni. These elements are typical markers of fuel oil combustion, strongly influenced by shipping emissions in the study region (Minguillón et al., 2014; Pey et al., 2013a).

Figures S3 and S4 and Table S1 show average concentrations of chemical components in PM_{10} at the continental background site of Puy de Dôme (Bourcier et al., 2012) and at the rural stations of Payerne and Magadino (Gianini et al., 2012). Nitrate and ammonium PM_{10} concentrations at MSC were slightly higher than those observed at Puy de Dôme, whereas the concentrations registered at MSY were lower than those measured at Payerne and Magadino. The similar sulfate concentrations across Europe in both continental and regional background areas corroborate that this component can remain in the atmosphere for a long time, homogenizing sulfate concentrations in Europe. Average PM_{10} concentrations of EC and OM in the WMB were lower than those measured at Payerne and Magadino, probably due to the higher influence of biomass burning in central Europe. This is confirmed by the higher concentrations of the well-known biomass burning tracer potassium (Pio et al., 2008) registered at the Swiss stations, most of it water soluble (Gianini et al., 2012). Mineral major ($Al + Ca + Mg$) and trace (Ti, Sr, La or Ce) elements at MSC and MSY were recorded in concentrations twice as high as those at Payerne and Magadino, indicating the higher influence of Saharan dust transport and regional dust resuspension in the Mediterranean area. As expected, a higher influence of SS particles was observed at MSC and MSY than at Payerne and Magadino owing to their closer location to the Mediterranean Sea. Conversely, the highest concentrations of typical anthropogenic trace elements – such as Ni, Cu, Zn, As, Cd, Sb, and Pb – were recorded at the Swiss stations, with the exception of V, which was higher at the Spanish sites. This can be partially attributed to a greater influence of emissions from fuel oil combustion, mostly from shipping emissions in the Mediterranean region (Pey et al., 2009).

3.2.2 Partitioning of major and trace components in PM_1 and PM_{1-10}

In the WMB region nitrate showed a prevalent coarse-grain size distribution (Figs. 3 and S5). PM_{1-10} nitrate compounds were partially associated with mineral dust and sea salt particles, since nitric acid and/or some other nitrogen compounds can react with these particles (Wall et al., 1988; Zhuang et al., 1999a). The resulting coarse sodium or calcium nitrate particles are much more stable than ammonium nitrate at high temperature and low humidity (Zhuang et al., 1999b).

As expected, sulfate was mainly fine at both sites (Figs. 3 and S5) and hence attributed to the presence of ammonium sulfate as deduced by the good ionic balance fitting between sulfate and ammonium ($R^2 = 0.899$). Nevertheless, PM_{1-10} sulfate was also detected at MSC and MSY, and it was partially attributed to mineral dust and sea salt particles, and partially attributed to the reaction product between sulfuric acid and/or sulfur dioxide (SO_2) and these natural particles (Wall et al., 1988; Zhuang et al., 1999a).

Ammonium showed a prevalent fine-grain size distribution at the continental and regional background sites (Figs. 3 and S5). Fine ammonium was attributed to the presence of both ammonium nitrate and ammonium sulfate. PM_{1-10} ammonium is most likely in $PM_{1-2.5}$ fraction, according to Querol et al. (2009).

OM was mainly fine at both sites (Figs. 3 and S5). A high number of studies have demonstrated the dominant secondary origin of the fine OM (Jimenez et al., 2009). In the study area, previous works found that secondary organic aerosol (SOA) accounted for 91 and 55 % of the OM at MSY and at the city of Barcelona, respectively (Minguillón et al., 2011; Mohr et al., 2012). Therefore, fine OM at MSC is expected to be dominated by SOA, even more than at MSY given its remote settlement. Furthermore, the presence of OM in PM_{1-10}, especially in spring and summer, suggests the impact of primary bioaerosols (Pöschl et al., 2010).

Although EC was mainly fine at both sites (Figs. 3 and S5), PM_{1-10} EC was also detected, suggesting a partial association between EC and MM by means of adsorption of anthropogenic pollutants onto dust.

As expected, most of the MM species and the mineral trace elements were encountered in the PM_{1-10} fraction, while

the concentrations in PM_1 were clearly lower at both sites (Figs. 3 and S6 and Table S1). Contrary to the rest of MM species, K was also abundant in PM_1 fraction and its concentrations were slightly higher at MSY than those at MSC (Figs. S6 and S7c). This indicates an additional source origin other than mineral (generally as K-feldspar and illite, a K-bearing clay mineral), such as biomass burning, especially over the regional background. The winter DAURE campaign (Determination of the sources of atmospheric Aerosols in Urban and Rural Environments in the western Mediterranean) performed in March 2009 revealed that biomass burning emissions accounted for 33 % of EC and 22 % of OM at MSY (Minguillón et al., 2011). Nevertheless, these contributions are much lower than those obtained at other European regions (Pio et al., 2011).

SS components were found mainly in the PM_{1-10} fraction at both sites (Figs. 3 and S5) as we expected. In the continental background, the lower variation of SS concentrations as a function of atmospheric episodes reflects the minor impact of marine aerosols in this continental background area.

