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Saharan dust contributions to PM10 and TSP levels in Southern and Eastern Spain

S. Rodríguez[a,]*, X. Querol[a], A. Alastuey[a], G. Kallos[b], O. Kakaliagou[b]

[a]*Institute of Earth Sciences "Jaume Almera", CSIC, C/ Lluis Solé i Sabarís, s/n, 08028 Barcelona, Spain*
[b]*Division of Applied Physics, University of Athens, University Athens Bldg PHYS-V, 15784 Athens, Greece*

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Abstract

The analysis of PM10 and TSP levels recorded in rural areas from Southern and Eastern Spain (1996–1999) shows that most of the PM10 and TSP peak events are simultaneously recorded at monitoring stations up to 1000 km apart. The study of the atmospheric dynamics by back-trajectory analysis and simulations with the SKIRON Forecast System show that these high PM10 and TSP events occur when high-dust Saharan air masses are transported over the Iberian Peninsula. In the January–June period, this dust transport is mainly caused by cyclonic activity over the West or South of Portugal, whereas in the summer period this is induced by anticyclonic activity over the East or Southeast Iberian Peninsula. Most of the Saharan intrusions which exert a major influence on the particulate levels occur from May to September (63%) and in January and October. In rural areas in Northeast Spain, where the PM10 annual mean is around 18 μg PM10 m^{-3}, the Saharan dust accounts for 4–7 annual daily exceedances of the forthcoming PM10-EU limit value (50 μg PM10 m^{-3} daily mean). Higher PM10 background levels are recorded in Southern Spain (30 μg PM10 m^{-3} as annual mean for rural areas) and very similar values are recorded in industrial and urban areas. In rural areas in Southern Spain, the Saharan dust events accounts for 10–23 annual daily exceedances of the PM10 limit value, a high number when compared with the forthcoming EU standard, which states that the limit value cannot be exceeded more than 7 days per year. The proportion of Sahara-induced exceedances with respect to the total annual exceedances is discussed for rural, urban and industrial sites in Southern Spain. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Air quality; PM10; Saharan dust; Mediterranean basin; Spain

1. Introduction

Suspended particle levels are monitored in ambient air quality networks because of their potential impact on human health, visibility and climate. In the last decade a review of the National Air Quality Standards has been carried out in certain countries, prompted by the results of a number of epidemiological studies showing a close relationship between fine particles and health effects (e.g. Dockery and Pope, 1996). In 1987, the monitoring of TSP (total suspended particles) was replaced by PM10 (particulate matter $<10\,\mu m$, aerodynamic diameter) measurements in the United States as a result of a review of the National Standards (US-EPA, 1986). More recently, a new review of the air quality standards resulted in the standardisation of PM2.5 (particulate matter $<2.5\,\mu m$, aerodynamic diameter) in addition to PM10 measurements (US-EPA, 1996).

In the European Community, the new Air Quality Directive (Directive 1999/30/EC) established the limit values for PM10, to be accomplished by 2010 (20 μg PM10 m^{-3} as annual daily value and not exceeding the daily value of 50 μg PM10 m^{-3} more than 7 days per year), following an intermediate stage (2005–2010) with more permissive limit values (40 μg PM10 m^{-3} as annual

* Corresponding author.
E-mail address: srodriguez@ija.csic.es (S. Rodríguez).

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daily value and not exceeding the daily value of 50 μg PM10 m^{-3} more than 35 days per year). As a result of the stringency of this new standard, exceedances of the PM10 limit values may not always be caused by anthropogenic emissions. In Southern European environments, natural PM10 sources may cause a number of PM10 exceedances. Thus, prior studies in the Western Mediterranean basin have shown that natural mineral particulate sources such as high-dust Saharan air mass intrusions interfere with the monitoring of the incidence of anthropogenic emissions on ambient air PM10 levels (Querol et al., 1998).

