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African dust outbreaks over the Mediterranean Basin during 2001–2011: PM₁₀ concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology

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**African dust
outbreaks over the
Mediterranean Basin
during 2001–2011**

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Abstract

The occurrence of African dust outbreaks over the whole Mediterranean Basin has been identified on an 11-yr period (2001–2011). In order to evaluate the impact of such mineral dust outbreaks on ambient concentrations of particulate matter, PM₁₀ data from regional and suburban background sites across the Mediterranean area were compiled. After identifying the daily influence of African dust, a methodology for estimating natural dust contributions on daily PM₁₀ concentrations was applied.

Our results reveal that African dust outbreaks occur with much higher frequency in southern areas of the Mediterranean, from 30 to 37 % of the annual days, whereas they take place less than 20 % of the annual days in northern sites. The central Mediterranean emerges as a transitional area, with slightly higher frequency of dust episodes in its lower extreme when compared to equivalent areas in western and eastern sides of the Basin. A decreasing south to north gradient of African dust contribution to PM₁₀ is patent across the Mediterranean. Our study demonstrates that this gradient may be mainly explained by the latitudinal position. A longitudinal increasing trend of African dust contribution to PM₁₀ is also observed from 25° E eastwards, and is due to the annual occurrence of intense dust episodes. Thus, the slightly higher frequency of African dust episodes over the lower part of Central Mediterranean is compensated by its moderately lower intensity. Concerning seasonality patterns and intensity characteristics, a clear summer prevalence is observed in the western part, with low occurrence of severe episodes (daily dust averages over 100 µg m⁻³ in PM₁₀); no seasonal trend is detected in the central region, with moderate-intensity episodes; and significantly higher contributions are common in autumn-spring in the eastern side, with yearly occurrence of various severe episodes. Overall, African dust emerges as the largest PM₁₀ source in regional background southern areas of the Mediterranean (35–50 % of PM₁₀), with seasonal peak contributions to PM₁₀ up to 80 % of the total mass.

The multi-year study of African dust episodes and their contributions to PM₁₀ concentrations allowed us to identify a consistent decreasing trend in the period 2006/2007

ACPD

12, 28195–28235, 2012

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

to 2011 in 4 of the 17 studied regions, all of them located in the NW of the Mediterranean. The observed trend is almost parallel to the NAO (North Atlantic Oscillation) index for the summer period, progressively more negative since 2006 onwards. As a consequence, a sharp change in the atmospheric circulation over the last 5 yr (a similar negative NAO period occurred in the 1950 decade) have affected the number of African dust episodes and their mean contribution to PM₁₀ in the NW part of the Mediterranean. The investigation of summer temperatures at 850 hPa suggest that warm air accomplishing African dust air masses moved anomalously through the central Mediterranean in the 2007–2008 period, whereas it was displaced atypically to the NW African coast and the Canary Islands in the 2009–2011 period.

1 Introduction

On a global scale, most of the atmospheric particles are emitted by natural sources, being mineral dust the second more abundant component after sea-spray derived aerosols (IPCC, 2007). These crustal aerosols are mainly released to the atmosphere from arid and semiarid regions located in sub-tropical areas in the Northern Hemisphere, being the Sahara-Sahel-Chad dust corridor in North Africa the largest source region (Prospero et al., 2002; Moreno et al., 2006).

In general, atmospheric circulation over north-western Africa is mainly controlled by the northeast trade winds and by the mid-tropospheric Saharan Air Layer. Southerly of the Saharan deserts, winds are usually mono-directional with a general westward transport. Along the year, the dust plume extension varies accordingly to the displacement of the Inter-Tropical Convergence Zone (ITCZ) (Prospero et al., 1981). In winter, when the ITCZ is at its southernmost position, dust originated in Sahara and Sahel deserts is transported towards the tropical Atlantic Ocean by such northeast trade winds (Alonso-Pérez et al., 2011). In summer, such trade winds more constrained owing to the northern displacement of the ITCZ (Prospero et al., 1981). Additionally, high insolation and temperatures over Sahara-Sahel area create strong surface winds and

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

large-scale convection processes, which lift dust particles at high atmospheric levels (up to 5 km). Such mineral dust particles, transported at high-altitude by the Saharan air layer, move partially towards the tropical Atlantic above the boundary layer (Bergametti et al., 1989b). A significant amount of the atmospheric dust is transported also towards the western Mediterranean describing an anticyclonic gyre over north-western Africa. Regardless of most of the African dust particles are exported westwards over the Atlantic (Viana et al., 2002; Alastuey et al., 2005; Alonso-Pérez et al., 2011), travelling for long distances and impacting very distant areas in the Caribbean and the United States (Arimoto et al., 1997; Prospero et al., 2002), a considerable amount of dust is also released northerly, affecting the Mediterranean region (Ganor and Mamane, 1982; Bergametti, et al., 1989a; Guerzoni, and Chester, 1996; Querol et al., 1998; Rodriguez et al., 2001; Escudero et al., 2005, Kallos et al., 2006; Mitsakou et al., 2008; Gerasopoulos et al., 2006; Koçak et al., 2007) and even other European areas (Klein et al., 2010).

African dust towards the Mediterranean region is usually mobilized by a number of meteorological scenarios widely described elsewhere (Rodriguez et al., 2001; Escudero et al., 2005, Gkikas et al., 2009, 2012). As summarized in Querol et al. (2009a), dust-storms affecting western and central Mediterranean are caused by low-pressure systems over the Atlantic or North Africa, high pressures over the Mediterranean, or high pressures at upper levels over NW Africa. Dust storms over the eastern Mediterranean are generally originated by cyclones moving eastwards throughout the Mediterranean, but also because of the combination of a low-pressure over North Africa with high pressures over Middle East. Details on these meteorological scenarios may be found in Escudero et al. (2005) for the western and central scenarios and in Kallos et al. (2006) for the eastern ones.

One of the most important health outcomes of African dust concerns its chemical and biological composition, as highlighted in a recent review study (Karanasiou et al., 2012). The meteorological scenarios favouring the export of African dust to the western or eastern Mediterranean implies that mineral particles emitted in regions located

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



in East Africa, such as the Bodele Depression, hardly reach western areas in the Mediterranean. Conversely, dusts from NW deserts usually affect eastern locations of the Mediterranean because of the sweep effect caused by low-pressure movement. Taking into account that significant differences in natural soil composition are observed from one region to another in North Africa (Moreno et al., 2006), variations in anthropogenic pollutants travelling with mineral dust are also observed (Perrino et al., 2010; Rodríguez et al., 2011) and the content in micro-organisms may be different, the potential effects on health (Pérez et al., 2008; Polymenakou et al., 2008; Tobías et al., 2011a), ecosystems (Arimoto, 2001) and climate (IPCC, 2007) may vary notably.

Yearly, variations in mean ambient temperature or rainfall amount are observed elsewhere. These changes are associated to alterations in the atmospheric circulation. Thus, it is expected that such variations in the atmospheric dynamics affect also other phenomena such as dust mobilization frequency and/or intensity. The study of long data series of dust contributions at multiple points across a wide area may reflect periodic or consistent tendencies. As an example, Cusack et al. (2012) have observed a clear decreasing trend in a number of components of $PM_{2.5}$ at a regional background site in NE Spain that is linked to the implantation of abatement measures in that region together with meteorological changes. Such meteorological changes or cycles essentially explain most of the decrease in PM levels observed North to South in western and central Europe (Barmpadimos et al., 2012; Cusack et al., 2012).

Bearing in mind that African dust is an important source of particulate matter pollution in specific areas, causing by itself or contributing to exceed the daily limit values of PM_{10} (in Europe being established in $50 \mu g m^{-3}$ by the 2008/50/EC Directive), and exerting negative health outcomes (Middleton et al., 2008; Kitsakou et al., 2008; Pérez et al., 2008; Jiménez et al., 2010; Mallone et al., 2011; Sajani et al., 2011; Samoli et al., 2011a, b; Tobías et al., 2011a, b), the identification of such episodes and the quantification of daily and annual contributions is currently necessary. Moreover, the identification of temporal trends in African dust contributions may be indicative of atmospheric changes.

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Previously, Querol et al. (2009a) performed a comprehensive work on African dust significance over the Mediterranean from a database of up to 6 yr for a few sites. In this work we have interpreted a much longer database of PM levels and African dust contributions (up to 11 yr) at multiple locations (17 areas with at least one monitoring site in each one) across the Mediterranean. The aim of our work was to characterize the phenomenology of African dust outbreaks across the Mediterranean, with special interest in identifying the significance of this natural PM source on ambient air PM₁₀ concentrations by studying daily and seasonal patterns. Moreover, an extended database of African dust episodes across the whole Mediterranean together with their contributions to PM₁₀ allows a confident study on inter-annual trends.