Most of the trace elements showed a prevalent fine-grain size distribution at MSC and MSY (Fig. S8), especially the ones included in the industrial and road traffic group and in the fuel oil combustion group. V, Cr, Co, Ni, and As were also found in the PM_{1-10} fraction because these elements are also present in clay mineral assemblages.

3.3 Atmospheric episodes affecting continental and regional background aerosol chemical composition in the western Mediterranean

The WMB is affected by peculiar atmospheric episodes which influence aerosol chemical composition. At the MSC site, a higher frequency of Atlantic advections was detected, whereas the MSY site was more influenced by regional episodes (Fig. 1). However, the seasonal distribution of the main atmospheric episodes throughout the year is very similar at both sites. The NAF episodes were more frequent from March to October (17 and 18 % of the days, at MSC and MSY, respectively), and very often they alternated with the SREG scenarios (12 and 27 % of the days) or both episodes occurred simultaneously, when the NAF air masses travel at high altitudes and the stagnation of air masses prevails at surface levels (Escudero et al., 2005). The WREG scenarios were detected from October to March (11 and 27 % of the days), as were the EU episodes (11 and 13 % of the days). Air masses from the Atlantic (AN, ANW, and ASW) affected the WMB throughout the year (62 and 41 % of the days). Conversely, the MED episodes were detected sporadically (4 % of the days at both sites), and therefore conclusions on their characteristics will not be drawn in the present study. Moreover, the WMB is affected by sporadic large wildfire events, especially during summer (Cristofanelli et al., 2009).

3.3.1 North African episodes (NAF)

The non-NAF-to-NAF increase in PM_1 concentrations is estimated to be 2.9 $\mu g\,m^{-3}$ and 4.6 $\mu g\,m^{-3}$ at MSC and MSY, respectively, whereas this increase in PM_{1-10} concentrations is estimated to be 14.3 $\mu g\,m^{-3}$ and 7 $\mu g\,m^{-3}$ (Figs. 4 and 5). The PM_1 non-NAF-to-NAF increase was attributed to the increment of PM_1 MM, sulfate, nitrate, ammonium, OM, and EC (Figs. S5 and S6). In relative contribution the highest difference in PM_1 concentrations was recorded for MM at MSC (Fig. 4), thus evidencing that NAF episodes also affect the fine fraction.

On the other hand, the higher concentrations of PM_{1-10} MM under NAF episodes accounted for the non-NAF-to-NAF increase of PM_{1-10} concentrations. Additionally, average concentrations of trace elements from the mineral group were higher at the continental background site than at the regional background site under NAF episodes (Fig. S6). Concentrations of nitrate and sulfate were high during NAF episodes at both sites when compared with the average (Figs. S5 and S7b). At MSY concentrations of coarse ammonium and EC also increased.

The higher impact of NAF scenarios on the continental than on the regional background aerosols in the WMB confirms that African dust travels preferentially at high altitudes. The concurrent increase of secondary pollutants (nitrate and sulfate) at MSC demonstrates that dust arrives together with industrial pollutants, as shown at the Canary Islands by Rodríguez et al. (2011). The relatively high concentrations of secondary pollutants and EC during NAF at MSY in PM_1 and PM_{1-10} can be related to the interaction of dust with anthropogenic pollutants. During NAF episodes a compression of the PBL is observed at regional scale (Alastuey et al., 2005; Pandolfi et al., 2013) (Fig. S9), and a dominance of southern winds during the whole day breaks the regular sea breeze circulation (Jorba et al., 2013). These processes enhance the concentration of regional pollutants in the lowest part of the troposphere and inhibit the sea breeze "clean-up" effect.

The net contribution of African dust to the PM_{10} concentrations was estimated to be 16 % at MSC and 11 % at MSY. This is in agreement with results presented by Pey et al. (2013b).

An example of the impact of NAF episodes on the continental and regional background aerosols was recorded on 26 March 2011 (Fig. 5b). Backward trajectory for this day clearly showed a North African air mass origin (Fig. S10), and the Navy Aerosol Analysis and Prediction System (NAAPS) indicated high dust surface concentrations over the Iberian Peninsula (Fig. S11). During this episode, dust transport more strongly affected MSC than MSY, with PM_{10} MM concentration reaching 16 $\mu g\,m^{-3}$ at MSC (more than 5 times higher than the annual average) and 9 $\mu g\,m^{-3}$ at MSY (about 3 times higher than the annual average). At MSC, concentrations of MM increased simultaneously with those of sulfate