A number of studies have focused on the long-range transport of Saharan air masses over Western and Eastern North Atlantic areas (Prospero and Ness, 1986; Condé-Gaussen et al., 1987; Bergametti et al., 1989; Muhs et al., 1990; Savoie et al., 1992; Chiapello et al., 1995; Rajkumar and Siung Chang, 2000), South America (Prospero, 1999) and the Mediterranean (Loÿe-Pilot et al., 1986; Molinaroli et al., 1993; Avila et al., 1997, 1998; Díaz and Miranda, 1997; Guerzoni et al., 1997). Although considerable literature is available on the mineral and chemical composition of Saharan dust, transport patterns of selected dust outbreak events and the influence on the deposition chemistry, there is limited information on the actual impact of Saharan dust on the records of air quality monitoring networks in Southern European countries.

This study presents preliminary results of a research project supported by the Spanish Ministry of the Environment and the Comisión Interministerial de Ciencia y Tecnología (CICYT) focusing on PM10 and PM2.5 source apportionment analysis in selected areas in Spain with different atmospheric and geographical patterns. In this regard, the present study focuses on the identification of high PM10/TSP events (1996–1999) caused by intrusions of Saharan dust over the Iberian Peninsula, and on the description of the transport patterns.

2. Methodology

The study area covers the Eastern and Southern regions of the Iberian Peninsula. The topography is characterised by a high central plateau, Eastern coastal ridges crossed by deep valleys, Southeastern high mountains and a Southwestern deep valley. Most of these areas are characterised by a poor soil coverage, with semi-arid conditions in the inner eastern areas and desert environments in several areas of the Southeastern regions.

2.1. Measurements

Measurements of PM10, TSP and gaseous pollutants (NO_x, SO_2, CO and O_3) from 1996 to 1999 were interpreted in this study. The data were obtained from the ambient air quality monitoring networks belonging to a number of Autonomous Governments of Eastern and Southern Spain (Catalonia, Valencia and Andalusia), Empresa Nacional de Electricidad S.A. (ENDESA) and from the European Monitoring Evaluation Program (EMEP) network. Seventeen rural, industrial and urban monitoring stations from different climatic areas were selected. The location and major characteristics of the monitoring stations are summarised in Table 1 and Figs. 3–6.

In the ambient air quality monitoring networks of Catalonia and Valencia, TSP measurements were carried out using automatic beta radiation attenuation monitors. The same measurement procedure was used by the Andalusia network for monitoring PM10 and TSP levels. The ENDESA network monitored PM10 levels by using automatic tapered oscillating microbalance system (TEOM). Finally, at the EMEP stations, TSP measurements were carried out using the manual gravimetric method with high-volume captors. Periodical manual gravimetric measurements performed simultaneously with the automatic measurements in most of the stations guarantee the accuracy of the measurements.

2.2. Meteorological analysis

Air back-trajectory analysis, synoptic charts and simulations using the SKIRON Forecast System (Kallos et al., 1997) were performed in this study. Air back-trajectories were computed by means of the Hybrid Single-Particle Lagrangian Integrated Trajectories (HYSPLIT) trajectory model (Draxler, 1994) using meteorological data supplied by the US National Climatic Data Centre. Five to ten day back-trajectories were obtained at 750, 1500 and 2500 m a.s.l. at a 6 h step. Synoptic charts from European Centre Medium Weather Forecast (ECMWF) were used to support trajectory analyses and to identify the synoptic pressure system giving rise to each transport pattern from the Sahara.

The SKIRON forecasting system is a version of the ETA/NCEP weather forecasting model developed at the University of Athens (Kallos et al., 1997; Nickovic et al., 1998a). The 'heart' of the system is the ETA/NCEP model developed initially at the University of Belgrade and the Yugoslav Hydrometeorological Service and further developed at NOAA/NCEP.

The system is fully portable and contains a series of modules for pre- and post-processing of data. A major feature of the SKIRON system is the modules describing the dust cycles in the atmosphere (uptake–transport–deposition (Nickovic and Dobricic, 1996; Nickovic et al., 1998b).