This study is part of a European project (MED-PARTICLES, Particles size and composition in Mediterranean countries: geographical variability and short-term health effects) financed by the LIFE program. Overall the MED-PARTICLES project aims at quantifying short-term health effects of particulate matter over the Mediterranean region by distinguishing different particle sizes, chemical components and sources, with special emphasis in the effects derived from African dust. Since the main motivation of the project is in evaluating health effects, the results of this study are crucial from an epidemiological point of view. These results will be used to estimate effects on health distinguishing between PM₁₀ from North Africa and that from local and/or regional sources.

2 Methodology

2.1 Data collection

To evaluate African dust contributions across the Mediterranean Basin (not including North African sites) and to assess on their impact on PM₁₀ levels, data from 19 regional background (RB) and sub-urban background (SUB) sites were obtained (Fig. 1): 7 in Spain covering central and eastern Iberia, and the Balearic islands; 2 in southern

France; 5 in Italy covering north to south of the peninsula, Sardinia and Sicily; 1 in Bulgaria; 3 in Greece including 2 in the continent and 1 in Crete; and 1 in Cyprus. Thus, these RB sites were distributed west to east and north to south of the Mediterranean region, as shown in Fig. 1. From all these regional background sites daily PM₁₀ concentrations have been obtained from 2001–2011 when available. In Table 1 it is shown a summary of station characteristics, data coverage, and measurement principles.

All the data used in this study were obtained from public European databases: Airbase (<http://acm.eionet.europa.eu/databases/airbase/>), EMEP (www.emep.int/) and EUSAAR (<http://www.eusaar.net/>).

2.2 African dust occurrence

The methodology used for identifying African dust episodes is akin to that used successfully in numerous studies (Rodríguez et al., 2001; Escudero et al., 2005 and 2007; Querol et al., 2009a; Pey et al., 2010). This procedure assures the identification of almost all the African dust episodes, independently of their intensity, and consists in the interpretation of a couple of tools: meteorological products (NCEP/NCAR : <http://www.esrl.noaa.gov/psd/data/composites/hour/>), aerosol maps (BSC-DREAM: <http://www.bsc.es/projects/earthscience/DREAM/>; NAAPS-NRL: <http://www.nrlmry.navy.mil/aerosol/>; SKIRON: <http://forecast.uoa.gr/dustindx.php>), satellite images (SeaWiFS: <http://oceancolor.gsfc.nasa.gov/SeaWiFS/>; MODIS: <http://modis.gsfc.nasa.gov/>), and air masses back-trajectories (HYSPLIT: http://ready.arl.noaa.gov/HYSPLIT_traj.php). It is important to remark that, in some cases when dust air masses are travelling at high altitude, African dust may affect PM levels at ground levels up to 2 days after the episode ends as reflected in the European Guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe (http://ec.europa.eu/environment/air/quality/legislation/pdf/sec_2011_0208.pdf). Thus, a final evaluation of PM levels at the different RB and SUB sites was conducted, which incorporated these possible delays.

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

2.3 African dust contribution to PM₁₀ concentrations

In order to ascertain on daily African dust contributions to PM₁₀ and PM_{2.5} concentrations, a statistical methodology applied to the PM data series have been used. This method is based on the application of 30 days moving 40th percentile to the PM₁₀ or PM_{2.5} data series, after excluding those days impacted by African dust. For those days affected by African dust it is obtained a percentile value which is assumed to be the theoretical background concentration of PM if African dust didn't occur. After that, the African dust contribution is obtained by difference between the experimental PM₁₀ or PM_{2.5} concentration value and the calculated 40th percentile value.

This methodology was preliminary published (Escudero et al., 2007) considering the 30th percentile. Subsequently this method was slightly modified by adopting the 40th percentile instead the 30th one only for conservative reasons. Currently, this is one of the official methods adopted by the European Commission for evaluating the occurrence of African dust outbreaks and quantifying its contributions (Commission staff working paper establishing guidelines for demonstration and subtraction of exceedances attributable to natural sources under the Directive 2008/50/EC on ambient air quality and cleaner air for Europe). It is important to remark that the feasibility of this method was demonstrated by comparing experimentally measured concentrations of mineral matter determined at three Spanish RB sites versus the estimated African dust contributions given by this procedure.

3 Results and discussions

3.1 PM levels across the Mediterranean

Annual averages of PM₁₀ at RB sites across the Mediterranean Basin (note that monitoring sites in North Africa were not studied) reflect a wide spatial variability (Fig. 2), with the highest concentrations in eastern-basin insular areas (21–24 µg m⁻³). On the

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



contrary, the lowest PM₁₀ concentrations (9–11 µg m⁻³) are observed at high-altitude sites West to East of the Mediterranean: San Pablo de los Montes, EMEP site in central Spain at 1241 m a.s.l.; Febbio, RB site in northern Italy at 1020 m a.s.l.; and Rojen Peak, EMEP site in the Rhodopes (Bulgaria) at 1750 m a.s.l. Intermediate PM₁₀ concentrations are recorded in the rest of RB sites, being sensibly higher when closer to densely populated and/or industrialized areas, and to North Africa. This is the case of: Fontechiari, in close proximity to Rome (20 µg m⁻³); Víznar, near Granada and North Africa (18 µg m⁻³); Lecce, in southern Italy (17 µg m⁻³); Drome Rurale Sud, close to the highly industrialized area of Marseille (16 µg m⁻³); and Montseny, in the vicinity of the Barcelona metropolitan and industrial agglomeration (15 µg m⁻³). Concerning average PM₁₀ concentrations at the suburban environments used in this study (selected because of the lack of RB sites covering that geographical areas), they are much higher in the Athens and Thessalonica influence areas (27–28 µg m⁻³) than in Sicily or Mallorca (18 µg m⁻³). See Fig. A1 to appreciate average PM₁₀ levels without the influence of African dust contributions.

Overall, there is an increasing PM gradient (occasionally broken as in the vicinity of Barcelona, Marseille and Rome, because of the high influence of anthropogenic emissions) from the NW to the SE of the Mediterranean. This augment is partially coincident with higher African dust contributions, but also to the increase of the regional pollution towards the eastern part of the Basin. This increment was formerly reported and chemically characterized by Querol et al. (2009a, b). In those studies, they found higher concentrations of sulphate and carbonaceous aerosols easterly in the Basin, both components with a prevalent anthropogenic origin at these areas.

3.2 PM levels across the Mediterranean: seasonal patterns

PM levels show clear seasonal patterns across the Mediterranean (Fig. 3). In general, a summer maximum is observed throughout the basin attributed to a number of factors including: (1) reduced dispersive conditions over the region; (2) enhanced formation of

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

secondary pollutants owing to intense solar radiation and humidity; (3) high frequency of wildfires in Mediterranean and surrounding areas; (4) elevated anthropogenic pressure since the Mediterranean is a common tourist destination; (5) higher shipping emissions owing to the increase in cruise traffic during the warm season. Specifically in the western side of the basin, higher frequencies of Saharan dust outbreaks (Querol et al., 1998; Rodríguez et al., 2001) and the effect of recirculation of air masses over that area (Millán et al., 1997; Rodríguez et al., 2002; Pey et al., 2009) contribute to increase the background levels. Likewise, the transport of polluted air masses from eastern-Europe accounts for such summer increase in the eastern part (Gerasopoulos et al., 2006; Koulouri et al., 2008). Similarly at both sides, the lowest PM concentrations are observed in December-January, coinciding with well-ventilated conditions, low photochemical activity and less frequency of Saharan dust episodes. The most noticeable difference between western and eastern sides corresponds to the late winter-early spring period, when PM concentrations in the eastern Mediterranean are at their maximum due to the impact of severe African dust outbreaks, whereas they are low or intermediate in the western part (it is usually a rainy season over this area).

3.3 African dust outbreaks: frequency

Figure 4a shows the average frequency of African dust outbreaks across the Mediterranean Basin during the period 2001–2010. It is evident the decreasing gradient of African dust outbreaks frequency South to North of the Mediterranean Basin. Among the investigated areas, the lowest frequencies of African dust events are observed in central and NE Spain, SE France and Northern Italy (17–18 %). On the contrary, the highest frequency is recorded in Sicily (37 %), followed by Cyprus (34 %). As a result, a similar frequency of African dust is observed in south-western and eastern Iberia (23–24 %) and in central Italy and northern Greece (24 %). Furthermore, there is a linear relation between mean frequency of African dust outbreaks and latitude (Fig. 4b). The linear relation is almost the same for areas located in the eastern or western part of the Mediterranean, whereas it is slightly different for the central part

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



of the Basin, where slightly higher frequency of dust episodes is observed in southern sites with respect to similar latitudinal points at both extremes of the Basin. This fact is directly related with the phenomenology of African dust episodes. As reviewed in Querol et al. (2009a), the western Mediterranean is more affected by African dust in summer. In contrast, the eastern Mediterranean is frequently impacted by African dust air masses in the autumn-spring period. Summer episodes usually affect the central part of the Mediterranean, especially from central Italy towards the South. Similarly, autumn-spring episodes impact often South Italy and Sicily. Thus, the central Mediterranean may be considered as a transitional area which is impacted in its lower part by African dust all over the year. An additional issue concerns the African dust export towards the Mediterranean. The transport of African dust towards the western Mediterranean mostly occurs at relatively high atmospheric levels, exported from NW African deserts (Escudero et al., 2005). Over the eastern Mediterranean, however, the transfer of African dust air masses take place at surface levels (Fig. A2). These features influence the intensity of the African dust outbreaks at both sides of the basin, as shown in following sections.

