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Joint analysis of continental and regional background environments in the Western Mediterranean: PM₁ and PM₁₀ concentrations and composition

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Abstract

The complete chemical composition of atmospheric particulate matter (PM₁ and PM₁₀) 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 for a relatively long-term series (January 2010–March 2013).

Differences on average PM concentration and composition and on seasonal variation between both sites were attributed to: distance to anthropogenic sources, altitude, height evolution of the planetary boundary layer (PBL) throughout the year, variations in the air mass origin, and changes in meteorology.

The diverse meteorological episodes showed different influence at regional and continental scale. 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 at high altitude layers. Conversely, the regional background was more influenced by regional processes. In winter, anticyclonic conditions enhance the stagnation of air masses with the consequent accumulation of pollutants at regional scale in the WMB, whereas the continental background site remains in the free troposphere. Totally different conditions drive the aerosol phenomenology in summer. Weak pressure gradients and elevated insolation generate recirculation of air masses and enhances the development of the PBL, causing the aging of aerosols and incrementing pollutant concentrations over a larger area, including the continental background. This is reflected in a more similar relative composition and absolute concentrations of PM components at both regional and continental background environments in the warmer season

Peculiarities of the WMB are: (a) high relevance of African dust transport and regional dust resuspension; (b) low biomass burning contribution; (c) low summer nitrate concentrations; and (d) high aerosol homogenization in summer.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



1 Introduction

The influence of atmospheric particulate matter (PM) on the Earth's radiative budget is of keen current 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 may also cause 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 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 define 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.

Despite there is not an established definition, continental background environments can be considered 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 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

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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 they mostly correspond to 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 is 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.

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 Jungfrauoch (Switzerland). Bourcier et al. (2012) studied PM_{10} and PM_1 water-soluble inorganic components over one 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 the 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

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

frequent (Pey et al., 2010, 2013b; Ripoll et al., 2014), and accumulation and recirculation processes are recurrently observed (Rodriguez et al., 2002).

For these reasons, results of PM₁₀ and PM₁ chemical characterization from Montsec (MSC) and Montseny (MSY) 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., 2008) and it is influenced by regional anthropogenic emissions in specific scenarios (Pérez et al., 2008; Pey et al., 2010). The record of a relatively long series of PM₁₀ and PM₁ 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 focus on regional and long-range transport processes. Daily and seasonal patterns of PM₁₀ and PM₁ 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 PM₁₀ and PM₁. To the authors' knowledge, no similar studies exist in the literature which compares 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

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



is situated at 50 km to the S of the Axial Pyrenees and at 140 km to the NW of Barcelona (Supplement 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, 02°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 (Supplement Fig. S1) (Pérez et al., 2008).

At MSC site, 24 h samples of PM₁₀ and PM₁ were collected every 4 days on 150 mm quartz micro-fiber filters (Pallflex QAT) using high volume samplers (30 m³ h⁻¹, MCV CAV-A/MSb) equipped with MCV PM₁₀ and PM₁ cut-off inlets. PM₁₀PM₁ sampling began in November 2009 and in March 2011, respectively. In this work we study the results from January 2010 (March 2011 for PM₁) to March 2013 are provided. In addition to the routine measurements, 5 intensive campaigns (daily sampling) were performed during March–April 2011, July–August 2011, January–February 2012, June–July 2012 and January–February 2013. A detailed sampling schedule is shown in Supplement Fig. S2. Overall, 391 and 235 samples of PM₁₀ and PM₁, respectively, were collected throughout the study period, and PM₁₋₁₀ concentrations were calculated by the difference of simultaneous PM₁ and PM₁₀ samples (190 samples).

At MSY site, 24 h samples of PM₁₀ and PM₁ were also collected from January 2010 to March 2013 using high volume samplers (30 m³ h⁻¹, DIGITEL-DH80) equipped with PM₁₀ and PM₁ cut-off inlet (also DIGITEL). A total of 351 and 335 samples of PM₁₀ and PM₁, respectively, were collected during the study period, and PM₁₋₁₀ samples were calculated for 147 days. In most cases, sampling days were coincident at MSC and at MSY.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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).

5 Acid digestion ($\text{HF} : \text{HNO}_3 : \text{HClO}_4$) of 1/4 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). Few mg of the reference material NIST 1633b were added to 1/4 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 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 EU-SAAR 2 protocol (Cavalli et al., 2010). Moreover, one blank filter was kept for each set of ten 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
15 Ca as $\text{CO}_3^{2-} = 1.5 \times \text{Ca}$; (b) Al_2O_3 , calculated from Al as $\text{Al}_2\text{O}_3 = 1.889 \times \text{Al}$; (c) SiO_2 , calculated as $\text{SiO}_2 = 2.5 \times \text{Al}_2\text{O}_3$, and (d) organic matter (OM) obtained applying a 2.2
20 factor to the OC concentrations following the suggestion for a high-elevation mountain site 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-} ,
25 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.

**Aerosols in
continental and
regional background
in the Western
Mediterranean**

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



At MSC the mineral matter (MM) determination was given by:

$$\text{MM} = \text{CO}_3^{2-} + \text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Ca} + \text{Fe} + \text{K} + \text{nss-Na} + \text{Mg} + \text{Mn} + \text{Ti} + \text{P} \quad (1)$$

Where nss-Na is the non-sea-salt sodium and it was calculated as $\text{nss-Na} = \text{Al}_2\text{O}_3 \times 0.067$ 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 ($\text{ss-Na} = \text{Na} - \text{nss-Na}$).

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

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

At MSY, Na concentrations were totally attributed to SS, given that it is located closer to the sea and therefore nss-Na concentrations are negligible.

2.3 Classification of meteorological episodes

The classification of the atmospheric episodes affecting 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 North West (ANW), (3) Atlantic South West (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).

Additionally, the boundary layer height was calculated at MSC and MSY sites using the HYSPLIT model from the NOAA Air Resources Laboratory (<http://www.ready.noaa.gov/READYamet.php>), which uses the information for stability time series. This was calculated every three hours during the whole period (Supplement Fig. S3).

3 Results and discussion

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

PM₁₀ and PM₁ average concentrations measured at MSC continental background reached 11.5 µg m⁻³ (January 2010–March 2013) and 7.1 µg m⁻³ (March 2011–March 2013), respectively, whereas at MSY regional background these concentrations were 15.5 and 8.2 µg m⁻³ (January 2010–March 2013) (Supplement Table S1). Thus, the continental to regional background increase is estimated in 4.0 µg m⁻³ for PM₁₀ and 1.1 µg m⁻³ for PM₁ in the WMB. The differences between both sites were caused by differences of: (a) altitude, (b) distance to anthropogenic sources, and (c) evolution of the PBL height throughout the year at both sites (Supplement Fig. S3).

In the colder months the lower vertical development of the PBL leaves the continental background site in the free troposphere (FT) on most days, due to its higher altitude (Supplement Fig. S3), and therefore very low PM concentrations were recorded at MSC. Conversely, during these months the regional background site is located most of the day within the PBL, and is more affected by polluted air masses, which enhanced the PM concentrations.

In the warmer months the development of the PBL at the continental background site is much more important than that at the regional background site (Supplement Fig. S3) owing to the higher convection at MSC, caused by: (a) the less cooling effect from the sea breeze at this inland site, and (b) the less forested areas than at MSY. The higher PBL development at MSC favors the dilution of aerosols, and therefore PM concentrations measured at MSC were lower than those recorded at MSY.

Despite the different effect of atmospheric dynamics throughout the year, both sites showed similar seasonal variation of PM concentrations, with maximum values in summer and minimum in winter (Ripoll et al., 2014). This has been concluded for different regional and continental background sites (e.g. Cozic et al., 2008; Querol et al., 1998;

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Rodríguez et al., 2003; Tositti et al., 2013) and it has been attributed to changes in the air mass origin from summer to winter, and to the different PBL height between seasons. However, at MSY a secondary maximum was observed in January–March.

A previous study in the WMB (Ripoll et al., 2014) has demonstrated that PM₁₀ concentrations at MSC showed a strong seasonal pattern driven by the transport of African dust and by the contribution of regional dust resuspension, both enhanced in summer. However, PM₁ showed a weaker seasonal variation but a better defined weekly pattern driven by the regional anthropogenic emissions. Hourly PM variations have been also studied for each month. In the continental background, summer and winter present different behaviors. In the warmer months, long-range transport of aerosols and summer recirculation processes over the WMB mask the diurnal cycles of PM produced by the mountain breezes. In the colder months, the background PM concentrations at MSC are extremely reduced and the PM daily variations are strongly driven by the influence of mountain breezes. By contrast, PM concentrations at the regional background showed clear daily patterns independently of the season, because MSY is located closer to pollution sources and it is more influenced by sea breeze regimes (Cusack et al., 2013; Pérez et al., 2008).