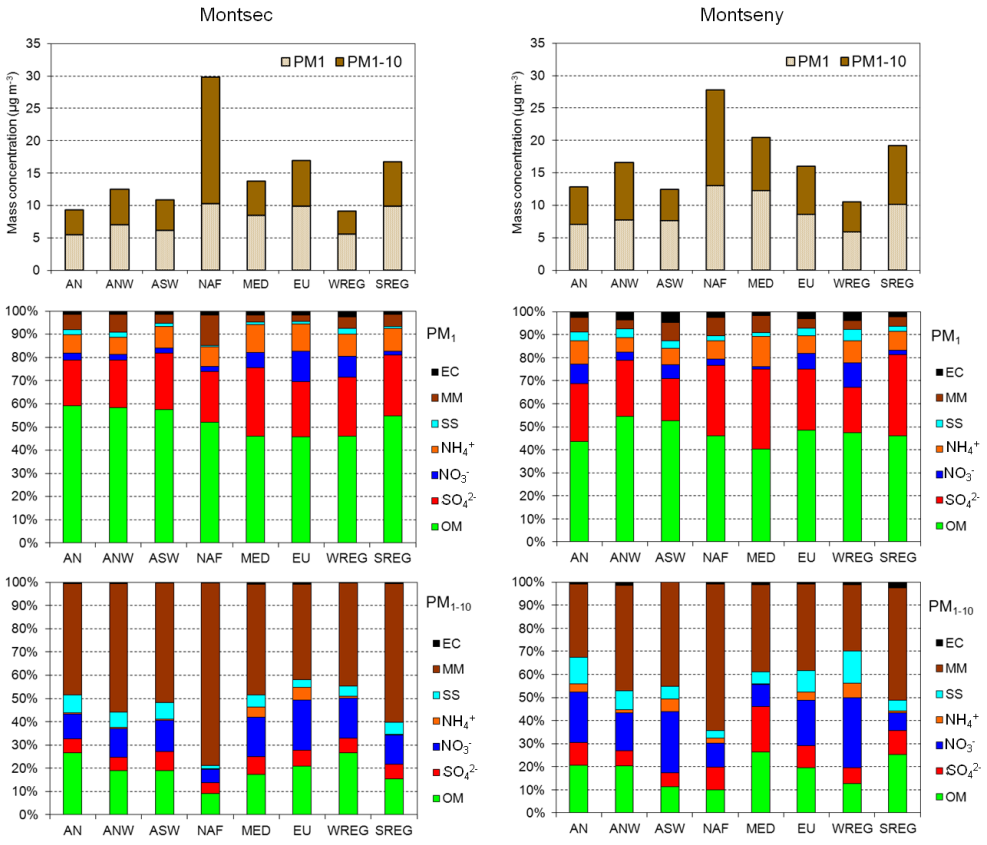

Figure 4. Average concentrations of PM_1 and PM_{1-10} mass and relative contribution of aerosol major components in PM_1 and PM_{1-10} fractions for different meteorological episodes at Montsec and Montseny based on daily measurements between January 2010 and March 2013.

(2 times higher than the annual average), nitrate (3 times), and other anthropogenic elements, such as Sb and EC, reflecting that dust is transported together with these pollutants. The increments of absolute concentrations of nitrate, sulfate, Sb, and EC were higher at MSY than MSC, probably due to the aforementioned effect of both the PBL compression and the breeze.

3.3.2 Summer regional episodes (SREG)

The importance of the SREG scenarios in the WMB has been studied in a number of works (e.g. Escudero et al., 2005; Gangoiti et al., 2001; Millan et al., 1997; Rodríguez et al., 2002). These episodes take place under a weak barometric gradient and a lack of advections in summer, which causes the recirculation of air masses over the WMB. Generally, these situations last for several days, favoring the accumulation of pollutants at regional scale, increasing the magnitude of convection and aging processes, and enhancing the dust resuspension and the formation of secondary organic and inorganic aerosols. For these reasons, under SREG episodes high concentrations of PM_1 and PM_{1-10} were measured at the regional background site (Fig. 4). The PM_1 increase was attributed to higher concentrations of PM_1 sulfate, OM, EC, and trace elements of industrial and road traffic group, and the PM_{1-10} increase was attributed to higher concentrations of PM_{1-10} MM, OM, and EC (Figs. S5, S6 and S8).

In spite of the high altitude of MSC, the continental background site was also affected by this type of episodes due to a higher development of the PBL over the continental areas (Fig. S9), which favors the transport of anthropogenic pollutants towards high-altitude sites such as MSC and enhances the dust resuspension (Figs. 4, S5, S6 and S8).

High concentrations of elements such as Ca, Mg, Sr, and Mn during SREG scenarios (Fig. S6) confirm the local/regional origin of these dust particles at both sites. However, the ratio of concentrations of these specific elements with respect to Al during SREG scenarios was higher at MSC compared to MSY, probably because of the calcareous (richer in Ca, Mg, Sr, and Mn) nature of the Montsec Range as opposed to the slate and granitic composition (richer in Al) of the Montseny Range.