3.4 African dust outbreaks contributions to ambient air PM₁₀

3.4.1 Average PM₁₀ concentrations

After identifying daily occurrence of dust outbreaks, the methodology described in Sect. 2.3 has been applied in order to estimate dust contributions to PM₁₀. Figure 5 shows the average African dust contributions to the mean ambient air PM₁₀ levels calculated for the existing periods (in most cases a 10–11 yr database was available). As seen in Fig. 5, African dust inputs are considerably higher in the eastern locations of the Mediterranean when compared to those observed in the western side. In general, average annual concentrations were found to be maximum in Cyprus and Greek Islands (7.3–8.4 µg m⁻³), and minimum in NE Spain, SE France and North Italy (1.1–1.5 µg m⁻³). In order to evaluate properly the factors governing this spatial distribution,

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



a crossover study between average African contributions versus latitude and longitude has been conducted (Fig. 6a, b). As seen in Fig. 6a, there is a clear dependence of African dust contribution in PM₁₀ with latitude. It is remarkable that this relation is not lineal but exponential, being defined by the following experimental Eq. (1)

$$y = 2E + 13x^{-8.007} \quad (1)$$

where y is the estimated African dust (in $\mu\text{g m}^{-3}$), and x is latitude (in decimal units).

Thus, for a given latitude across the Mediterranean Basin, the expected average African dust contribution may be calculated by applying this experimental equation. It is important to highlight that this equation is only applicable to estimate average annual concentrations for long periods because, as shown in next sections, annual contributions of African dust may experience significant inter-annual variations.

Complementarily, the effect of the longitudinal position with respect to the average frequency of African dust outbreaks has been evaluated (Fig. 6b). Despite that slight differences have been found between the position within the Basin and the frequency of African dust (Fig. 4b, Fig. 6b), locations situated at the same latitude and different longitude across the Mediterranean register on average similar African dust concentrations (Fig. 6c) if they are between 10° W and 25° E. At more eastern longitudes (> 25° E) it is evident a higher African dust contribution (Fig. 6c), having in mind that the frequency of African dust outbreaks does not increment. This is strongly related with the severity of some African dust episodes in the eastern part of the Basin, as mentioned in following sections.

Overall, mean annual African dust contributions in PM₁₀ varied with respect to the latitude. A longitudinal effect is patent from 25° E eastwards. The decreasing gradient of African dust towards the North is not lineal but exponential.

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

3.4.2 Seasonal patterns

As mentioned previously, African dust transport occurs in different seasons in western and eastern sides of the Mediterranean. These seasonal patterns exert a clear influence in the African dust contributions along the year.

Figure 7 shows the seasonal contributions of African dust on PM_{10} , and the partitioning between bulk ambient air PM_{10} and African dust. On average, African dust may occur all along the year across the Mediterranean. However, African dust inputs in the western side of the Mediterranean are considerably higher between May and October, and in March, when compared to the rest of the year. On the contrary, such inputs are clearly higher between November and May in the eastern part of the Mediterranean. An intermediate outcome is observed for central locations in the Mediterranean, where only slightly higher summer contributions are detected.

In relative terms, African dust accounts for 50 % of PM_{10} in Sicily, between 35 and 39 % in SE Spain, Greek Islands and Cyprus, between 17 and 26 % in SW, eastern and central Spain, the Balearic Islands, Sardinia, Bulgaria and South of Greece, and between 10 and 16 % in the rest of the areas. These percentages experience large variations along the year. As a result, in the south-western part of the Mediterranean African dust may account for about 50 % of PM_{10} mass in mid summer, and less than 10 % in winter. Similarly, in the southern-central side of the basin between 55 and 65 % of the PM_{10} in summer is mineral African dust. More evident is the impact of the African dust in the south-eastern locations of the Mediterranean, where up to 80 % of the PM_{10} recorded in the period February–April is given by external mineral dust. By contrast, mineral dust from desert regions account for less than 10 % of PM_{10} in mid summer. The mentioned trends are repeated in the northern areas across the basin, although a substantial diminution in the dust contributions on ambient PM_{10} concentrations is evident.

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

3.4.3 Intensity of dust outbreaks

One of the most interesting aspects of African dust episodes concerns to their intensity. By analyzing extended databases of African dust events across the Mediterranean it is possible to identify the occurrence and recurrence of intense African dust episodes.

5 Figure 8 represents the percentage of African dust days according to their intensity (mean contributions in $\mu\text{g m}^{-3}$ in 9 concentration ranges, the highest being 30–44, 44–99 and > 100) for the whole Mediterranean. The occurrence of extreme dust events (with daily PM_{10} African dust contributions higher than $100 \mu\text{g m}^{-3}$) with respect to the total number of episodes is infrequent. However, the occurrence of such severe
10 outbreaks is relatively frequent (2–5 % of the African dust days) in the south-eastern part of the Basin (Greek Islands, South of Greece and Cyprus). As a result, some of these events occur randomly every year. The occurrence of extreme events is much reduced (< 1 –1 % of the African dust days) in equivalent (in latitude) locations west-erly of the Mediterranean (southern Spain, southern Italy, Sicily). Moderate to intense
15 events (30 – $99 \mu\text{g m}^{-3}$ PM_{10} dust) are observed both in western, central and eastern areas, accounting for 15 to 25 % of the dust events in southern areas, 10–15 % in inter-mediate latitudes, and 5–10 % in northern areas. Low-intensity episodes (1 – $10 \mu\text{g m}^{-3}$) prevail in northern locations of the western and central Mediterranean, accounting for 50 to 70 % of the African dust episodes.

20 Overall, the intensity characteristics described in this section are strongly related with the transport patterns of African air masses. Dust transfer over western and eastern sides of the Mediterranean is caused by different transport mechanisms. In addition, the Atlas mountainous barrier, with a 2500 km extension from western Sahara towards Tunisia and peak altitudes up to more than 4000 m a.s.l., plays a dominant role in local and meso-scale atmospheric circulation patterns. As a result, African dust episodes
25 over the western and central part of the Mediterranean are very frequent in summer (Rodríguez et al., 2001; Escudero et al., 2005), although moderate in intensity given the intricate transport mechanisms. The situation in the eastern part of the Mediterranean

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



is very different. African dust transport is typically induced by cyclones moving eastwards across the Mediterranean and/or North Africa, transporting dust at surface levels. These flows may be enhanced during specific scenarios (Moulin et al., 1998) if air masses are canalized southerly of the Atlas Mountains (with a north-eastern prevalent direction), giving to the occurrence of short but intense dust episodes in the eastern side of the Mediterranean. In addition, dust plumes leaving north-African deserts eastwards of Tunisia do not reach elevated atmospheric layers (Nastos, 2012) since they do not find any significant geographical barrier.

In order to assess on the relation between African dust occurrence and intensity, Fig. 9 has been elaborated. Most of the patterns observed in Fig. 9 have been already discussed in previous paragraphs. Nevertheless, there are new specific features to be described and interpreted. In general, there is no clear relationship between occurrence and intensity, with significant seasonal distribution of dust episodes but flat variation in the intensity along the year. Only in the eastern side of the Mediterranean, intensity and occurrence are varying in the same way. Concerning intensity, although flat variation is observed, February–March and October–November episodes over the western and central Mediterranean are usually more intense than those of other months. This fact is due to the transport mechanisms of the dust, always at ground level, and induced by two main scenarios already described in Escudero et al. (2005): (1) cyclone systems moving from western Iberia towards the Mediterranean; (2) widespread anticyclone over the western and central Mediterranean that moves slightly easterly. Commonly, the first scenario is typical of autumn, whereas the second is frequently observed in late autumn and early spring.

3.5 African dust outbreaks: inter-annual variability

Long-term series of annual averages of African dust contributions are helpful to recognize cyclic or climatic tendencies, which would be related to fast changes in atmospheric circulation patterns, as African dust is usually mobilized by specific meteorological mechanisms. Also variations in the emissions from source areas could be

recognized by exploring these long-term databases. Figure 10 summarizes the annual averages of African dust contributions from 2001 to 2011 for selected areas across the Mediterranean. Although it was not possible to find a very complete PM₁₀ database for the whole Mediterranean to quantify the African dust content, usual data coverage ranges between 7 to 11 yr.