PM concentrations measured in the WMB continental background were higher than those obtained at other continental background sites in Central Europe, such as Puy de Dôme at 1465 m a.s.l. in France (Bourcier et al., 2012) (Supplement Table S1) and Jungfraujoch at 3454 m a.s.l. in Switzerland (Cozic et al., 2008). Such higher PM concentrations at MSC are related to the important impact of Saharan dust particles over this area (Ripoll et al., 2014); and to the more polluted atmosphere in the warmer months as a result of the summer recirculation of air masses over the WMB (Millan et al., 1997). By contrast, PM concentrations reported in the WMB regional background were lower than those reported at the rural stations of Payerne and Magadino in Switzerland (Gianini et al., 2012) (Supplement Table S1), probably because of the specific alpine location of these sites which does not favor pollution dispersion.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3.2 Continental vs. regional background aerosols chemical composition in the Western Mediterranean

On average, MSC and MSY showed very similar relative average PM chemical composition (Supplement Fig. S4). OM was the foremost PM₁ constituent at both sites (34–39%), followed by sulfate (17–21%), ammonium (6–7%), MM (4–5%), nitrate (3%), SS (1–2%), and EC (1–2%). The undetermined mass ranged between 27 and 28%. The PM_{1–10} fraction was mainly made up of MM (39–53%), OM (14–15%), nitrate (9–11%), sulfate (5–7%), SS (4–5%), ammonium (1–2%) and EC (0.4–1%). The undetermined mass was 20% at MSY and 13% at MSC. Absolute concentrations were also very similar at both sites, especially in PM₁ (Supplement Table S1) and except for winter (Fig. 1). In the PM_{1–10} fraction, MM values were higher at the continental background site, and nitrate levels were slightly higher at the regional background site. The closer compositional similarities for PM₁ fraction points to the suitability of using PM₁ as indicator of regional anthropogenic pollution in Europe, and reflects the wider spatial representativeness of the fine PM.

In spite of these results, more differences are evident concerning concentrations of specific major and trace PM components recorded at MSC and MSY. In agreement with the seasonal variation of PM concentrations, major and trace PM components showed a marked seasonal variation, with the highest concentrations during the warmer months, except for nitrate. Possible causes will be discussed in the following sections.

3.2.1 Nitrate (NO₃⁻)

Average concentrations of nitrate were slightly higher at the regional background site than those at the continental background site for the PM₁₀ fraction (Supplement Table S1, 1.2 vs. 0.8 μg m⁻³) and more similar for the PM₁ fraction (Supplement Table S1, 0.3 vs. 0.2 μg m⁻³). Nitrate showed a prevalent coarse grain size distribution (Figs. 2 and 3). PM_{1–10} nitrate compounds were partially associated with mineral dust and sea

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Aerosols in
continental and
regional background
in the Western
Mediterranean**

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



salt particles at both sites, since nitric acid and/or some other nitrogen compounds can react with these particles (Wall et al., 1988; Zhuang et al., 1999a), and they are much more stable than ammonium nitrate at high temperature and low humidity (Zhuang et al., 1999b). However, in the present study, part of the nitrate in the PM_{1-10} fraction was also ascribed to ammonium nitrate, as PM_{1-10} ammonium concentrations were also detected especially during the winter episodes (WREG and EU). Such coarse ammonium nitrate is most likely in $PM_{1-2.5}$ fraction, according to Querol et al. (2009), who found that $PM_{2.5-10}$ concentrations of ammonium nitrate were negligible at MSY.

Concentrations of nitrate decreased in summer at both sites, especially in PM_1 (2 and 3 times lower than the winter concentrations at MSC and MSY, respectively), whereas maxima were registered in February–April and in October at MSC, and from October to March at MSY (Fig. 2). The decrease of nitrate during the warmer months was attributed to the high volatility of ammonium nitrate (Pey et al., 2009) at low humidity and high temperature (Zhuang et al., 1999b).

Maxima nitrate concentrations can also be explained by the development of regional pollution episodes together with ambient conditions favoring the stability of ammonium nitrate. However, these maxima are not registered simultaneously at both environments (Fig. 2) due to the different origin of air masses affecting MSC and MSY sites throughout the year (Supplement Fig. S5). At MSY, the highest nitrate concentrations are related to the occurrence of winter anticyclonic episodes (WREG). Under these episodes, polluted air masses are transported from the Barcelona metropolitan area towards MSY (Pey et al., 2010) increasing the levels of anthropogenic PM components (Fig. 3 and Supplement Fig. S6a).

Figure 4d shows an example of a WREG episode affecting the WMB, during which the nitrate concentrations increased at the regional background site. In this episode, backward trajectories showed a regional air mass origin (Supplement Fig. S7) and PM_{10} nitrate daily concentrations increased from $0.2 \mu\text{g m}^{-3}$ on 13 January to $1.3\text{--}5.0 \mu\text{g m}^{-3}$ between 14–19 January 2012. Simultaneously, the continental background

site was almost unaffected by such polluted air masses and nitrate concentrations in PM_{10} remained between $0.1\text{--}0.6\ \mu\text{g m}^{-3}$ during this episode (Fig. 4d).

At MSC, the highest nitrate concentrations (more than 2 times the annual average concentrations) were associated with air masses from mainland Europe (Fig. 3 and Supplement Fig. S6a), more frequent in February–April and in October (Supplement Fig. S5). European air masses may be transported towards the WMB from Central and Eastern Europe, one of the most polluted regions in the continent. This transport occurs preferentially at high altitude layers (between 915–1930 m), as observed by Sicard et al. (2011). This may explain the higher impact at MSC than at MSY (Fig. 3).

Figure 4c shows an example of EU air mass transport affecting nitrate concentrations at the continental background. This episode occurred on 15 October 2011 and PM_{10} nitrate daily concentration at MSC reached $5\ \mu\text{g m}^{-3}$ (6 times higher than the annual average). During this episode the air mass remained 3 days over Central Europe before reaching MSC, as shown by the backward trajectories (Supplement Fig. S7). Simultaneously, the regional background site was less affected by this episode as PM_{10} nitrate concentration at MSY was $2.3\ \mu\text{g m}^{-3}$ ($1\ \mu\text{g m}^{-3}$ higher than the annual average).

In agreement with PM concentrations, average nitrate PM_{10} concentrations at MSC were slightly higher than those observed at the continental background site of Puy de Dôme (Bourcier et al., 2012), whereas the concentrations registered at MSY were lower than those measured at the rural stations of Payerne and Magadino (Gianini et al., 2012) (Supplement Table S1 and Fig. 5). Contrary to the seasonal variation observed in the WMB, the maximum nitrate concentrations were observed in summer at the continental background stations of Puy de Dôme (Bourcier et al., 2012), Mt. Cimone (Carbone et al., 2014) and Jungfraujoch (Cozic et al., 2008), probably due to their different meteorological conditions in summer, with lower temperatures at which the nitrate is stable in the atmosphere.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



3.2.2 Sulfate (SO₄²⁻)

As for nitrate, average concentrations of sulfate were slightly higher at the regional background site than those at the continental background site for the PM₁₀ fraction (Supplement Table S1, 1.9 vs. 1.3 μg m⁻³) and more similar for the PM₁ fraction (1.5 vs. 1.2 μg m⁻³). As expected, sulfate was mainly fine (Fig. 3), 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, PM₁₋₁₀ sulfate was also detected at both sites, and it was partially associated with mineral dust and sea salt particles, and partially attributed to the reaction product between sulfuric acid and/or sulfur dioxide (SO₂) and these natural particles (Wall et al., 1988; Zhuang et al., 1999a).

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. 2). 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 linked to the higher photochemistry in the atmosphere that enhances the SO₂ 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 (Millan et al., 1997). Sporadically, the higher sulfate concentrations in summer could be also linked to the higher frequency of NAF episodes (Supplement Fig. S5), during which a compression of the PBL is observed (Alastuey et al., 2005; Pandolfi et al., 2013) (Supplement Fig. S9), with the consequent increase in the concentration of pollutants in the WMB (Fig. 3 and Supplement Fig. S6b). Moreover, Jorba et al. (2013) found that during NAF episodes south synoptic flows dominate during the whole day breaking the regular sea breeze circulation. This process inhibits the sea breeze “clean up” effect causing a continuous transport of polluted air masses, especially at MSY.

An example of the impact of NAF episodes on the concentrations of sulfate was recorded on 26 March 2011 (Fig. 4a), when PM₁₀ sulfate daily concentration at MSY

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



reached $5.2 \mu\text{g m}^{-3}$ (almost 3 times higher than the annual average) and backward trajectories showed a North African air mass origin (Supplement Fig. S7). The increase in sulfate concentrations at the continental background site was less important, since these reached $2.9 \mu\text{g m}^{-3}$ (2 times higher than the annual average), even though MSC was more affected by the NAF episode as demonstrated by the higher MM concentrations.

Additionally, higher summer sulfate concentrations in the regional background may be partially due to the more intense sea breeze circulation affecting MSY, which may enhance the transport of shipping emissions from the Mediterranean to the continental areas (Fig. 3). Moreover, the increase of PM_{1-10} sulfate concentrations under SREG and NAF episodes, especially at MSY (Fig. 3), could be partially associated with mineral dust and sea salt particles, and partially linked to the aforementioned processes, which enhance the reaction of SO_2 with mineral dust at a regional scale.