Simulations with the SKIRON system were carried out for selected high PM10/TSP events recorded at rural

Table 1
TSP and PM10 monitoring stations used in the present study. See location in Figs. 3–6

Station	Parameter	Location	Environment
Monagrega, MON	PM10	40.5°N, 0.2°W, 600 m a.s.l.	Rural
Carboneras, CAR	PM10	37.0°N, 1.9°W, 11 m a.s.l.	Rural
Coratxar, COR	TSP	40.4°N, 0.0°, 1235 m a.s.l.	Rural
Vilafranca, VIL	TSP	39.7°N, 0.1°W, 1125 m a.s.l.	Rural
Espiel, ESP	TSP	38.0°N, 5.0°W, 520 m a.s.l.	Rural
San Pablo, SPA	TSP	39.2°N, 4.2°W, 917 m a.s.l.	Rural, EMEP
Logroño, LOG	TSP	42.4°N, 2.5°W, 503 m a.s.l.	Rural, EMEP
Campisábalos, CAM	TSP	41.2°N, 3.0°W, 1405 m a.s.l.	Rural, EMEP
Roquetas, RQT	TSP	40.5°N, 0.0°W, 46 m a.s.l.	Rural, EMEP
Fornells, FOR	TSP	41.6°N, 2.6°E, 101 m a.s.l.	Urban
Sagrera, SAG	TSP	41.2°N, 2.1°E, 60 m a.s.l.	Urban, Barcelona city
L'Hospitalet, L'HO	PM10	41.2°N, 2.0°E, 70 m a.s.l.	Urban, Barcelona area
Onda, OND	TSP	39.8°N, 0.1°W, 193 m a.s.l.	Industrial
Linares, LIN	PM10	38.0°N, 3.0°W, 280 m a.s.l.	Urban, Linares city
Palos de la Frontera, PLF	PM10	37.1°N, 6.5°W, 24 m a.s.l.	Industrial
Constitución, CON	PM10	37.1°N, 3.4°W, 688 m a.s.l.	Urban, Granada city
Madrid, MAD	PM10	40.2°N, 3.4°W, 650 m. a.s.l.	Urban, Madrid city

stations and identified as Saharan dust intrusions by back-trajectory and the synoptic charts analysis. Output files were taken every 6 h for winds at standard pressure levels, dust load (the vertically integrated dust concentration in $g\,m^{-2}$) and precipitation data. Studied episodes showing a good agreement between the transport patterns obtained from SKIRON simulations and the direct satellite observations of Saharan intrusions over the Mediterranean are reported by Nickovic et al. (1997, 1998b).

For the SKIRON simulations, the ECMWF analysis data fields ($0.5^\circ \times 0.5^\circ$ latitude × longitude) at standard pressure levels were used for initialisation and for nudging the boundaries domain every 6 h. Moreover, climatological sea-surface temperature fields ($1^\circ \times 1^\circ$ SST), topography database ($30'' \times 30''$), soil textural class data set ($1^\circ \times 1^\circ$) and gridded vegetation data ($30'' \times 30''$) were used. The modelling domain covered an area ranging from the Western Atlantic to the Central Mediterranean, and from Central Europe to Southern Algeria. The defined mesh had a grid increment of approximately 24 km horizontal ($0.25^\circ \times 0.25^\circ$) and 32 vertical levels up to about 16 km. The simulations covered a period of at least 5 days.

2.3. Data treatment

The following methodology was used.

Step 1: simultaneous high particulate events (coincident particulate peaks) identified at the records of daily PM10 and TSP levels from distant rural stations were selected (pre-selection).

Step 2: air back-trajectories were computed for these events and episodes showing a North African long-range transport pattern were finally selected for subsequent analysis.

Step 3: the impact of the Saharan dust on the PM10/TSP levels of urban and industrial stations for the selected events was studied to assess the relative importance of the Saharan dust input versus the anthropogenic emissions.

Step 4: SKIRON simulations were run for most of the selected events to confirm the mineral dust supply from the Sahara as a major origin of the high PM10 and TSP concentrations.

2.4. Source apportionment analysis

One of the Saharan events (25–28 August 1999) forecasted by the SKIRON model was selected for a PM10 source apportionment analysis to quantify the impact of the Saharan input on the ambient air PM10 levels simultaneously recorded at the distant urban, industrial and rural stations of Madrid-L'Hospitalet (Barcelona), Onda and Monagrega, respectively. The sampling was performed by high-volume samplers with PM10 cut-off inlets and quartz fibreglass filters. Sample treatment and analytical procedures are described by Querol et al. (2001). The results allowed us to differentiate the following elemental groups with a definite origin: (a) crustal elements (addition of Al and Si oxides, carbonate, Ca, K, Na, Ti, P, Fe, Mg and other minor natural elements); (b) elemental and organic carbon obtained from the subtraction of mineral C (stoichiometrically obtained from Ca levels); (c) secondary inorganic particles and heavy metals (sulphate, nitrate, ammonium and major anthropogenic heavy metals such as Zn, Pb, Cu, V, Mn and Ni); and (d) marine aerosol (sodium, marine sulphate, chloride).