In general over central and southern areas (central and southern Spain, central Italy and Sicily, Greece, and Cyprus), there are no evident increasing neither decreasing trends of African dust contribution to ambient PM₁₀, with sporadic annual peaks as a result of the occurrence of severe episodes. Thus, South Spain in 2004 registered a number of episodes above 150 µg m⁻³; Sicily also recorded a very intense event in February 2004, with a daily concentration of more than 300 µg m⁻³ of African dust; South Greece experienced also severe African dust episodes in February, May and November 2004, with daily peak concentrations up to 200 µg m⁻³; Cyprus suffered numerous and very intense dust episodes in February–March, August and December 2008, 12 days with dust daily concentrations exceeding 100 µg m⁻³.

The situation in the northern locations of the western and central Mediterranean (NE Spain, the Balearic Islands, SE France, and North Italy) is clearly different. In these areas, a prominent decreasing trend on ambient PM₁₀ contributions is patent from 2007 onwards. Before 2007, alternate contributions are observed, with annual peaks caused by intense episodes. From 2007, a marked and continuous decreasing trend is observed in these areas. Because this trend is not observed in the southern areas, a change in the emissions of dust from source regions is not feasible. Nevertheless, a modification in the atmospheric circulation over these areas may be possible. Actually, a recent study on evaluating trends of PM and chemical composition across Europe (Cusack et al., 2012) has demonstrated a partial relation between the observed trends and the North Atlantic Oscillation (NAO) index. The NAO index is accounting for the intensity of the westerly circulation over the North Atlantic (Barnston and Livezey, 1987) that is mainly defined by the position of the two leading atmospheric systems, the Azores high and the Scandinavian low. This NAO index is widely used to interpret

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

wintertime weather anomalies in temperature and precipitation over northern, central and western areas across Europe and the United States (Hurrell et al., 2003). In our case we have calculated the NAO index for the summer period, when the highest frequency of African dust outbreaks is detected in those areas where the decreasing trend is obvious. As seen in Fig. 11a there is a clear change in NAO index from 2006–2007 onwards. In the period 2001–2005, summer NAO indexes were close to 0 whereas there became consistently more negative from 2006–2007 to be at their lowest in 2011 (–0.90). A wider temporal scale of this summer NAO index (Fig. 11b) allows identifying only one persistent negative period in the 50's. Thus, it is evident that the last 5-year summer period has been governed by a peculiar atmospheric pattern. When contrasting the summer NAO index for the period 2006/2007–2010/2011 with respect to the average annual African dust contributions to PM₁₀ levels for NE Spain, SE France, the Balearic Islands and North Italy, a high correlation is observed (Fig. 12). The correlation observed for these areas is higher in the Spanish areas ($R^2 = 0.9$) than in SE France ($R^2 = 0.8$) or North Italy ($R^2 = 0.5$), thus indicating a progressive lesser influence of the summer NAO index towards the East. Negative NAO phases are related to a displacement of the storm trajectories towards the South. Generally in summer, westerly winds blow above the Alps, leaving the Mediterranean region under weak gradient conditions (Millán et al., 1997). During negative NAO phases the track of westerly winds is observed at lower latitude, thus preventing subtropical air masses to reach the NW Mediterranean. In order to address on the influence of African dust air masses over summer periods, the temperature at 850 hPa (approximately at 1500 m a.s.l.) has been calculated from 2001 to 2011 (Fig. 13). It is clear in this Fig. 13 that elevated temperatures at 850 hPa are emerging from North Africa, accomplished with mineral dust particles. In many cases from 2001 to 2006 these warm air masses attained the western Mediterranean. In 2007 and 2008 it is observed a displacement of such warm air towards the central Mediterranean, although still affecting in 2007 the NE Spain and the Balearic Islands. From 2009 to 2011 North-African warm air masses affected the Iberian Peninsula progressively with lower extent at the same time that such warm

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



air was essentially located over the NW side of the African continent and the Canary Islands.

4 Conclusions

A comprehensive study on African dust outbreaks occurrence across the Mediterranean Basin and on the influence of such natural source on ambient air PM₁₀ levels has been completed. The results of this work reinforce those previously reported by Querol et al. (2009a). On the other hand, this work adds new findings that are summarized and discussed in this section:

1. African dust outbreaks across the Mediterranean occur in different seasons and are caused by distinct meteorological scenarios western to eastern of the Mediterranean, which condition their intensity. Intricate and slow-moving transport mechanisms (convective injection of dust in North Africa coupled with anticyclonic conditions at upper atmospheric levels) dominate African dust occurrence, higher in summer, in the western Mediterranean. On the contrary, conventional transport scenarios (low pressure systems) provoke a rapid transport of dust towards the eastern Mediterranean, usually from autumn to spring. An intermediate situation occurs in the central part of the Basin, resulting in no significant seasonality but slightly higher number of events in its lower part since is affected marginally by a number of dust outbreaks covering western and eastern parts of the Basin. Accordingly, a number of severe episodes are annually recorded in the eastern part of the Mediterranean, whereas these are scarce or absent in the central and western part.
2. Mean African dust contributions to ambient PM₁₀ concentrations across the Mediterranean Basin are varying according to the latitude. A longitudinal effect is patent from 25° E eastwards; with sensibly higher concentrations of African dust

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

in the eastern side of the Basin. Such eastward trend is caused by the occurrence of 2–5 annual severe dust episodes over this area.

3. African dust may represent up to 50 % of PM_{10} , as in Sicily, and around 35–40 % of PM_{10} in SE Spain, Greek Islands and Cyprus. Thus, such inputs are considered to be the largest PM_{10} source affecting the regional background of the southern part of the Mediterranean Basin. Moreover, these African dust contributions may constitute up to 80 % of the PM_{10} in the dusty period of the mentioned areas.
4. Significant inter-annual variations in African dust contributions are observed in the NW of the Mediterranean from 2006/2007 onwards. Such variations are perfectly correlated with the summer NAO index, exceptionally and consistently more negative from 2006 to 2011. Only a similar negative period was observed in the 1950 decade. The averaged temperatures calculated at 850 hPa for the summer periods from 2001 to 2011 reveal a dislocation of the subtropical warm air (coupled with mineral dust) towards the central Mediterranean in 2007–2008, and towards the NW part of Africa and the Canary Islands in 2009–2011.

Appendix A

It is well known that African dust exerts an important influence on PM concentrations over some of these areas (Querol et al., 2009a). In order to compare the RB PM_{10} concentrations avoiding such natural contributions, we have subtracted the African dust estimated in PM_{10} (Fig. A1). Similarly, the high-altitude sites register the lowest PM_{10} concentrations ($7\text{--}9\text{ }\mu\text{g m}^{-3}$). Immediately afterwards, western Mediterranean sites show intermediate PM concentrations ($10\text{--}11\text{ }\mu\text{g m}^{-3}$). Subsequently, central and eastern Mediterranean locations display higher concentrations ($13\text{--}15\text{ }\mu\text{g m}^{-3}$). Accordingly, there is a PM_{10} increase of $3\text{--}4\text{ }\mu\text{g m}^{-3}$ from western towards central and eastern locations across the Mediterranean. Finally, the highest PM_{10} concentrations

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



(13–18 $\mu\text{g m}^{-3}$) correspond to those RB sites located in areas subjected to severe anthropogenic influence (Rome, Marseille and Barcelona).

As described in Querol et al. (2009a), there are a number of meteorological scenarios favouring the African dust transport towards the Mediterranean region. Among these scenarios, two of them are responsible for more than 50 % of the African dust outbreaks accounted in the period 2001–2010: (1) transport of African dusty air masses at relatively high atmospheric levels, principally in summer, and impacting over the western Mediterranean; (2) African dust export at surface levels, frequently in from October to May, and impacting commonly over central and eastern of the Mediterranean basin. As seen in Fig. A2, these 2 scenarios are repeated every year. The spring scenario is characterized by low pressures over central and eastern of the Mediterranean, giving to the transport of African air masses towards the central and eastern of the Mediterranean. The summer scenario is evident at upper atmospheric levels, usually above 1500 m a.s.l., characterized by high pressures over Tunisia at such atmospheric layers. This meteorological configuration favours the mobilization of African dust (previously injected to the atmosphere as a consequence of the intense convective dynamics) towards western and central of the Mediterranean.

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African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

References

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ACPD

12, 28195–28235, 2012

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Escudero, M., Querol, X., Pey, J., Alastuey, A., Pérez, N., Ferreira, F., Alonso, S., Rodríguez, S., and Cuevas, E.: A methodology for the quantification of the net African dust load in air quality monitoring networks, *Atmos. Environ.*, 41, 5516–5524, 2007.

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ACPD

12, 28195–28235, 2012

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Table 1. Location, type (regional background: RB; sub-urban background: SUB), data availability and measurement methods used in the monitoring sites of this study.