The relatively low sulfate concentrations during WREG at MSY may be an indicator of the low emissions of SO_2 nearby this area, as most of the PM under these winter anticyclonic episodes has a nearby origin. Moreover, in winter the SO_2 oxidation rate is much lower than in summer (Querol et al., 1999).

Comparison of these results with those from other sites in Central Europe shows that PM_{10} sulfate concentrations were very similar at all sites (Supplement Table S1 and Fig. 5). These similar sulfate concentrations across Europe in both continental and regional background areas corroborate that this PM component can remain in the atmosphere for long time, homogenizing sulfate concentrations in Europe.

3.2.3 Ammonium (NH_4^+)

Average concentrations of ammonium at the regional and continental background sites were the same (Supplement Table S1, $0.5 \mu\text{g m}^{-3}$ in PM_{10} and $0.5 \mu\text{g m}^{-3}$ in PM_1). Ammonium showed a prevalent fine grain size distribution (Figs. 2 and 3). Fine ammonium was attributed to the presence of both ammonium nitrate and ammonium sulfate.

PM₁₋₁₀ ammonium was also ascribed to ammonium nitrate in the PM_{1-2.5} fraction, as described above.

Ammonium concentrations did not follow a clear seasonal pattern (Fig. 2). Nevertheless, at MSC lower ammonium concentrations in November–January were recorded, as MSC is mostly within the FT in this period. During the rest of the colder months at MSC, and in winter period at MSY, ammonium was principally associated with nitrate ($R^2 = 0.77$ and 0.80 at MSC and MSY, respectively), because ammonium nitrate is thermodynamically stable at low temperatures and high humidity (Zhuang et al., 1999b), and due to a lesser competence with ammonium sulfate formation. At MSY, sporadically high values of ammonium in winter were associated with the aforementioned WREG episodes (Figs. 3 and 4d), whereas at MSC they were linked to air masses from Central and Eastern Europe (Figs. 3 and 4c). During the warmer months ammonium was mostly associated with sulfate and thus it was linked to the higher photochemistry in the atmosphere and to the SREG episodes.

3.2.4 Organic matter (OM)

Average concentrations of OM were slightly higher at the regional background site than those at the continental background site in PM₁₀ (Supplement Table S1, 4.0 vs. $3.2 \mu\text{g m}^{-3}$) and similar in PM₁ (2.9 vs. $2.8 \mu\text{g m}^{-3}$). OM was found mainly in the fine fraction (Figs. 2 and 3). A high number of studies have demonstrated the dominant secondary origin of the fine OM (Jimenez et al., 2009). In the study area, the origin of OM has been recently investigated (Minguillón et al., 2011; Mohr et al., 2012), and it has been found that secondary organic aerosols (SOA) accounted for 91 % and 55 % of the OM at MSY and at the city of Barcelona, respectively. 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 PM₁₋₁₀, especially in spring and summer, suggests the impact of primary bioaerosols (Pöschl et al., 2010).

OM 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

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



concentrations at MSC and MSY, respectively) (Fig. 2). The summer maximum was linked 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 episodes; (3) the greater biogenic emissions from vegetation (Seco et al., 2011); and (4) the higher frequency of wildfires. These processes could also explain why high PM_{10} OM concentrations at both sites were measured during NAF episodes (Fig. 3), since these episodes are more frequent in summer (Supplement Fig. S5).

Furthermore, at MSY a secondary maximum of OM concentrations occurred in October–March (Fig. 2), linked to the occurrence of WREG episodes (Fig. 4d). The continental background site was unaffected by this type of episodes, although, as for the case of nitrate, high OM concentrations were measured at MSC under EU episodes (Figs. 3 and 4c).

3.2.5 Elemental Carbon (EC)

Average concentrations of EC were higher at the regional background site than those at the continental background site (Supplement Table S1, 0.23 vs. 0.12 $\mu\text{g m}^{-3}$ in PM_{10} and 0.17 vs. 0.09 $\mu\text{g m}^{-3}$ in PM_{10}). This difference confirms that MSC is located at a sufficient altitude and distance from large urban/industrial agglomerations to avoid direct anthropogenic influence (Ripoll et al., 2014). EC showed a prevalent fine grain size distribution (Figs. 2 and 3). However, PM_{1-10} EC was also detected during SREG, NAF, MED and EU episodes. This suggests a partial association between EC and MM by means of adsorption of anthropogenic pollutants onto dust. According to the findings by Querol et al. (2009), this process may happen more likely in $PM_{1-2.5}$ than in $PM_{2.5-10}$ as OM + EC concentrations in the latest were negligible at MSY.

As expected, EC at both sites showed the same seasonal pattern described for BC by Ripoll et al. (2014), with high concentrations in the warmer months at MSC and a less marked seasonal variation observed at MSY (Fig. 2). The smoother seasonal variation at MSY reflects the regional anthropogenic influence on the levels of this

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



component, since anthropogenic emissions occur all along the year. The higher summer EC concentrations at both sites (more elevated at MSY) were attributed to the impact of the SREG episodes and to the higher occurrence of wildfires in North Africa and/or in the WMB (Cristofanelli et al., 2009). Sporadically, they could also be linked to the higher frequency of NAF episodes (Supplement Fig. S5), which increases the concentration of pollutants due to the compression of the PBL (Supplement Fig. S9), and causes the continuous transport of polluted air masses owing to the change in the regular sea breeze circulation (Jorba et al., 2013), especially at MSY (Figs. 3 and 4b). The difference in summer EC concentrations between both sites could be associated with the more intense sea breeze circulation affecting MSY, which may enhance the transport of shipping emissions from the Mediterranean to the continental areas (Fig. 3).

Figure 4e illustrates the impact of a wildfire, which took place in Eastern Europe on 27–28 March 2012, on the OM and EC levels recorded at both MSY and MSC. The NAAPS model shows high smoke surface concentrations over SE Europe (Supplement Fig. S8) and the air mass backward trajectories confirm the pathway of air mass being transported from SE Europe (Supplement Fig. S7). During this episode, daily PM₁₀ OM concentration at MSY reached 7.9 μg m⁻³ (almost 2 times higher than the annual average) and it was 9.1 μg m⁻³ at MSC (almost 3 times higher than the annual average). EC concentrations also increased up to 0.3 μg m⁻³ at both sites (1.5 and 3 times the annual average).

As for the case of nitrate, the increase of EC during the colder months was not registered simultaneously at both environments (Fig. 2). In the regional background this increase was observed from October to March and was attributed to the aforementioned WREG episodes (Figs. 3 and 4d). By contrast, in the continental background it was observed from February to April and in October, and was associated with EU episodes (Figs. 3 and 4c).

Average PM₁₀ concentrations of EC in the WMB were lower than those measured at Payerne and Magadino (Gianini et al., 2012) (Supplement Table S1 and Fig. 5), probably due to the higher influence of biomass burning at these Swiss sites.

3.2.6 Mineral Matter (MM)

As expected, most of the MM was encountered in the PM_{1-10} fraction, being the PM_1 concentrations very low at both sites ($< 0.3 \mu\text{g m}^{-3}$). Average concentrations of PM_{1-10} MM were higher in the continental background than those in the regional background site (4.0 vs. $3.3 \mu\text{g m}^{-3}$), probably due to the prevalence of long-range dust transport at higher altitude layers (Sicard et al., 2011).

MM concentrations are driven by the local and regional dust resuspension and by the contribution of African dust outbreaks, resulting in higher values during the warmer months and the lowest during the colder months, with sporadic high concentrations in March–April (Fig. 2). African dust outbreaks, which transport dust particles over WMB are more frequent in summer and in March–April (Supplement Fig. S5). Furthermore, dust resuspension in summer is enhanced by the dryness of soils, low rainfall, and high convective dynamics (Rodriguez et al., 2002). High concentrations of elements such as Ca, Mg, Sr, and Mn during SREG scenarios (Fig. 6) further 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 for the continental background compared to the regional background, probably because of the calcareous (richer in Ca, Mg, Sr and Mn) nature of Montsec range in front of the slate and granitic composition (richer in Al) of Montseny range.

The average concentrations of MM-related elements during NAF episodes were higher in the continental background than those in the regional background (Fig. 6), which corroborates that the long-range dust transport occurs preferentially at high altitude layers. A specific example can be seen in Fig. 4b. On 26 March 2011 PM_{10} MM concentration at MSC reached $16 \mu\text{g m}^{-3}$ (more than 5 times higher than the annual average) and it was $9 \mu\text{g m}^{-3}$ (about 3 times higher than the annual average) at MSY. Backward trajectory for this day clearly showed a North African air mass origin (Supplement Fig. S7).