3. Results and discussion

3.1. Saharan dust events and meteorological contexts

The Saharan intrusions over the study area occur when air masses over the Sahara desert move northward over the Iberian Peninsula. This results in an increase in PM10 and TSP levels at the air quality stations due to the high mineral load of the Saharan air masses. Table 2 summarises the particulate events with a major Saharan origin in the period 1996–1999. The interpretation of the meteorological mechanisms giving rise to each Sahara intrusion is also included in this table. Fig. 1 shows the daily PM10 and TSP levels recorded at the rural stations of Monagrega (NE Spain, MON in Fig. 6), Carboneras (SE Spain, CAR in Fig. 6), and the EMEP San Pablo station (SW Spain, SPA in Fig. 6), where the Saharan events are highlighted. For the sake of brevity, only some examples of Saharan events will be discussed to illustrate the results of this study.

Saharan air masses reach the study area when the synoptic situation is governed by depressions located to the West or Southwest of Iberian Peninsula (coast of Portugal or in the region between the Saint Vincent Cape, the straits of Gibraltar and the Canary Islands, Fig. 2a) or when the North African anticyclone (usually placed between Algeria and Egypt at the 850 hPa level) shifts to the East or Southeast of the Iberian Peninsula (Fig. 2b). The combination of both cyclone and anticyclone systems is also a scenario favouring the dust transport from North Africa towards Spain (Fig. 2c). Air back-trajectories of these transport scenarios are shown in Fig. 2d.

From January to June most of the Saharan events were induced by depressions located off Portugal (events 96-1, 96-2, 96-4, 97-3, 97-4, 98-1, 98-4, 99-3, 99-5 and 99-6 of Table 2 and Fig. 1) or between the Saint Vincent Cape, the Canary Islands and the straits of Gibraltar (97-1, 98-5 and 99-1, 99-2). Moreover, four events were induced by the North African anticyclone (97-2, 98-2, 98-3 and 99-4). The Saharan events caused by depressions are characterised by sharp (2–3 days) particulate peaks. PM10 levels one day before and one day after the particulate peak reached 12 and 25 μg PM10 m^{-3} at MON and CAR stations on average for all the events, whereas maximum average levels reached 41 and 57 μg PM10 m^{-3}, respectively. Low PM10 concentrations before the Saharan events are caused by the arrival of Atlantic air masses preceding the northward high particulate flow. The sharp and rapid increase in particulate levels is due to the plume-like behaviour of the Saharan intrusion under these scenarios. Furthermore, the rapid decrease in particulate levels recorded at the end of the events are frequently due to rainfall scavenging following most of the events produced under this scenario. This is the origin of the well-known Saharan red rains in Spain (Avila et al., 1997, 1998).

The event of 20–23 January 1997 is a typical case of Saharan dust transport affecting Eastern Spain (Fig. 3). On 19 January the depression located over the Saint Vincent Cape induced a Southwest flow over the Iberian Peninsula, resulting in an injection of Atlantic oceanic air masses and rainfalls. As a consequence low PM10 and TSP levels were registered in Southern and Eastern Spain. On 20 January the strong pressure differences between the Atlantic and the Western Mediterranean resulted in a persistent northward flow until 22 January (Fig. 2a). Thus, a Saharan plume expanded along the Eastern coast of Spain and high PM10 and TSP levels were registered in Eastern Spain. However, low PM10 values were simultaneously measured at the Southwestern stations, outside of the influence of the plume. The subsequent particulate scavenging by rainfall resulted in a sharp reduction in the particulate levels on 23 and 24 January in Northeastern and Southeastern Spain, respectively. PM10 daily levels exceeding the new limit values of the EU Standard were registered at the rural station of CAR during this event (147 and 116 μg PM10 m^{-3} on 22 and 23 January, respectively).