Country	Code	Type	Latitude	Longitude	Altitude (m.a.s.l.)	Start	End	Method
SPAIN	MSY	RB	41°45′36″ N	02°35′00″ E	728	2002	2011	GRAV
	MON	RB	40°56′48″ N	00°17′27″ W	570	2001	2011	BETA
	ZAR	RB	39°05′10″ N	01°06′07″ W	885	2001	2011	GRAV
	VIZ	RB	37°14′18″ N	03°28′28″ W	1265	2001	2011	GRAV
	BAR	RB	38°28′33″ N	06°55′22″ W	393	2001	2011	GRAV
	SPM	RB	39°31′29″ N	04°21′09″ W	1241	2001	2011	GRAV
	BEL	SUB	39°33′50″ N	02°37′22″ E	117	2001	2011	BETA
FRANCE	GEN	RB	45°43′55″ N	04°58′50″ E	235	2001	2010	TEOM
	DRS	RB	44°31′15″ N	05°05′24″ E	460	2004	2010	TEOM
ITALY	FEB	RB	44°45′04″ N	10°26′05″ E	1020	2005	2010	BETA
	FON	RB	41°40′48″ N	13°40′48″ E	393	2001	2010	BETA
	LEC	RB	40°27′32″ N	18°06′58″ E	10	2009	2010	BETA
	CST	RB	39°03′52″ N	08°27′26″ E	270	2005	2010	BETA
	BOC	SUB	38°07′13″ N	13°18′07″ E	141	2001	2010	BETA
GREECE	PAN	SUB	40°35′20″ N	23°01′54″ E	363	2001	2010	BETA
	APK	SUB	37°59′36″ N	23°49′10″ E	290	2001	2010	BETA
	FKL	RB	35°20′00″ N	25°40′00″ E	150	2004	2010	GRAV
BULGARIA	ROJ	RB	41°41′45″ N	24°44′19″ E	1750	2005	2010	BETA
CYPRUS	AYM	RB	35°02′21″ N	33°03′29″ E	532	2003	2010	GRAV

MSY: Montseny; MON: Monagrega; ZAR: Zarra; VIZ: Víznar; BAR: Barcarrota; SPM: San Pablo de los Montes; BEL: Castillo de Bellver; GEN: Genas; DRS: Drôme Rurale Sud; FEB: Febbio; FON: Fontechiari; LEC: Lecce; CST: Censt; BOC: Boccadifalco; PAN: Panorama; APK: Agia Pareskevi; FKL: Finokalia; ROJ: Rojen Peak; AYM: Ayia Marina GRAV: Gravimetric; BETA: Beta Attenuation monitor; TEOM: Tapered Element Oscillating Microbalance.

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

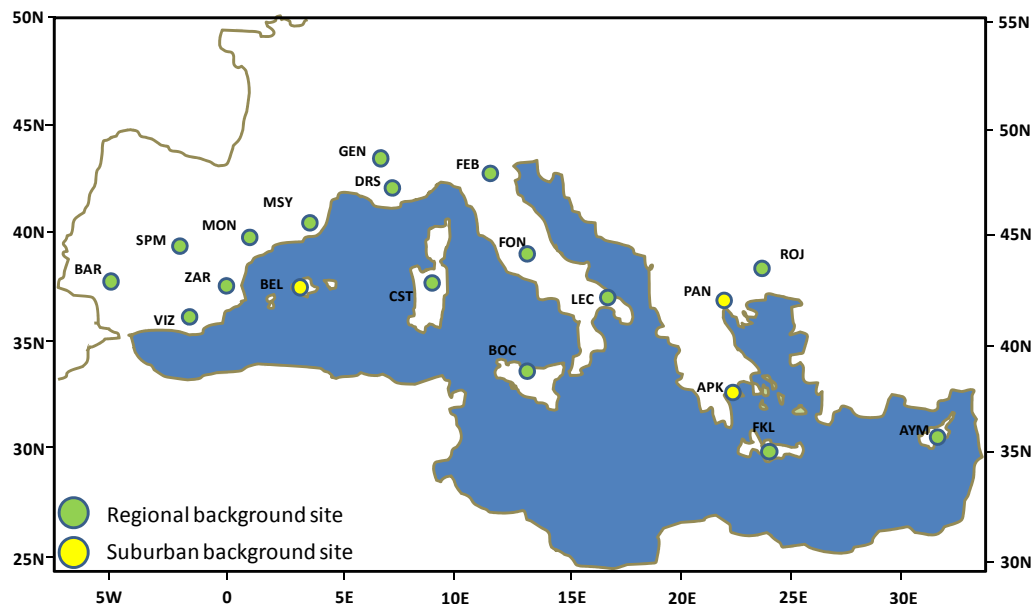
Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

**Fig. 1.** Location of regional and suburban background sites providing data for this study.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

**African dust
outbreaks over the
Mediterranean Basin
during 2001–2011**

J. Pey et al.

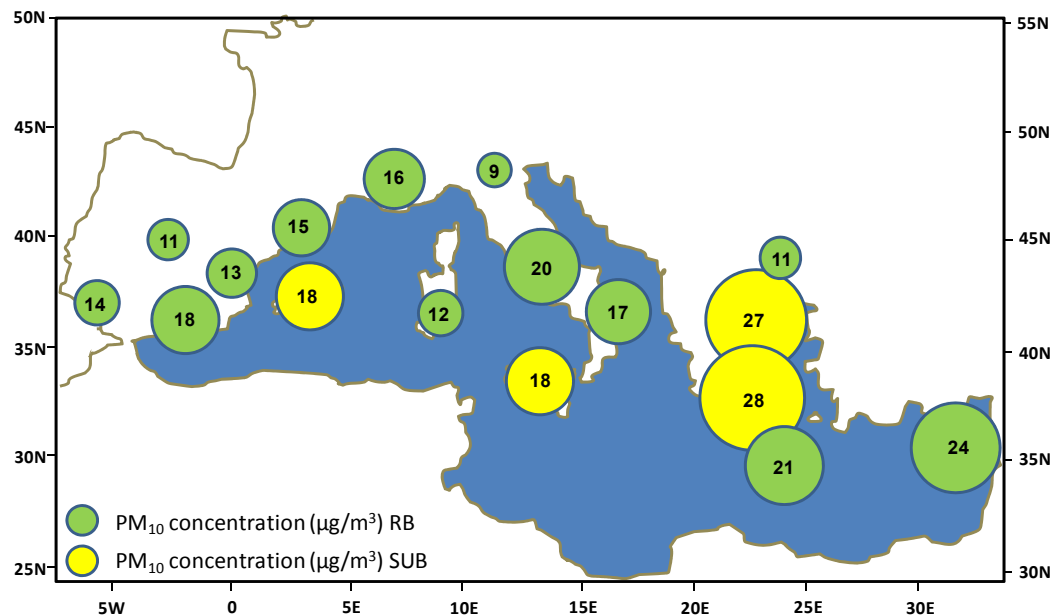


Fig. 2. Annual PM₁₀ levels (µg m⁻³) at regional (RB) and suburban (SUB) background sites across the Mediterranean for the period 2001–2011.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

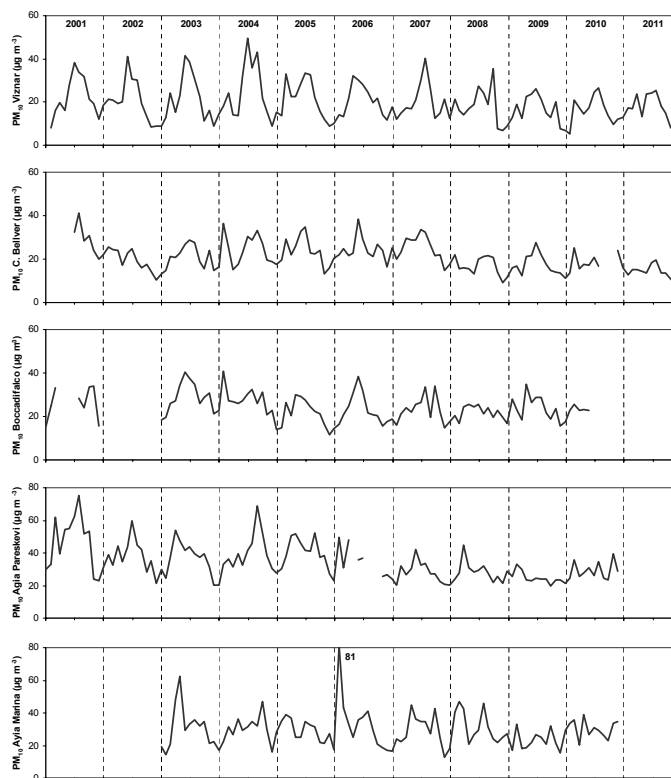


Fig. 3. Monthly PM_{10} levels ($\mu\text{g m}^{-3}$) at selected regional and suburban background sites across the Mediterranean Basin in the period 2001–2011: Víznar (SE Spain); C. Bellver (Balearic Islands); Boccadifalco (Sicily); Agia Pareskevi (Athens); Ayia Marina (Cyprus).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