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Opposite to the rest of MM species, K was also abundant in PM₁ fraction (Fig. 6), and its concentrations were slightly higher at MSY than those at MSC (Fig. 6 and Supplement Fig. S6c). 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 performed in March 2009 revealed that biomass burning emissions accounted for 33 % of EC and 22 % of OM at MSY (Minguillón et al., 2012). These contributions are much lower than those obtained at other European regions (Pio et al., 2011). Although it is not a major source in the WMB, the partial contribution of biomass burning is observed, as shown in Fig. 4e. During this episode, K concentrations in PM₁₀ at MSY reached 0.27 μg m⁻³ (2 times higher than the annual average), with a prevalent fine partitioning (PM₁/PM₁₀ = 0.55), instead of its main coarse occurrence (PM₁/PM₁₀ = 0.28) during NAF episodes.

Mineral elements (Al + Ca + Mg) at MSC and MSY were recorded in concentrations twice as high as those at Payerne and Magadino (Gianini et al., 2012) (Supplement Table S1 and Fig. 5), indicating the higher influence of Saharan dust transport and regional dust resuspension in the Mediterranean area. Conversely, the total potassium concentrations were higher at the Swiss stations than those at MSY and MSC (Fig. 5). Considering that potassium was mainly water soluble at the Swiss sites (70 %, Gianini et al., 2012) and that this PM component is a tracer of biomass burning (Pio et al., 2008), it corroborates the higher influence of biomass burning in Central European than in the WMB.

3.2.7 Sea Salt (SS)

Average concentrations of SS were higher at the regional background site than those at the continental background site (Supplement Table S1, 0.14 vs. 0.07 μg m⁻³ in PM₁, and 0.5 vs. 0.3 μg m⁻³ in PM₁₀), because MSY is located closer to the coast and more influenced by sea breezes. As expected, SS components were found mainly in the PM₁₋₁₀ fraction (Fig. 3). In the continental background, the lower variation of SS

concentrations as a function of meteorological episodes reflects the minor impact of marine aerosols in this area.

As expected, MSC and MSY sites showed a higher influence of SS particles than Payerne and Magadino (Gianini et al., 2012) (Supplement Table S1 and Fig. 5), owing to their closer location to the Mediterranean Sea.

3.2.8 Trace elements

Average concentrations of the majority of trace elements were higher at the regional background site than those at the continental background site in PM₁₀ and PM₁, except for the MM-related elements (Supplement Table S1). Most of the trace elements were mainly found in the fine fraction (Supplement Fig. S10). In order to provide a global picture of the origin and variability of trace elements in the study region, a Principal Component Analysis (PCA), using Varimax rotation (Thurston and Spengler, 1985), was performed. This exploration permitted the identification of three main common groups of trace elements in PM₁₀ at the continental and regional background sites (Supplement Tables S2 and S3). Ordered by their contribution to the total mass of trace elements in PM₁₀ these groups were: mineral, industrial + road traffic and fuel oil combustion related elements (Fig. 7).

Mineral trace elements

Typical crustal elements (Ti, Mn, Li, and Sr) were included in this group, but also V, Cr, Co, Ni, and As (Supplement Tables S2 and S3) were partially associated with this factor since these elements, usually associated to anthropogenic sources, are also found in clay mineral assemblages. The typical crustal elements were encountered in the PM₁₋₁₀ fraction, being the PM₁ concentrations negligible ($< 1.1 \text{ ng m}^{-3}$) (Supplement Table S1). These elements follow the same seasonal variation described for the MM concentrations.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Industrial + road traffic trace elements

The group enriched in Cu, Zn, As, Cd, Pb, Sb and Sn was associated with industrial + road traffic sources (Supplement Tables S2 and S3), 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, (b) Cu and Sn as tracers of vehicle exhaust, and (c) Cu 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. Most of these elements were found mainly in the PM₁ fraction, except for Cu, As and Sb, which were also found in the PM₁₋₁₀ fraction (Supplement Fig. S10). The seasonal pattern of this group of elements showed low variations, especially at MSY (Fig. 7) confirming the anthropogenic origin of these elements. Furthermore, the highest concentrations of these elements were measured under NAF episodes at both sites (Supplement Fig. S10 and Fig. 4b), probably due to the compression of the PBL (Supplement Fig. S9) and to the change in the regular sea breeze circulation (Jorba et al., 2013) during this type of episodes. High concentrations of these elements were also measured under SREG and EU episodes at both sites and under MED episodes at MSY (Supplement Fig. S10), as MSY is located downwind of large emissions sources during easterly Mediterranean advections.

Fuel oil combustion trace elements

This group was better identified at MSY than at MSC, and it was traced by V and Ni (Supplement Tables S2 and S3). 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). V and Ni showed a prevalent fine grain size distribution (PM₁/PM₁₀ = 0.6 and 0.8, respectively, Supplement Fig. S10), although they were also found in the PM₁₋₁₀ fraction, as described above. Average concentrations of these elements were higher in the regional background than those in the continental

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



background, especially in the PM_1 fraction (Supplement Table S1), as MSY is located closer to the sea. This group of 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 frequent and intense sea breeze circulation, which enhances the transport of air masses from the Mediterranean. This could also explain why high concentrations of V and Ni were measured under SREG episodes at both sites (Supplement Fig. S10).

As for the mineral matter concentrations, the typical crustal trace elements, such as Ti, Sr, La or Ce, were higher at MSC and MSY than at Payerne and Magadino (Fig. 8). 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 (Fig. 8), 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).

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 (WMB).

In this particular region of the WMB, the continental to regional background increase has been estimated in $4.0 \mu\text{g m}^{-3}$ for PM_{10} and $1.1 \mu\text{g m}^{-3}$ for PM_1 . Relative chemical composition and absolute concentrations of PM 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, 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

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

of PM₁ concentration and composition through the region, allowing us to consider PM₁ as a more suitable indicator of anthropogenic impact than PM₁₀. Nevertheless, 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 components.

The seasonal variation of major and trace PM 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 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. Moreover, during NAF episodes a compression of the PBL and a change in the wind regime towards a permanent southern flow increments 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 components is usually higher in the continental background, since this transport from Europe occurs preferentially at

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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.

Meteorological variables such as temperature, humidity and solar radiation are also connected to the time variation displayed by the major and trace PM components. In the WMB, the higher temperature and solar radiation in the warmer months augments atmospheric photochemistry, promoting the formation of secondary inorganic and organic aerosols and thus incrementing markedly the concentration of certain PM components such as sulfate and OM. Additionally, the higher temperatures in the warmer months enhance sea breeze circulation which favors the transport of shipping emissions from the Mediterranean to the continental areas increasing concentration of PM components such as 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.

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. This is reflected in more elevated concentrations of mineral elements across the Mediterranean, with the only exception of 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 is probably linked to the long residence time of sulfate aerosols in the atmosphere. On

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



the contrary, 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.

Monitoring of relevant properties of aerosols in these sites give us a general picture of atmospheric aerosols for a wide area in the WMB. In view of the relatively high concentrations of atmospheric aerosols from a variety of sources, both natural and anthropogenic, and the importance of atmospheric processes resulting in a complex vertical distribution with a wide horizontal representativeness, the characterization of atmospheric aerosols in this region provides valuable information for air quality and climate models.

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References

- Alastuey, A., Querol, X., Castillo, S., Escudero, M., Avila, A., Cuevas, E., Torres, C., Romero, P., Exposito, F., and Garcia, O.: Characterisation of TSP and PM_{2.5} at Izaña and Sta. Cruz de Tenerife (Canary Islands, Spain) during a Saharan Dust Episode (July 2002), *Atmos. Environ.*, 39, 4715–4728, doi:10.1016/j.atmosenv.2005.04.018, 2005.
- Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Perez, N., and Hopke, P. K.: Quantifying road dust resuspension in urban environment by Multilinear Engine: a comparison with PMF2, *Atmos. Environ.*, 43, 2770–2780, doi:10.1016/j.atmosenv.2009.02.039, 2009.
- Andrews, E., Ogren, J. A., Bonasoni, P., Marinoni, A., Cuevas, E., Rodríguez, S., Sun, J. Y., Jaffe, D. A., Fischer, E. V., Baltensperger, U., Weingartner, E., Coen, M. C., Sharma, S., Macdonald, A. M., Leaitch, W. R., Lin, N.-H., Laj, P., Arsov, T., Kalapov, I., Jefferson, A., and Sheridan, P.: Climatology of aerosol radiative properties in the free troposphere, *Atmos. Res.*, 102, 365–393, doi:10.1016/j.atmosres.2011.08.017, 2011.
- Belis, C. A., Karagulian, F., Larsen, B. R., and Hopke, P. K.: Critical review and meta-analysis of ambient particulate matter source apportionment using receptor models in Europe, *Atmos. Environ.*, 69, 94–108, doi:10.1016/j.atmosenv.2012.11.009, 2013.
- Bourcier, L., Sellegri, K., Chausse, P., Pichon, J. M., and Laj, P.: Seasonal variation of water-soluble inorganic components in aerosol size-segregated at the puy de Dôme station (1465 m a.s.l.), France, *J. Atmos. Chem.*, 69, 47–66, doi:10.1007/s10874-012-9229-2, 2012.
- Burkhardt, J. and Pariyar, S.: Particulate pollutants are capable to “degrade” epicuticular waxes and to decrease the drought tolerance of Scots pine (*Pinus sylvestris* L.), *Environ. Pollut.*, 184, 659–67, doi:10.1016/j.envpol.2013.04.041, 2014.
- Carbone, C., Decesari, S., Mircea, M., Giulianelli, L., Finessi, E., Rinaldi, M., Fuzzi, S., Marinoni, a., Duchi, R., Perrino, C., Sargolini, T., Vardè, M., Sprovieri, F., Gobbi, G. P., Angelini, F., and Facchini, M. C.: Size-resolved aerosol chemical composition over the Italian Peninsula during typical summer and winter conditions, *Atmos. Environ.*, 44, 5269–5278, doi:10.1016/j.atmosenv.2010.08.008, 2010.
- Carbone, C., Decesari, S., Paglione, M., Giulianelli, L., Rinaldi, M., Marinoni, A., Cristofanelli, P., Didiodato, A., Bonasoni, P., Fuzzi, S., and Facchini, M. C.: 3-year chemical composition of free tropospheric PM₁ at the Mt. Cimone GAW global station – South Europe – 2165 m a.s.l., *Atmos. Environ.*, 87, 218–227, doi:10.1016/j.atmosenv.2014.01.048, 2014.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Aerosols in
continental and
regional background
in the Western
Mediterranean**