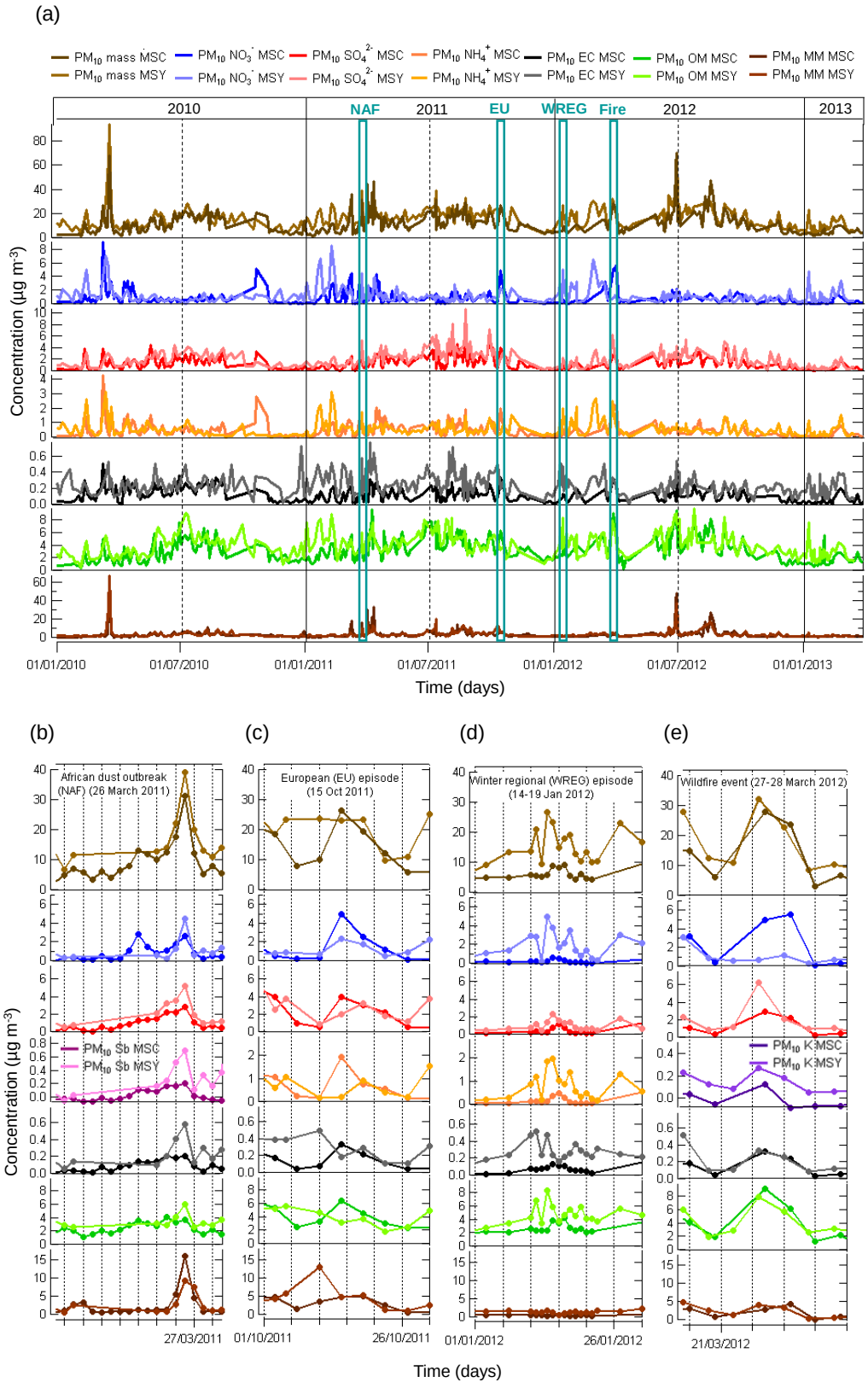

Figure 5. (a) Time series of daily PM_{10} mass and major PM_{10} chemical components concentrations at Montsec (MSC) and Montseny (MSY) between January 2010 and March 2013. Green bands indicate four examples of different episodes affecting the study area. Zoom of the four selected meteorological episodes – **(b)** African dust outbreak, **(c)** European episode, **(d)** winter regional episode, and **(e)** wildfire event – with daily PM_{10} mass and PM_{10} chemical components concentrations.

3.3.3 Winter regional episodes (WREG)

The WREG episodes affecting the WMB have been described by Pey et al. (2010) as winter anticyclonic episodes (WAEs), based on a comparison between urban and regional background sites. In the present study the WREG episodes showed low concentrations of PM_1 and PM_{1-10} on average (Fig. 4), but the chemical composition revealed the higher relative contribution of anthropogenic sources and the lower relative proportion of natural emissions, with high concentrations of PM_1 nitrate, ammonium, OM, and EC and the lowest concentrations of PM_{1-10} OM and MM, especially at MSY (Figs. S5, S6 and S7a).

The impact of these episodes on the continental and regional background aerosols can be explained by the persistence of anticyclonic conditions in the WMB. During these situations the stagnation of air masses occurs, which increases the pollution around the emission sources (mainly urban and industrial areas); therefore regional and continental background areas are presumably free of this anthropogenic pollution. However, the meteorological conditions (sunny days and thermal inversion) together with the topography of the WMB (very mountainous) may develop mountain breezes. In this case, the anthropogenic pollution accumulated over the adjacent valleys reaches regional background sites and rarely continental background areas as intense pollution episodes.