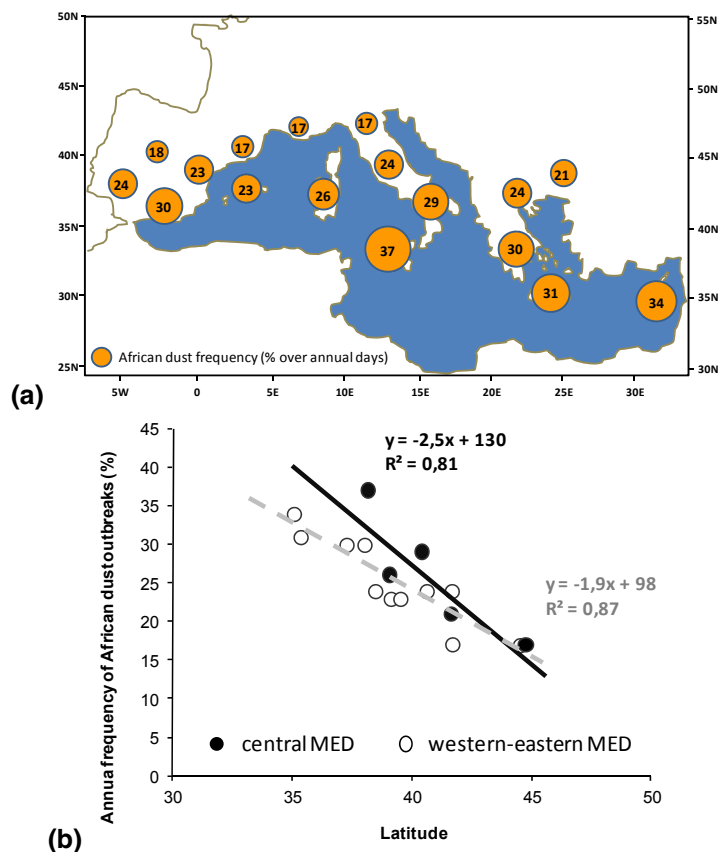


Fig. 4. (a) Mean frequency of African dust outbreaks (%) across the Mediterranean Basin during the period 2001–2011. (b) Mean frequency of African dust outbreaks (%) versus latitude across the Mediterranean Basin during the period 2001–2011.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

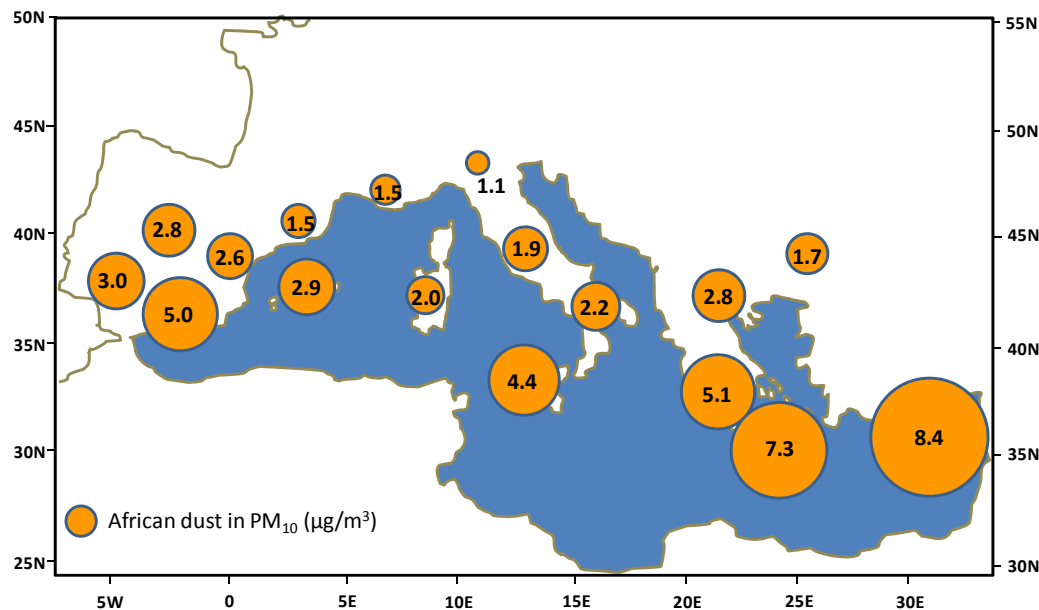


Fig. 5. Mean African dust contributions to PM₁₀ (in $\mu\text{g m}^{-3}$) across the Mediterranean (average values for the periods when data are available, in most cases from 2001–2010).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

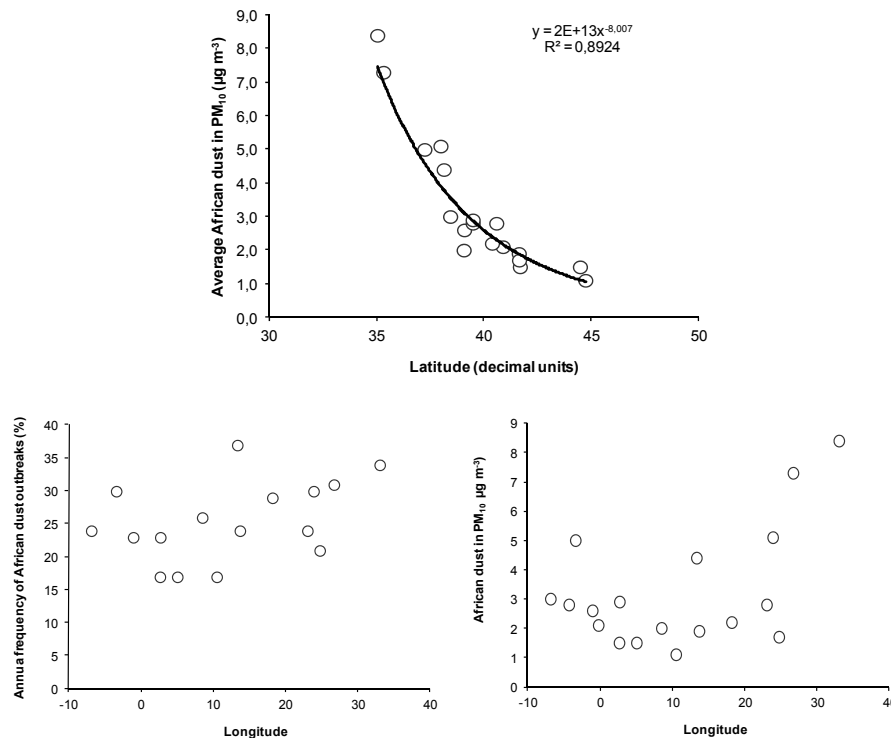


Fig. 6. (a) Top-left: mean annual African dust contributions in PM_{10} (in $\mu g m^{-3}$) versus latitude; (b) bottom-left: mean annual frequency of African dust outbreaks (%) versus longitude; (c) bottom-right: mean annual African dust contributions in PM_{10} (in $\mu g m^{-3}$) versus longitude.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

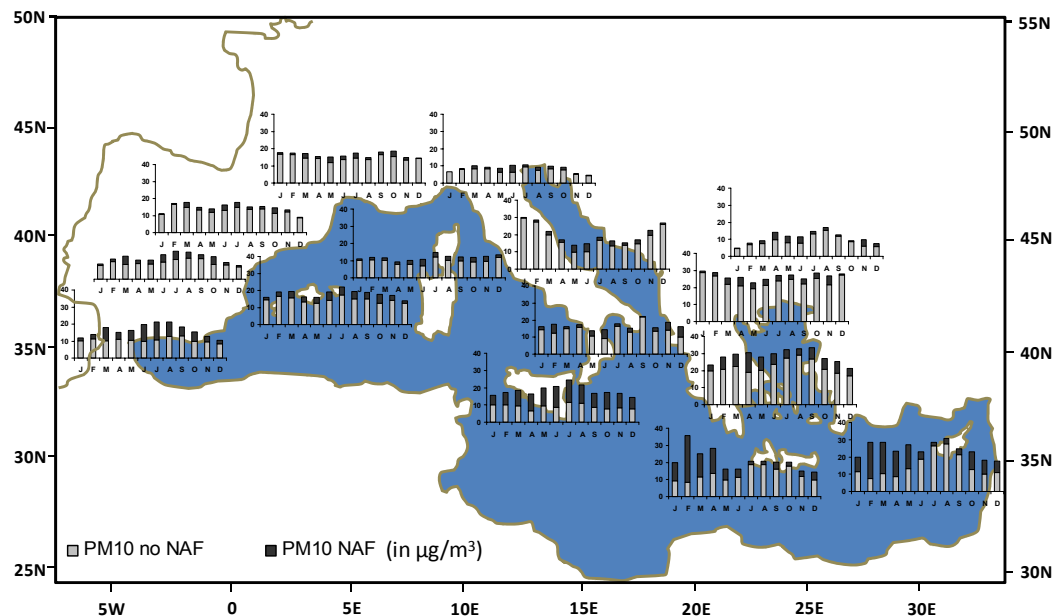


Fig. 7. Seasonal partitioning of PM_{10} (in $\mu\text{g m}^{-3}$), considering the influence of African dust, (average values for the periods where data are available, in most cases from 2001–2010) across the Mediterranean Basin.