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Cavalli, F., Viana, M., Yttri, K. E., Genberg, J., and Putaud, J.-P.: Toward a standardised thermal-optical protocol for measuring atmospheric organic and elemental carbon: the EUSAAR protocol, *Atmos. Meas. Tech.*, 3, 79–89, doi:10.5194/amt-3-79-2010, 2010.

Cozic, J., Verheggen, B., Weingartner, E., Crosier, J., Bower, K. N., Flynn, M., Coe, H., Henning, S., Steinbacher, M., Henne, S., Collaud Coen, M., Petzold, A., and Baltensperger, U.: Chemical composition of free tropospheric aerosol for PM₁ and coarse mode at the high alpine site Jungfraujoch, *Atmos. Chem. Phys.*, 8, 407–423, doi:10.5194/acp-8-407-2008, 2008.

Cristofanelli, P., Marinoni, A., Arduini, J., Bonafè, U., Calzolari, F., Colombo, T., Decesari, S., Duchi, R., Facchini, M. C., Fierli, F., Finessi, E., Maione, M., Chiari, M., Calzolari, G., Messina, P., Orlandi, E., Roccatò, F., and Bonasoni, P.: Significant variations of trace gas composition and aerosol properties at Mt. Cimone during air mass transport from North Africa – contributions from wildfire emissions and mineral dust, *Atmos. Chem. Phys.*, 9, 4603–4619, doi:10.5194/acp-9-4603-2009, 2009.

Cusack, M., Alastuey, A., Pérez, N., Pey, J., and Querol, X.: Trends of particulate matter (PM_{2.5}) and chemical composition at a regional background site in the Western Mediterranean over the last nine years (2002–2010), *Atmos. Chem. Phys.*, 12, 8341–8357, doi:10.5194/acp-12-8341-2012, 2012.

Cusack, M., Pérez, N., Pey, J., Wiedensohler, A., Alastuey, A., and Querol, X.: Variability of sub-micrometer particle number size distributions and concentrations in the Western Mediterranean regional background, *Tellus B*, 65, 1–19, doi:10.3402/tellusb.v65i0.19243, 2013.

EC: Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on Ambient Air Quality and Cleaner Air for Europe (OJ L 152, 11.6.2008, pp. 1–44), available at: <http://eur-lex.europa.eu/homepage.html> (last access: 16 June 2014), 2008.

Gianini, M. F. D., Gehrig, R., Fischer, A., Ulrich, A., Wichser, A., and Hueglin, C.: Chemical composition of PM₁₀ in Switzerland: an analysis for 2008/2009 and changes since 1998/1999, *Atmos. Environ.*, 54, 97–106, doi:10.1016/j.atmosenv.2012.02.037, 2012.

IPCC 2013: Climate Change 2013: The Physical Science Basis (Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change), edited by: Stocker, T. F., Qin, D., Plattner, G. K., Tignor, M. M. B., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midgley, P. M., Cambridge Univ. Press, UK and New York, USA, 2013.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Jimenez, J. L., Canagaratna, M. R., Donahue, N. M., Prevot, A. S. H., Zhang, Q., Kroll, J. H., DeCarlo, P. F., Allan, J. D., Coe, H., Ng, N. L., Aiken, A. C., Docherty, K. S., Ulbrich, I. M., Grieshop, A. P., Robinson, A. L., Duplissy, J., Smith, J. D., Wilson, K. R., Lanz, V. A., Hueglin, C., Sun, Y. L., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J. M., Collins, D. R., Cubison, M. J., Dunlea, E. J., Huffman, J. A., Onasch, T. B., Alfarra, M. R., Williams, P. I., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J. Y., Zhang, Y. M., Dzepina, K., Kimmel, J. R., Sueper, D., Jayne, J. T., Herndon, S. C., Trimborn, A. M., Williams, L. R., Wood, E. C., Middlebrook, A. M., Kolb, C. E., Baltensperger, U., and Worsnop, D. R.: Evolution of organic aerosols in the atmosphere, *Science*, 326, 1525–1529, doi:10.1126/science.1180353, 2009.

Jorba, O., Pandolfi, M., Spada, M., Baldasano, J. M., Pey, J., Alastuey, A., Arnold, D., Sicard, M., Artiñano, B., Revuelta, M. A., and Querol, X.: Overview of the meteorology and transport patterns during the DAURE field campaign and their impact to PM observations, *Atmos. Environ.*, 77, 607–620, doi:10.1016/j.atmosenv.2013.05.040, 2013.

Laj, P., Klausen, J., Bilde, M., Plaß-Duelmer, C., Pappalardo, G., Clerbaux, C., Baltensperger, U., Hjorth, J., Simpson, D., Reimann, S., Coheur, P.-F., Richter, A., De Mazzière, M., Rudich, Y., McFiggans, G., Torseth, K., Wiedensohler, A., Morin, S., Schulz, M., Allan, J. D., Attié, J.-L., Barnes, I., Birmili, W., Cammas, J. P., Dommen, J., Dorn, H.-P., Fowler, D., Fuzzi, S., Glasius, M., Granier, C., Hermann, M., Isaksen, I. S. A., Kinne, S., Koren, I., Madonna, F., Maione, M., Massling, A., Moehler, O., Mona, L., Monks, P. S., Müller, D., Müller, T., Orphal, J., Peuch, V.-H., Stratmann, F., Tanré, D., Tyndall, G., Abo Riziq, A., Van Roozendaal, M., Villani, P., Wehner, B., Wex, H., and Zardini, A. A.: Measuring atmospheric composition change, *Atmos. Environ.*, 43, 5351–5414, doi:10.1016/j.atmosenv.2009.08.020, 2009.

Marenco, F., Bonasoni, P., Calzolari, F., Ceriani, M., Chiari, M., Cristofanelli, P., D'Alessandro, A., Fermo, P., Lucarelli, F., Mazzei, F., Nava, S., Piazzalunga, A., Prati, P., Valli, G., and Vecchi, R.: Characterization of atmospheric aerosols at Monte Cimone, Italy, during summer 2004: source apportionment and transport mechanisms, *J. Geophys. Res.*, 111, D24202, doi:10.1029/2006JD007145, 2006.