Figure 5d shows an example of a WREG episode affecting the WMB during the period 14–19 January 2012. Backward trajectory of air masses for these days corroborates a WREG situation (Fig. S10). During this episode PM_{10} nitrate, ammonium, EC, and OM concentrations at the regional background site increased from 0.2 $\mu g\,m^{-3}$, 0.2 $\mu g\,m^{-3}$, 0.2 $\mu g\,m^{-3}$, and 3.4 $\mu g\,m^{-3}$ on 13 January to 1.3–5.0 $\mu g\,m^{-3}$, 0.6–1.9 $\mu g\,m^{-3}$, 0.2–0.5 $\mu g\,m^{-3}$, and 3.7–8.4 $\mu g\,m^{-3}$ between 14 and19 January 2012, respectively. Simultaneously, the continental background site was almost unaffected by such polluted air masses since nitrate, ammonium, EC, and OM concentrations in PM_{10} remained 0.1–0.6 $\mu g\,m^{-3}$, 0.1–0.5 $\mu g\,m^{-3}$, 0.05–0.13 $\mu g\,m^{-3}$, and 2.3–4.2 $\mu g\,m^{-3}$, respectively.

3.3.4 European episodes (EU)

During EU episodes air masses from central and eastern Europe are transported towards the WMB, crossing the whole continent. This type of episode is associated with cold meteorological conditions and polluted air masses (Pey et al., 2010). For this reason, under EU episodes high concentrations of PM_1 and PM_{1-10} were measured at both sites (Fig. 4). This increase was attributed to higher concentrations of nitrate, ammonium, OM, EC, and trace elements of industrial and road traffic group at both fractions PM_1 and PM_{1-10}, especially at MSC (Figs. S5 and S8).

The higher impact of EU scenarios on the continental background aerosols than on the regional background aerosols confirms that the transport of pollutants occurs preferentially at high altitude layers (915–1930 m), as observed by Sicard et al. (2011).

An example of the impact of EU episodes on the continental and regional background aerosols was recorded on 15 October 2011 (Fig. 5c). During this episode the air mass remained 3 days over central Europe before reaching the WMB, as shown by the backward trajectory (Fig. S4). Under these meteorological conditions PM_{10} nitrate, sulfate, ammonium, EC, and OM daily concentration at MSC reached 5 $\mu g\,m^{-3}$ (6 times higher than the annual average), 4 $\mu g\,m^{-3}$ (about 3 times higher than the annual average), 2 $\mu g\,m^{-3}$ (more than 4 times higher than the annual average), 0.3 $\mu g\,m^{-3}$ (almost 3 times higher than the annual average), and 6.4 $\mu g\,m^{-3}$ (about 2 times higher than the annual average), respectively. Simultaneously, the regional background site was less affected by this episode as PM_{10} nitrate, sulfate, ammonium, EC, and OM concentrations at MSY were 2.3 $\mu g\,m^{-3}$ (1 $\mu g\,m^{-3}$ higher than the annual average), 2.0 $\mu g\,m^{-3}$, 0.2 $\mu g\,m^{-3}$, 0.2 $\mu g\,m^{-3}$, and 3.2 $\mu g\,m^{-3}$, respectively.

3.3.5 Atlantic advections (AN, ANW, ASW)

The WMB is strongly affected by oceanic air masses owing to the large influence of Atlantic winds over this region, which is favored by the displacement of the Azores High westwards or southwards, allowing the movement of depression systems towards the Mediterranean (Lopez-Bustins et al., 2008). This type of episodes is associated with increased precipitation and intense winds, which favor the atmospheric wet-scavenging processes, leading to the renovation of the regional aged air masses.

These meteorological conditions explain why low PM_1 and PM_{1-10} concentrations were measured under Atlantic advections at both sites (Fig. 4) and for most of the aerosol chemical components (Figs. S5, S6, and S8).

3.3.6 Wildfire events

Although biomass burning is not a major source in the WMB, some contribution of biomass burning is observed and partially attributed to wildfires. Therefore, high concentrations of PM_1 OM and EC during NAF and SREG episodes (Fig. S5) were partially attributed to a higher frequency of wildfires, since these episodes very often occur simultaneously.

Figure 5e illustrates the impact of a wildfire on continental and regional background aerosols in the WMB. This event took place in eastern Europe on 27–28 March 2012, as shown by the NAAPS model with high smoke surface concentrations over SE Europe (Fig. S11). Backward trajectory for these days clearly showed a transport of air masses

from SE Europe towards the WMB (Fig. S10). During this episode, daily PM_{10} OM and EC concentration at MSY reached 7.9 $\mu g\,m^{-3}$ (almost 2 times higher than the annual average) and 0.3 $\mu g\,m^{-3}$ (1.5 times higher than the annual average), respectively, and at MSC it was 9.1 $\mu g\,m^{-3}$ (almost 3 times higher than the annual average) and 0.3 $\mu g\,m^{-3}$ (3 times higher than the annual average). Moreover, K concentrations in PM_{10} at MSY reached 0.27 $\mu g\,m^{-3}$ (2 times higher than the annual average), with a prevalent fine partitioning ($PM_1/PM_{10} = 0.55$), instead of its main coarse occurrence ($PM_1/PM_{10} = 0.28$) during NAF episodes, which confirms the biomass burning origin.

3.4 Seasonal variation of continental and regional background aerosol chemical composition in the western Mediterranean

In addition to the atmospheric episodes discussed above, the aerosols in the WMB are also affected by the different evolution of the PBL height and differences in the meteorological parameters throughout the year. In the warmer months (April–September) the development of the PBL at MSC is much more relevant than that at MSY (Figs. S2 and S9), owing to the higher convection at the continental background sites (Rodríguez et al., 2002) and to the greater cooling effect from the sea breeze at the regional background sites. On the other hand, in the colder months (October–March) the lower vertical development of the PBL at the inland sites leaves MSC in the FT on most days, whereas the regional background site is located most of the day within the PBL (Fig. S2).