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

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

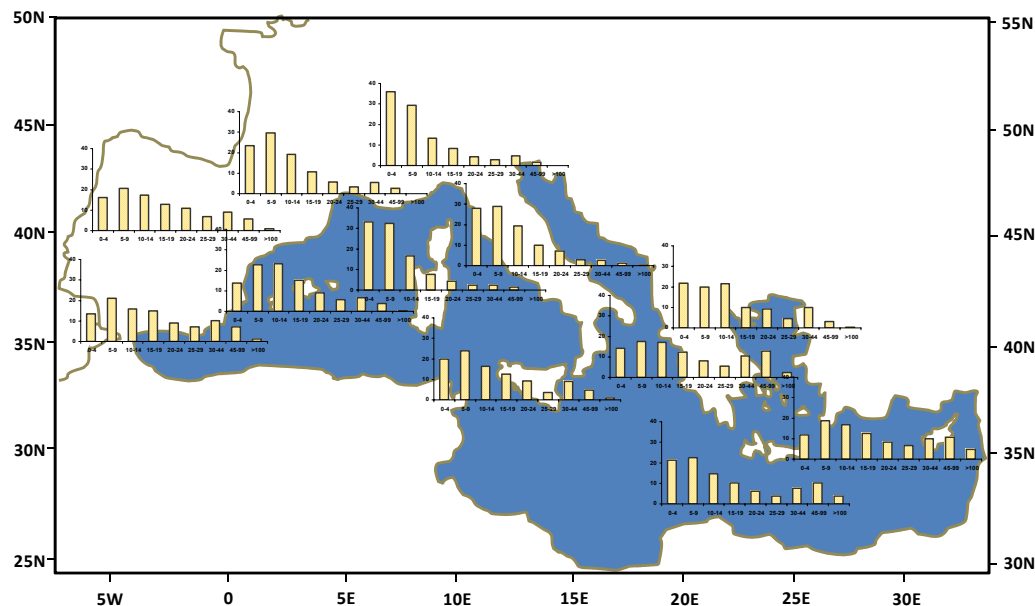


Fig. 8. Percentage of African dust episodes according to their intensity (divided in 9 concentration intervals from 0–4 to $> 100 \mu\text{m}^{-3}$) in selected areas across the Mediterranean Basin.

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

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

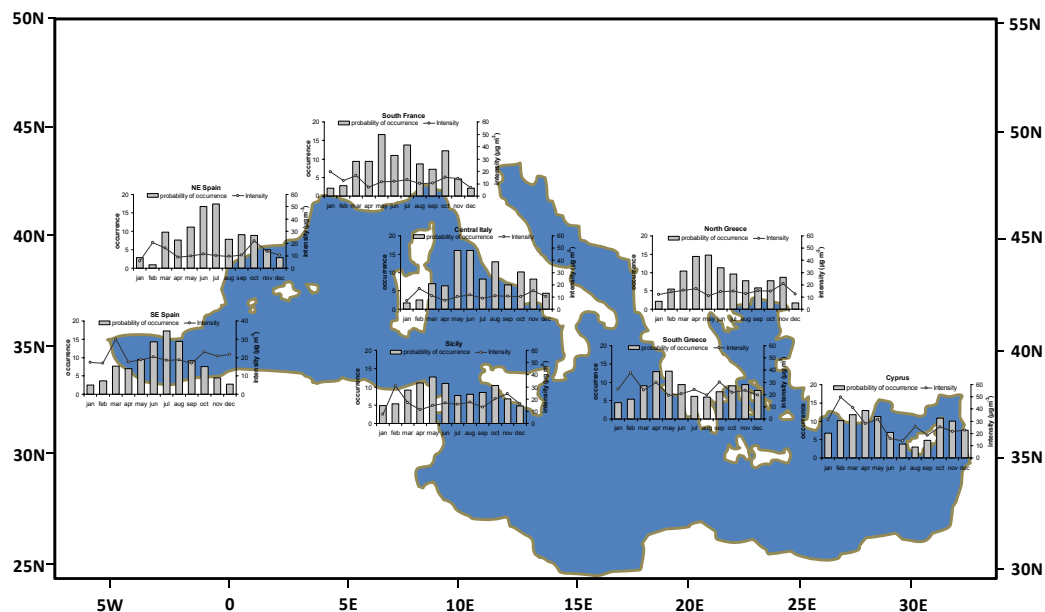


Fig. 9. Relation between probability of occurrence of African dust episodes (%) and intensity ($\mu\text{g m}^{-3}$) in selected areas across the Mediterranean Basin.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

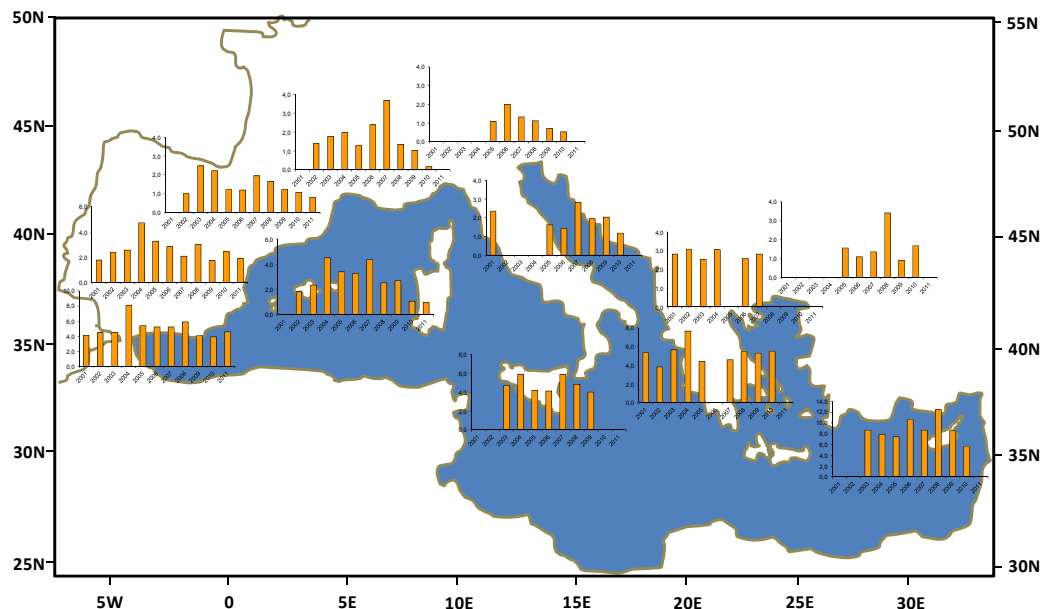


Fig. 10. Mean annual African dust contributions to PM_{10} (in $\mu\text{g m}^{-3}$) across the Mediterranean from 2001 to 2011 when available.

**African dust
outbreaks over the
Mediterranean Basin
during 2001–2011**

J. Pey et al.

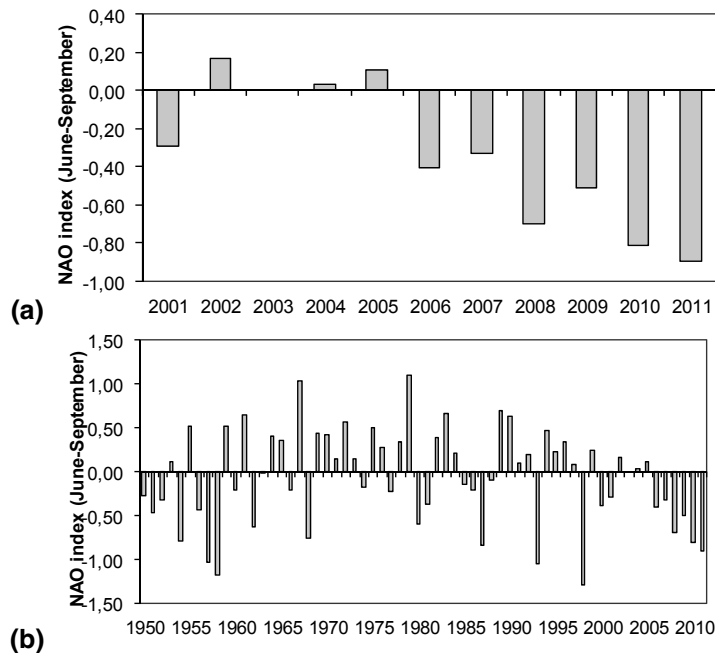


Fig. 11. (a) NAO index calculated for the period June-September from 2001 to 2011. **(b)** NAO index calculated for the period June-September from 1950 to 2011.