**Aerosols in
continental and
regional background
in the Western
Mediterranean**

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

- Millan, M. M., Salvador, R., Mantilla, E., and Kallos, G.: Photooxidant dynamics in the Mediterranean basin in summer: results from European research projects, *J. Geophys. Res.*, 102, 8811–8823, 1997.
- Minguillón, M. C., Perron, N., Querol, X., Szidat, S., Fahrni, S. M., Alastuey, A., Jimenez, J. L., Mohr, C., Ortega, A. M., Day, D. A., Lanz, V. A., Wacker, L., Reche, C., Cusack, M., Amato, F., Kiss, G., Hoffer, A., Decesari, S., Moretti, F., Hillamo, R., Teinilä, K., Seco, R., Peñuelas, J., Metzger, A., Schallhart, S., Müller, M., Hansel, A., Burkhardt, J. F., Baltensperger, U., and Prévôt, A. S. H.: Fossil versus contemporary sources of fine elemental and organic carbonaceous particulate matter during the DAURE campaign in Northeast Spain, *Atmos. Chem. Phys.*, 11, 12067–12084, doi:10.5194/acp-11-12067-2011, 2011.
- Minguillón, M. C., Querol, X., Baltensperger, U., and Prévôt, A. S. H.: Fine and coarse PM composition and sources in rural and urban sites in Switzerland: local or regional pollution?, *Sci. Total Environ.*, 427–428, 191–202, doi:10.1016/j.scitotenv.2012.04.030, 2012.
- Minguillón, M. C., Cirach, M., Hoek, G., Brunekreef, B., Tsai, M., de Hoogh, K., Jedynska, A., Kooter, I. M., Nieuwenhuijsen, M., and Querol, X.: Spatial variability of trace elements and sources for improved exposure assessment in Barcelona, *Atmos. Environ.*, 89, 268–281, doi:10.1016/j.atmosenv.2014.02.047, 2014.
- Mohr, C., DeCarlo, P. F., Heringa, M. F., Chirico, R., Slowik, J. G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Peñuelas, J., Jiménez, J. L., Crippa, M., Zimmermann, R., Baltensperger, U., and Prévôt, A. S. H.: Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data, *Atmos. Chem. Phys.*, 12, 1649–1665, doi:10.5194/acp-12-1649-2012, 2012.
- Moreno, T., Querol, X., Castillo, S., Alastuey, A., Cuevas, E., Herrmann, L., Mounkaila, M., Elvira, J., and Gibbons, W.: Geochemical variations in aeolian mineral particles from the Sahara-Sahel Dust Corridor, *Chemosphere*, 65, 261–70, doi:10.1016/j.chemosphere.2006.02.052, 2006.
- Nyeki, S., Baltensperger, U., Colbeck, I., Jost, D. T., Weingartner, E., and Gäggeler, H. W.: The Jungfrauoch high-alpine research station (3454 m) as a background clean continental site for the measurement of aerosol parameters, *J. Geophys. Res.*, 103, 6097–6107, 1998.
- Pandolfi, M., Martucci, G., Querol, X., Alastuey, A., Wilsenack, F., Frey, S., O'Dowd, C. D., and Dall'Osto, M.: Continuous atmospheric boundary layer observations in the coastal urban area of Barcelona during SAPUSS, *Atmos. Chem. Phys.*, 13, 4983–4996, doi:10.5194/acp-13-4983-2013, 2013.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Pérez, N., Pey, J., Castillo, S., Viana, M., Alastuey, A., and Querol, X.: Interpretation of the variability of levels of regional background aerosols in the Western Mediterranean, *Sci. Total Environ.*, 407, 527–40, doi:10.1016/j.scitotenv.2008.09.006, 2008.

Pey, J., Pérez, N., Castillo, S., Viana, M., Moreno, T., Pandolfi, M., López-Sebastián, J. M., Alastuey, A., and Querol, X.: Geochemistry of regional background aerosols in the Western Mediterranean, *Atmos. Res.*, 94, 422–435, doi:10.1016/j.atmosres.2009.07.001, 2009.

Pey, J., Pérez, N., Querol, X., Alastuey, A., Cusack, M., and Reche, C.: Intense winter atmospheric pollution episodes affecting the Western Mediterranean, *Sci. Total Environ.*, 408, 1951–1959, doi:10.1016/j.scitotenv.2010.01.052, 2010.

Pey, J., Pérez, N., Cortés, J., Alastuey, A., and Querol, X.: Chemical fingerprint and impact of shipping emissions over a Western Mediterranean metropolis: primary and aged contributions, *Sci. Total Environ.*, 463–464, 497–507, doi:10.1016/j.scitotenv.2013.06.061, 2013a.

Pey, J., Querol, X., Alastuey, A., Forastiere, F., and Stafoggia, M.: African dust outbreaks over the Mediterranean Basin during 2001–2011: PM₁₀ concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology, *Atmos. Chem. Phys.*, 13, 1395–1410, doi:10.5194/acp-13-1395-2013, 2013b.

Pio, C. A., Legrand, M., Alves, C. A., Oliveira, T., Afonso, J., Caseiro, A., Puxbaum, H., Sanchez-Ochoa, A., and Gelencsér, A.: Chemical composition of atmospheric aerosols during the 2003 summer intense forest fire period, *Atmos. Environ.*, 42, 7530–7543, doi:10.1016/j.atmosenv.2008.05.032, 2008.

Pio, C., Cerqueira, M., Harrison, R. M., Nunes, T., Mirante, F., Alves, C., Oliveira, C., Sanchez de la Campa, A., Artíñano, B., and Matos, M.: OC/EC ratio observations in Europe: rethinking the approach for apportionment between primary and secondary organic carbon, *Atmos. Environ.*, 45, 6121–6132, doi:10.1016/j.atmosenv.2011.08.045, 2011.

Pöschl, U., Martin, S. T., Sinha, B., Chen, Q., Gunthe, S. S., Huffman, J. A., Borrmann, S., Farmer, D. K., Garland, R. M., Helas, G., Jimenez, J. L., King, S. M., Manzi, A., Mikhailov, E., Pauliquevis, T., Petters, M. D., Prenni, A. J., Roldin, P., Rose, D., Schneider, J., Su, H., Zorn, S. R., Artaxo, P., and Andreae, M. O.: Rainforest aerosols as biogenic nuclei of clouds and precipitation in the Amazon, *Science*, 329, 1513–1516, doi:10.1126/science.1191056, 2010.

Putaud, J. P., Van Dingenen, R., Alastuey, A., Bauer, H., Birmili, W., Cyrys, J., Flentje, H., Fuzzi, S., Gehrig, R., Hansson, H. C., Harrison, R. M., Herrmann, H., Hittenberger, R., Hüglin, C., Jones, A. M., Kasper-Giebl, A., Kiss, G., Kousa, A., Kuhlbusch, T. A. J.,

**Aerosols in
continental and
regional background
in the Western
Mediterranean**

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Löschau, G., Maenhaut, W., Molnar, A., Moreno, T., Pekkanen, J., Perrino, C., Pitz, M., Puxbaum, H., Querol, X., Rodriguez, S., Salma, I., Schwarz, J., Smolik, J., Schneider, J., Spindler, G., ten Brink, H., Tursic, J., Viana, M., Wiedensohler, A., and Raes, F.: A European aerosol phenomenology – 3: Physical and chemical characteristics of particulate matter from 60 rural, urban, and kerbside sites across Europe, *Atmos. Environ.*, 44, 1308–1320, doi:10.1016/j.atmosenv.2009.12.011, 2010.

Querol, X., Alastuey, A. S., Puigercus, J. A., Mantilla, E., Miro, J. V, Lopez-soler, A., Plana, F., and Artíñano, B.: Seasonal evolution of suspended particles around a large coal-fired power station: chemical characterization, *Atmos. Environ.*, 32, 719–731, 1998.

Querol, X., Alastuey, A., Lopez-soler, A., Plana, F., and Puigercus, J. A.: Daily evolution of sulphate aerosols in a rural area, northeastern Spain – elucidation of an atmospheric reservoir effect, *Environ. Pollut.*, 105, 397–407, doi:10.1016/S0269-7491(99)00037-8, 1999.

Querol, X., Alastuey, A., Rodriguez, S., Plana, F., Mantilla, E., and Ruiz, C. R.: Monitoring of PM₁₀ and PM_{2.5} around primary particulate anthropogenic emission sources, *Atmos. Environ.*, 35, 845–858, doi:10.1016/S1352-2310(00)00387-3, 2001.

Querol, X., Viana, M., Alastuey, A., Amato, F., Moreno, T., Castillo, S., Pey, J., de la Rosa, J., Sánchez de la Campa, A., Artíñano, B., Salvador, P., García Dos Santos, S., Fernández-Patier, R., Moreno-Grau, S., Negral, L., Minguillón, M. C., Monfort, E., Gil, J. I., Inza, A., Ortega, L. A., Santamaría, J. M., and Zabalza, J.: Source origin of trace elements in PM from regional background, urban and industrial sites of Spain, *Atmos. Environ.*, 41, 7219–7231, doi:10.1016/j.atmosenv.2007.05.022, 2007.

Querol, X., Alastuey, A., Pey, J., Cusack, M., Pérez, N., Mihalopoulos, N., Theodosi, C., Gerasopoulos, E., Kubilay, N., and Koçak, M.: Variability in regional background aerosols within the Mediterranean, *Atmos. Chem. Phys.*, 9, 4575–4591, doi:10.5194/acp-9-4575-2009, 2009.

Ripoll, A., Pey, J., Minguillón, M. C., Pérez, N., Pandolfi, M., Querol, X., and Alastuey, A.: Three years of aerosol mass, black carbon and particle number concentrations at Montsec (southern Pyrenees, 1570 m a.s.l.), *Atmos. Chem. Phys.*, 14, 4279–4295, doi:10.5194/acp-14-4279-2014, 2014.

Rodríguez, S., Querol, X., Alastuey, A., and Mantilla, E.: Origin of high summer PM₁₀ and TSP concentrations at rural sites in eastern Spain, *Atmos. Environ.*, 36, 3101–3112, doi:10.1016/S1352-2310(02)00256-X, 2002.

**Aerosols in
continental and
regional background
in the Western
Mediterranean**

A. Ripoll et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Rodríguez, S., Querol, X., Alastuey, A., Viana, M.-M., and Mantilla, E.: Events affecting levels and seasonal evolution of airborne particulate matter concentrations in the Western Mediterranean, *Environ. Sci. Technol.*, 37, 216–222, doi:10.1021/es020106p, 2003.