Nitrate concentrations decreased in summer at both sites, especially in PM_1 (Fig. 6) (2 and 3 times lower than the winter concentrations at MSC and MSY, respectively). This decrease was attributed to the high volatility of ammonium nitrate (Pey et al., 2009) at low humidity and high temperature (Zhuang et al., 1999b). During the colder months higher nitrate concentrations are associated with WREG episodes at MSY and with EU episodes at MSC, with the exception of the November-to-January period, when MSC is mostly within the FT and therefore low nitrate concentrations were registered.

Sulfate showed similar seasonal variations at both sites and relatively similar absolute concentrations, with the highest values during the warmer months (3 and 2 times higher than the winter concentrations at MSC and MSY, respectively) (Fig. 6). The squared Pearson correlation coefficient between the daily sulfate concentrations at MSC and MSY was 0.71. This similarity reflects the high stability of sulfate and its longer residence time in the atmosphere, resulting in a homogeneous sulfate concentration in the WMB. The summer maximum was likely due to the higher photochemistry in the atmosphere that enhances the SO_2 oxidation (6 % h^{-1} in summer vs. < 1 % h^{-1} in winter; Querol et al., 1999) and to the accumulation of pollutants over the WMB as a result of the SREG and NAF episodes. Additionally, regional background sulfate aerosols in summer could be affected by the transport of shipping emissions from the Mediterranean to the continental areas, due to the more intense sea breeze circulation at MSY. The lower concentrations during the colder months were attributed to the lower rate of SO_2 oxidation (Querol et al., 1999), and at MSC to the FT conditions.

Ammonium concentration did not follow a clear seasonal pattern (Fig. 6) due to its association with both sulfate and nitrate.

Organic matter concentrations followed a similar seasonal variation at both sites, with the highest values during the warmer months (1.8 and 1.5 times higher than the winter concentrations at MSC and MSY, respectively) (Fig. 6). The summer maximum was due to (1) the higher temperature and photochemistry in the atmosphere that enhances the formation of SOA; (2) the accumulation of pollutants over the WMB owing to the occurrence of SREG and NAF episodes; (3) the greater biogenic emissions from vegetation (Seco et al., 2011); and (4) the higher frequency of wildfires. Furthermore, at MSY a secondary maximum of OM concentrations occurred in October–March, linked to the occurrence of WREG episodes. The continental background site was less affected by this type of episodes, since MSC is mostly within the FT in winter.

Elemental carbon showed high concentrations in the warmer months at MSC and a less marked seasonal variation at MSY (Fig. 6). The smoother seasonal variation at MSY reflects the regional anthropogenic influence on the levels of this component, since anthropogenic emissions occur throughout the year. The higher summer EC concentrations at both sites (more elevated at MSY) were attributed to the impact of the SREG and NAF episodes, and to the higher occurrence of wildfires. Additionally, at MSY high concentrations of EC in summer may be caused by the greater transport of shipping emissions. The increase of EC during the colder months was not registered simultaneously at both environments (Fig. 6) because at the regional background site it was attributed to the impact of WREG episodes, whereas at the continental background site it was associated with EU episodes.

Mineral matter concentrations and mineral trace elements in the WMB are driven by the local and regional dust resuspension and by the contribution of African dust outbreaks, both enhanced in the warmer months. Consequently, the highest values were measured in summer and the lowest in winter, with sporadic high concentrations in March–April (Figs. 6 and 7).

Trace elements concentrations of the industrial and road traffic group showed low variations, especially at MSY (Fig. 7), since anthropogenic emissions occur throughout the year. Fuel oil combustion elements showed a marked seasonal pattern at both sites, with the highest values in summer (Fig. 7) due to the higher shipping emissions and the more