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

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

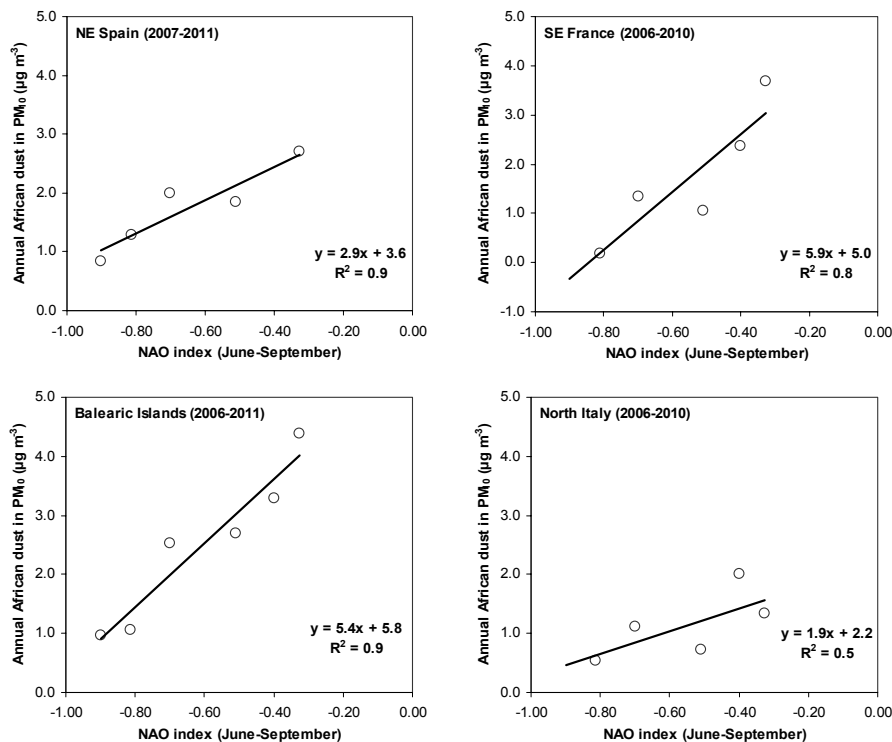


Fig. 12. Scatter-plots between summer NAO index with respect to annual African dust contribution to PM_{10} (in $\mu g m^{-3}$) for NE Spain, SE France, the Balearic Islands and North Italy for the period 2006/2007–2010/2011.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

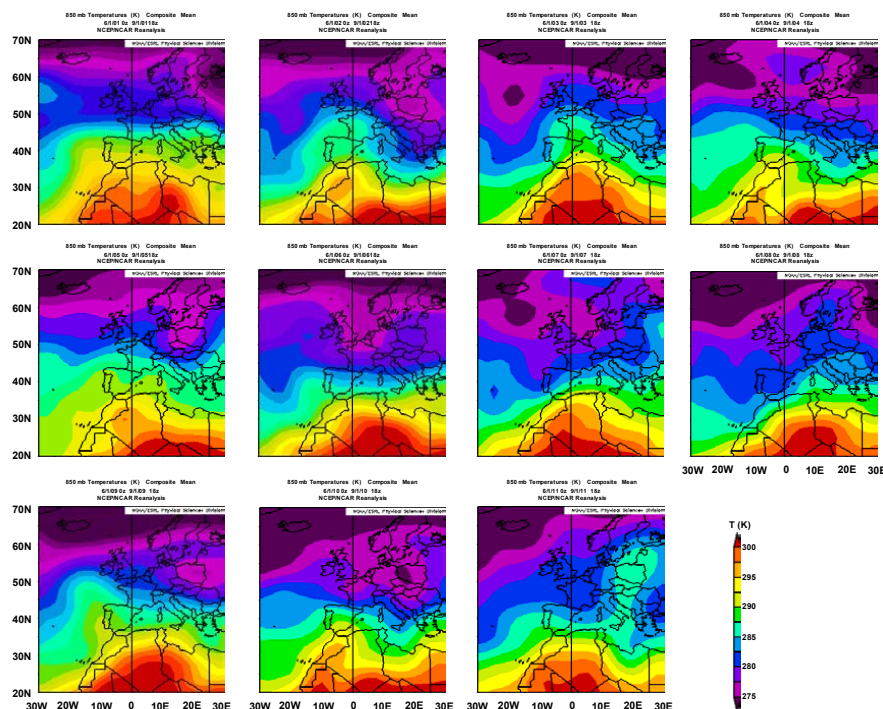


Fig. 13. Mean temperatures during summer periods (2001–2011), at a geopotential height of 850 hPa.

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

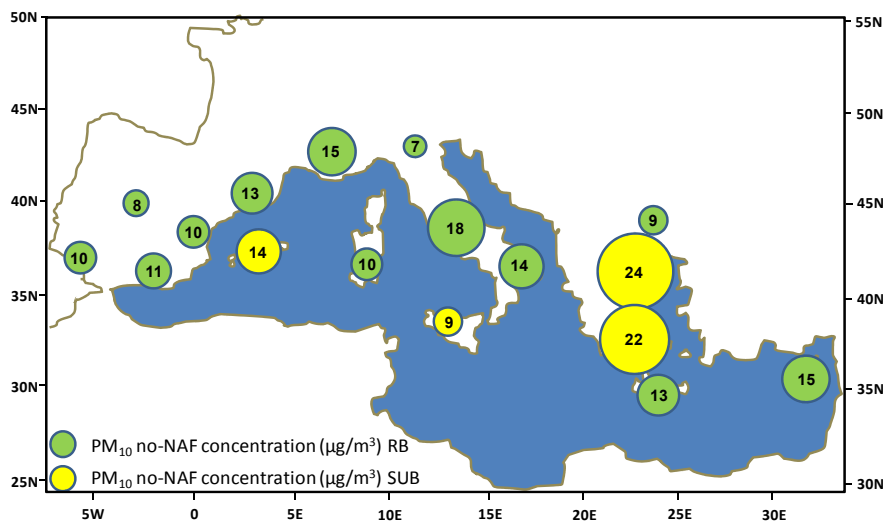


Fig. A1. Annual PM₁₀ levels (µg m⁻³), without African dust, at regional (RB) and suburban (SUB) background sites across the Mediterranean for the period 2001–2011.

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

African dust outbreaks over the Mediterranean Basin during 2001–2011

J. Pey et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

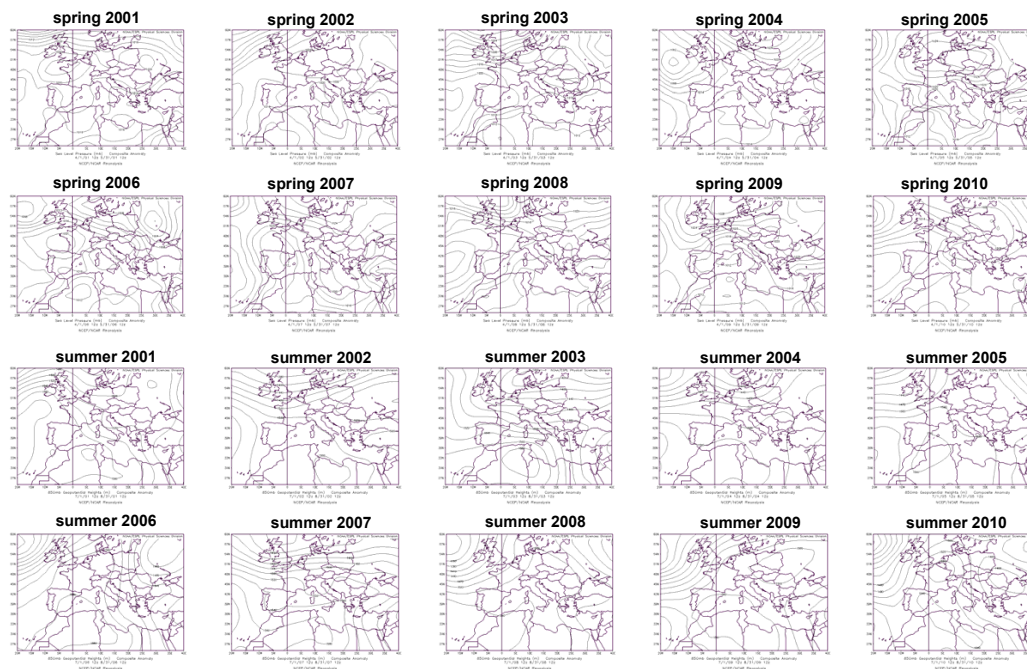


Fig. A2. Top: mean sea level pressure at surface calculated for spring periods from 2001 to 2010. Bottom: mean geopotential height at 850 hPa for summer periods from 2001 to 2010.