Seco, R., Peñuelas, J., Filella, I., Llusà, J., Molowny-Horas, R., Schallhart, S., Metzger, A., Müller, M., and Hansel, A.: Contrasting winter and summer VOC mixing ratios at a forest site in the Western Mediterranean Basin: the effect of local biogenic emissions, *Atmos. Chem. Phys.*, 11, 13161–13179, doi:10.5194/acp-11-13161-2011, 2011.

Sicard, M., Rocadenbosch, F., Reba, M. N. M., Comerón, A., Tomás, S., García-Vízcaino, D., Batet, O., Barrios, R., Kumar, D., and Baldasano, J. M.: Seasonal variability of aerosol optical properties observed by means of a Raman lidar at an EARLINET site over Northeastern Spain, *Atmos. Chem. Phys.*, 11, 175–190, doi:10.5194/acp-11-175-2011, 2011.

Takahama, S., Schwartz, R. E., Russell, L. M., Macdonald, A. M., Sharma, S., and Leaitch, W. R.: Organic functional groups in aerosol particles from burning and non-burning forest emissions at a high-elevation mountain site, *Atmos. Chem. Phys.*, 11, 6367–6386, doi:10.5194/acp-11-6367-2011, 2011.

Thurston, G. D. and Spengler, J. D.: A quantitative assessment of source contributions to inhalable particulate matter pollution in metropolitan Boston, *Atmos. Environ.*, 19, 9–25, 1985.

Tositti, L., Riccio, a., Sandrini, S., Brattich, E., Baldacci, D., Parmeggiani, S., Cristofanelli, P., and Bonasoni, P.: Short-term climatology of PM₁₀ at a high altitude background station in southern Europe, *Atmos. Environ.*, 65, 142–152, doi:10.1016/j.atmosenv.2012.10.051, 2013.

Wall, S. M., John, W., and Ondo, J. L.: Measurement of aerosol size distributions for nitrate and major ionic species, *Atmos. Environ.*, 22, 1649–1656, doi:10.1016/0004-6981(88)90392-7, 1988.

WHO: Review of Evidence on Health Aspects of Air Pollution – REVIHAAP, First Results, WHO's Regional Office for Europe, Copenhagen, 28 pp., available at: http://www.euro.who.int/__data/assets/pdf_file/0020/182432/e96762-final.pdf (last access: 16 June 2014), 2013.

Zhuang, H., Chan, C. K., Fang, M., and Wexler, A. S.: Formation of nitrate and non-sea-salt sulfate on coarse particles, *Atmos. Environ.*, 33, 4223–4233, doi:10.1016/S1352-2310(99)00186-7, 1999a.

Zhuang, H., Chan, C. K., Fang, M., and Wexler, A. S.: Size distributions of particulate sulfate, nitrate, and ammonium at a coastal site in Hong Kong, *Atmos. Environ.*, 3, 843–853, doi:10.1016/S1352-2310(98)00305-7, 1999b.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

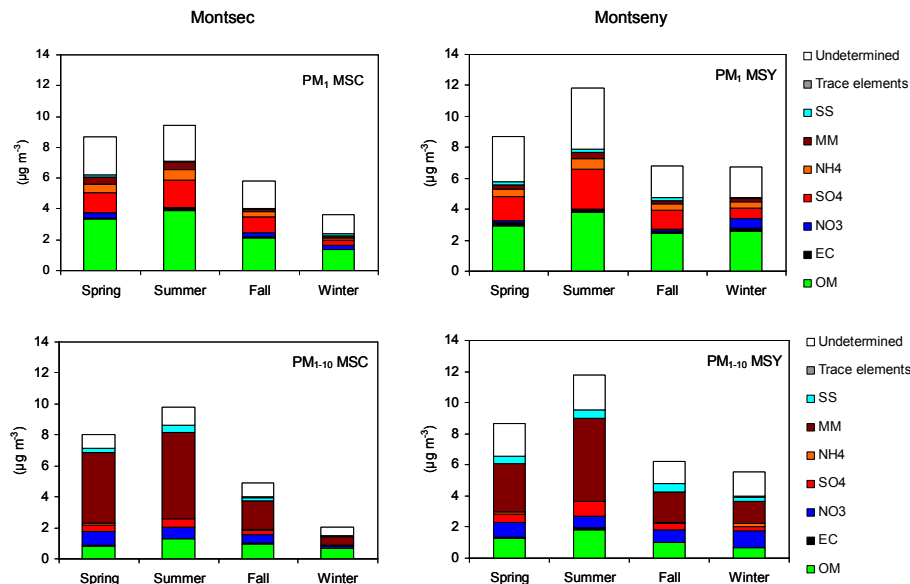


Figure 1. Average concentrations ($\mu g m^{-3}$) of aerosol major components and trace elements in PM_{10} and PM_{1-10} fractions at Montsec and Montseny for the different seasons based on daily measurements between January 2010 and March 2013.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

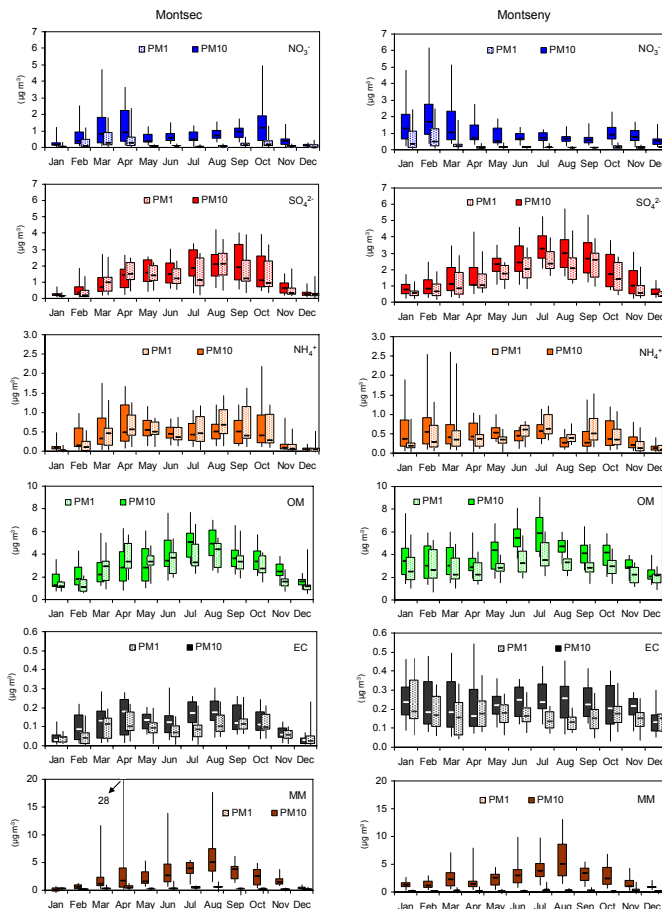


Figure 2. Monthly median (black line within the boxes) and percentiles (5-25-75-95, boxes and whiskers) of major PM₁ and PM₁₀ chemical components concentrations at Montsec and Montseny based on daily measurements between January 2010 and March 2013.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

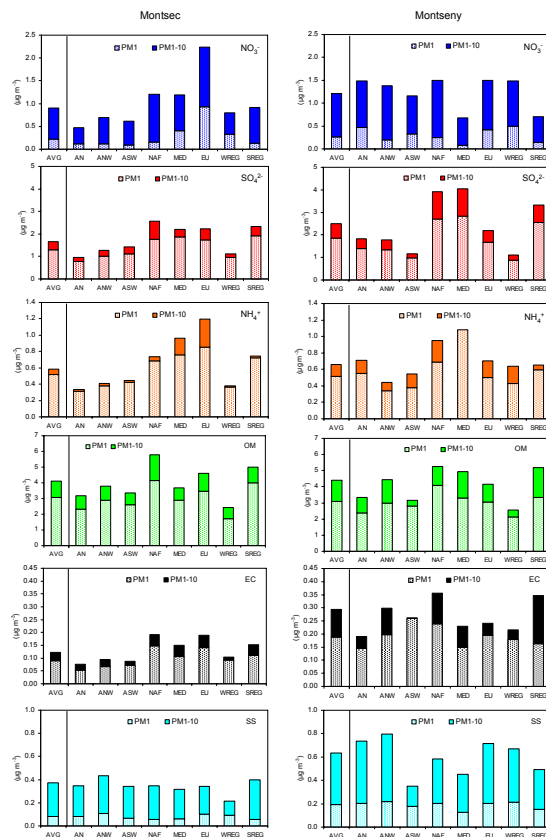


Figure 3. Average (AVG) concentrations of PM_{10} and PM_{1-10} nitrate (NO_3^-), sulfate (SO_4^{2-}), ammonium (NH_4^+), elemental carbon (EC), organic matter (OM) and sea salt (SS) at Montsec and Montseny for different air mass origins based on daily measurements between January 2010 and March 2013.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