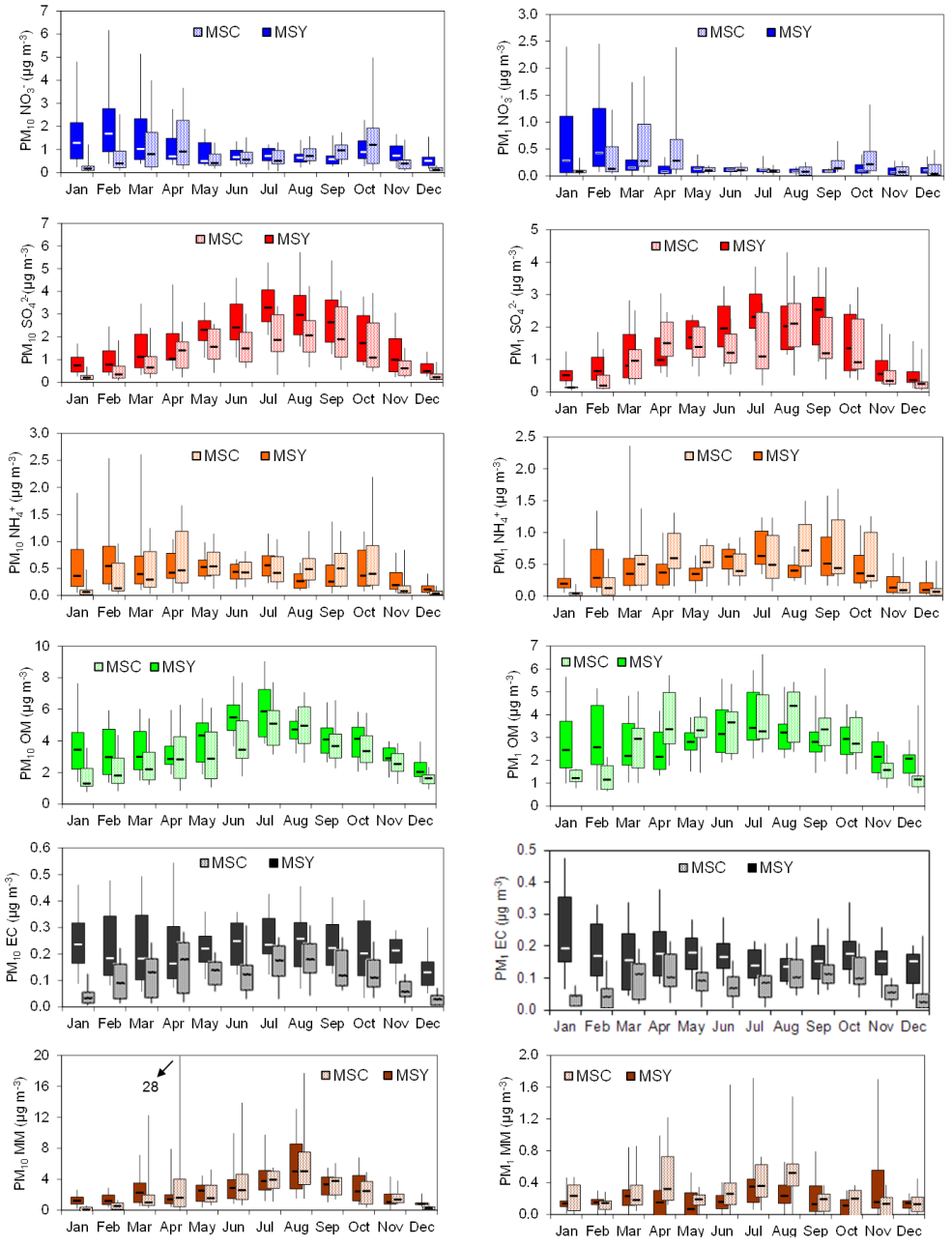

Figure 6. Monthly median (black line within the boxes) and percentiles (5–25–75–95, boxes and whiskers) of major PM_{10} and PM_1 chemical components concentrations at Montsec (MSC) and Montseny (MSY) based on daily measurements between January 2010 and March 2013.

frequent and intense sea breeze circulation, which enhances the transport of air masses from the Mediterranean Sea.

4 Conclusions

Aerosol chemical characterization (PM_1, PM_{1-10}, and PM_{10}) and its time variation were studied during January 2010–March 2013 simultaneously at a continental (Montsec, MSC) and a regional (Montseny, MSY) background site in the western Mediterranean Basin.

In this particular region of the WMB, the continental-to-regional background increase was estimated to be 4.0 µg m^{-3} for PM_{10} and 1.1 µg m^{-3} for PM_1. Relative chemical composition and absolute concentrations of PM_X showed very similar values at both environments, especially in PM_1, in spite of their altitudinal and longitudinal differences. The similarities are more pronounced in the warmer months, when recirculation processes at a regional scale are recurrent in the WMB,

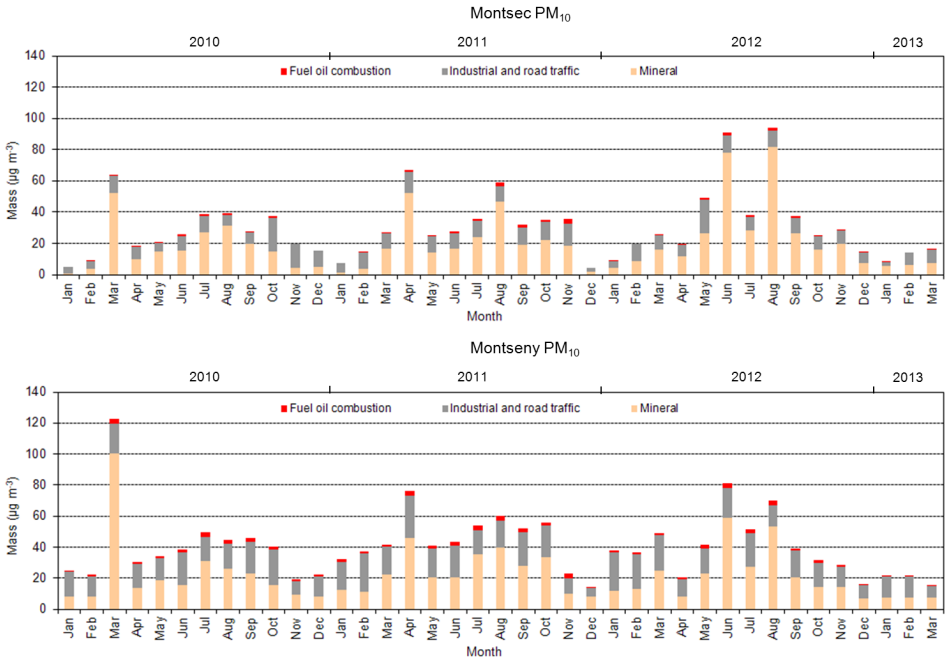

Figure 7. Monthly average concentrations of trace element groups at Montsec and Montseny based on daily measurements of PM_{10} between January 2010 and March 2013.