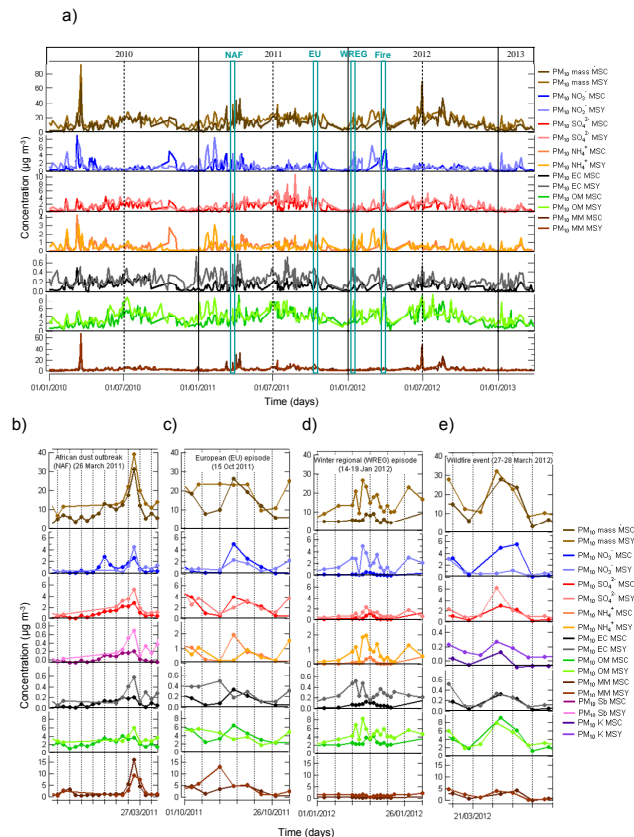


Figure 4. (a) Time series of daily PM₁₀ mass and major PM₁₀ chemical components concentrations at Montsec (MSC) and Montseny (MSY) between January 2010 and March 2013. Green bands indicate 4 examples of different episodes affecting the study area. Zoom of the 4 selected episodes (b) African dust outbreak, (c) European episode, (d) winter regional episode, and (e) wildfire event, with daily PM₁₀ mass and PM₁₀ chemical components concentrations.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

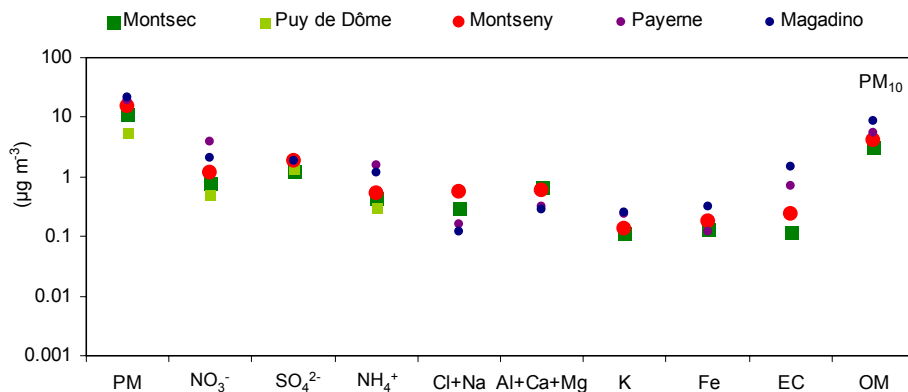


Figure 5. Average concentrations ($\mu\text{g m}^{-3}$) of PM_{10} major compounds at Montsec, Puy de Dôme (Bourcier et al., 2012), Montseny, Payerne and Magadino (Gianini et al., 2012).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

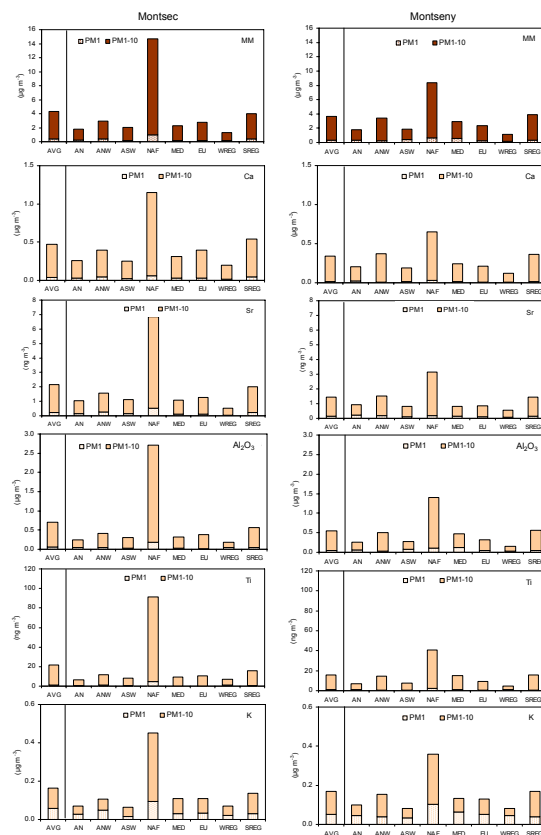


Figure 6. Average (AVG) concentrations of PM₁ and PM₁₋₁₀ mineral matter (MM), calcium (Ca), strontium (Sr), aluminium oxide (Al₂O₃), titanium (Ti) and potassium (K) at Montsec and Montseny for different air mass origins based on daily measurements between January 2010 and March 2013.

Aerosols in
continental and
regional background
in the Western
Mediterranean

A. Ripoll et al.

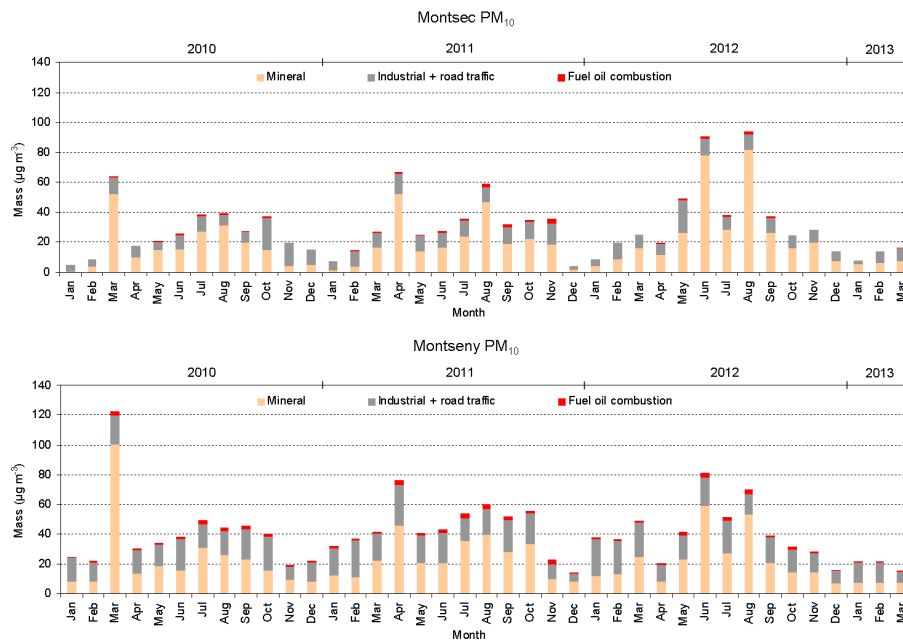


Figure 7. Monthly average concentrations of trace element groups at Montsec and Montseny based on daily measurements of PM₁₀ between January 2010 and March 2013.

Aerosols in continental and regional background in the Western Mediterranean

A. Ripoll et al.

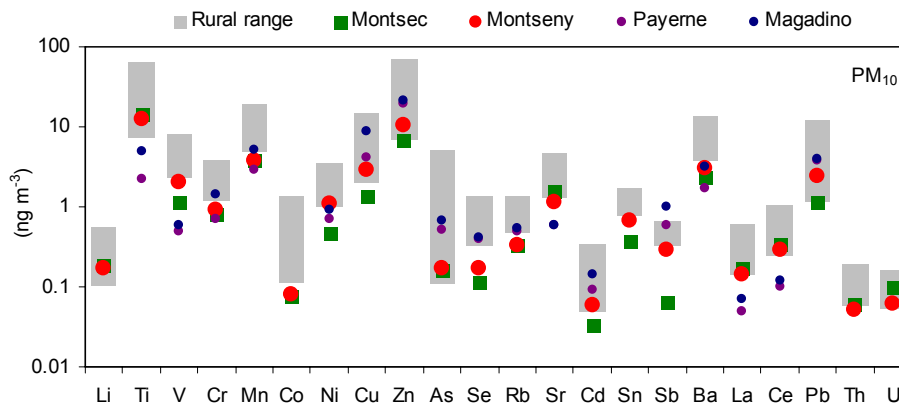


Figure 8. Average concentrations (ng m^{-3}) of PM_{10} trace elements at Montsec, Montseny, Payerne and Magadino (Gianini et al., 2012), and range of Spanish rural concentrations (updated from Querol et al., 2007).

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Printer-friendly Version	
Interactive Discussion	