and a strong development of the PBL occurs over continental areas, favoring the transport of anthropogenic pollutants towards remote sites such as MSC. These processes cause a homogenization of PM_1 concentration and composition throughout the region, allowing us to consider PM_1 as a more suitable indicator of anthropogenic impact than PM_{10}. Moreover, the higher temperature and solar radiation in the warmer months augment atmospheric photochemistry, promoting the formation of secondary inorganic and organic aerosols, and thus incrementing markedly the concentration of certain components such as sulfate and OM. Additionally, sea breeze circulation is enhanced, favoring the transport of shipping emissions from the Mediterranean to the continental areas and thereby increasing the concentrations of sulfate, EC, and fuel-oil-combustion-related trace elements, especially at the regional background since it is located closer to the coast. Furthermore, the occurrence of wildfires across the WMB increases in summer, which contributes to an extra increment of the OM and EC concentrations. Conversely, nitrate is not abundant in summer due to the high volatility of ammonium nitrate at high temperatures and low humidity. In the colder months the lower vertical development of the PBL leaves MSC in the FT on most days, whereas MSY is frequently located within the PBL due to its lower elevation. As a result, very low concentrations of all chemical components are recorded at MSC in winter, while MSY is regularly affected by nearby polluted air masses, which enhanced the concentrations of PM_X components.

The seasonal variation of major and trace PM_X components was also governed by changes in the air mass origin from summer to winter. Whereas southern flows and regional recirculation episodes are more frequent in summer, Atlantic advections and northeastern winds from mainland Europe are more common in winter. As a result, African dust outbreaks and regional dust resuspension increase MM concentrations over the WMB in the warmer months. This MM increase affects both PM_1 and PM_{1-10}, and it is frequently more pronounced at MSC, since long-range transport of dust occurs preferentially at high altitude layers and dust resuspension is enhanced by the drier surface and higher convection at this site. During NAF episodes concentrations of nitrate and sulfate also increase, demonstrating that dust arrives together with industrial pollutants. Moreover, a compression of the PBL and a change in the wind regime towards a permanent southern flow increase the concentrations of regional pollutants (sulfate, EC, and industrial and traffic tracers) in the lowest part of the troposphere. Regional recirculation of air masses (SREG episodes) also accounts for the accumulation of airborne particulates, increasing the concentrations of sulfate, OM, EC, industrial, traffic, and fuel oil combustion tracers at both continental and regional background environments.

In the colder months, the predominance of clean Atlantic advections prevents the accumulation of regional pollution and consequently reduces the concentration of all chemical components at both sites. However, the sporadic transport of polluted air masses from central and eastern Europe towards

the WMB increases the concentrations of nitrate, OM, EC, and industrial and traffic-related trace elements. The impact of these polluted air masses on the concentrations of PM_X components is usually higher in the continental background, since this transport from Europe occurs preferentially at high altitude layers. Occasionally, intense peaks of nitrate, OM, and EC are measured at the regional background site during the winter anticyclonic episodes (WREG). These stagnant situations cause the accumulation of pollutants around the emission sources (such as the Barcelona metropolitan area), and pollutants can be transported towards relatively nearby areas under favorable conditions. The distance from MSC to large anthropogenic sources and its altitude are restricting factors for the occurrence of this process.

Finally, the comparison of these results with those from other continental and regional background sites in central Europe shows that African dust transport and regional dust resuspension are much more important in the western Mediterranean area. The net contribution of African dust to the PM_{10} concentrations was estimated to be 16 % at MSC and 11 % at MSY. This is reflected in more elevated concentrations of mineral elements across the Mediterranean, with the only exception being potassium, higher in central Europe due to the contribution of biomass burning emissions. The surprising similar sulfate concentrations across Europe in both continental and regional background environments are probably linked to the long residence time of sulfate aerosols in the atmosphere. However, nitrate and ammonium showed different concentrations as a function of site, and nitrate maximum concentration was observed in winter in the WMB, whereas in the central Europe continental environments it was measured in summer. Moreover, the highest concentrations of typical anthropogenic trace elements were recorded at some European rural environments, with the exception of V, which was higher in the Mediterranean area due to the greater influence of shipping emissions.

The concurrent monitoring of aerosol properties in continental and regional background sites in the WMB provides a complete picture of the aerosol phenomenology of this region. In view of the relatively high concentrations of atmospheric aerosols from a variety of natural and anthropogenic sources, and taking into account the importance of atmospheric processes, the simultaneous characterization of atmospheric aerosols at continental and regional background sites provides valuable information to policy makers and air quality and climate models.

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