Another interesting example showing the influence of the Saharan plume on the daily PM10 and TSP levels occurred on 4 and 5 June 1998 (Fig. 4). In this case, the Northeast stations were the most affected. This event was caused by a depression crossing from the Atlantic ocean to the Western Mediterranean over the Atlas Mountain Range. The dust plume reached the North-eastern Spanish coast with the consequent increase in the particulate levels on 4 and 5 June. Daily levels of 49 and 71 μg PM10 m^{-3} were recorded at rural MON station during this event. As described for the January 1997 event, a sharp decrease in particulate levels was induced by rainfalls on 6 and 7 June and by the eastward displacement of the Saharan plume. In Southern Spain low PM10 levels were recorded owing to the strong influence of Atlantic air masses.

Summer meteorological conditions are characterised by the development of thermal lows over North Africa and the Iberian Peninsula (induced by the intense heating of ground) and by the intensification of the North African anticyclone (in the 850 hPa level; Font, 1956). The thermal convective activity over the Sahara desert forces the injection of particles to high atmospheric levels (Carlson and Prospero, 1972; Prospero and Carlson, 1972; Westphal et al., 1988). This important dust load at high altitudes is transported Southwestward as far as the Caribbean Islands (Prospero and Ness, 1986). The

Table 2
PM10/TSP peaks events induced by Saharan events in the period 1996–1999. *D*, duration of the event in days. The events may be induced by depressions centred off Portugal (type L(p)), in the region between Sant Vicent Cape, Canaries and Gibraltar (type L(c)), high pressure at the South or Southeast of Iberian Peninsula (H) or by the simultaneous presence of a Western or South-western depression and a Eastern anticyclone (type L + H)

Dates	No.	Duration (No. days)	Synoptic meteorology
1996			
15–17 January	96-1	3	L(p)
22–23 + 28 January	96-2	2 + 1	L(p) + L(p)
24–25 March	96-3	3	L(p) + H
20–22 April	96-4	3	L(p)
6–13 June	96-5	8	H
23–29 July	96-6	7	H
14–20 August	96-7	7	L(c)
23–29 October	96-8	7	H
1997			
22–23 + 26–27 + 31 January–1 February	97-1	2 + 2 + 2	L(p + c) + L(c) + L(p)
3–7 March	97-2	5	H
5–7 May	97-3	3	L(p)
27–30 May	97-4	4	L(p)
14–17 July	97-5	4	L(c)
9–10 August	97-6	2	L(p) + H
21–22 August	97-7	2	H
11–13 September	97-8	3	L(p)
29 September–3 October	97-9	5	L(c)
18–20 October	97-10	3	L(p)
1998			
11–13 January	98-1	3	L(p)
15–19 February	98-2	5	H
3–7 March	98-3	5	H
8–11 May	98-4	4	L(p)
4–5 June	98-5	2	L(c)
22–24 June	98-6	3	H
27 June–1 July	98-7	5	H
18–21 July	98-8	4	L(c) + H
11–15 August	98-9	5	H
25–28 August	98-10	4	H
30 August–2 September	98-11	4	H
1999			
7–9 January	99-1	3	L(c)
14–16 January	99-2	3	L(c)
8–14 March	99-3	7	L(p)
10–14 May	99-4	5	H
24–27 May	99-5	4	L(p)
28 May–3 June	99-6	7	L(p)
29 June–4 July	99-7	6	H
14–18 August	99-8	5	H
23–28 August	99-9	6	H
1–3 September	99-10	3	L(p) + H
27–30 October	99-11	4	L(c) + H

Saharan dust reaches the Iberian Peninsula when the North Atlantic anticyclone (Azores high) is displaced westward and the North African high is centred over Algeria. Most of the events which occurred in July and August were produced by this mechanism (96-5, 96-6, 97-7, 98-6, 98-7, 98-9, 98-10, 98-11, 99-7, 99-8 and 99-9, Table 2). The strong convective activity under the Iberian thermal low conditions (Millán et al., 1997) lead to the abatement of the Saharan air masses over the Iberian Peninsula. Dayan and Miller (1989) demonstrated that

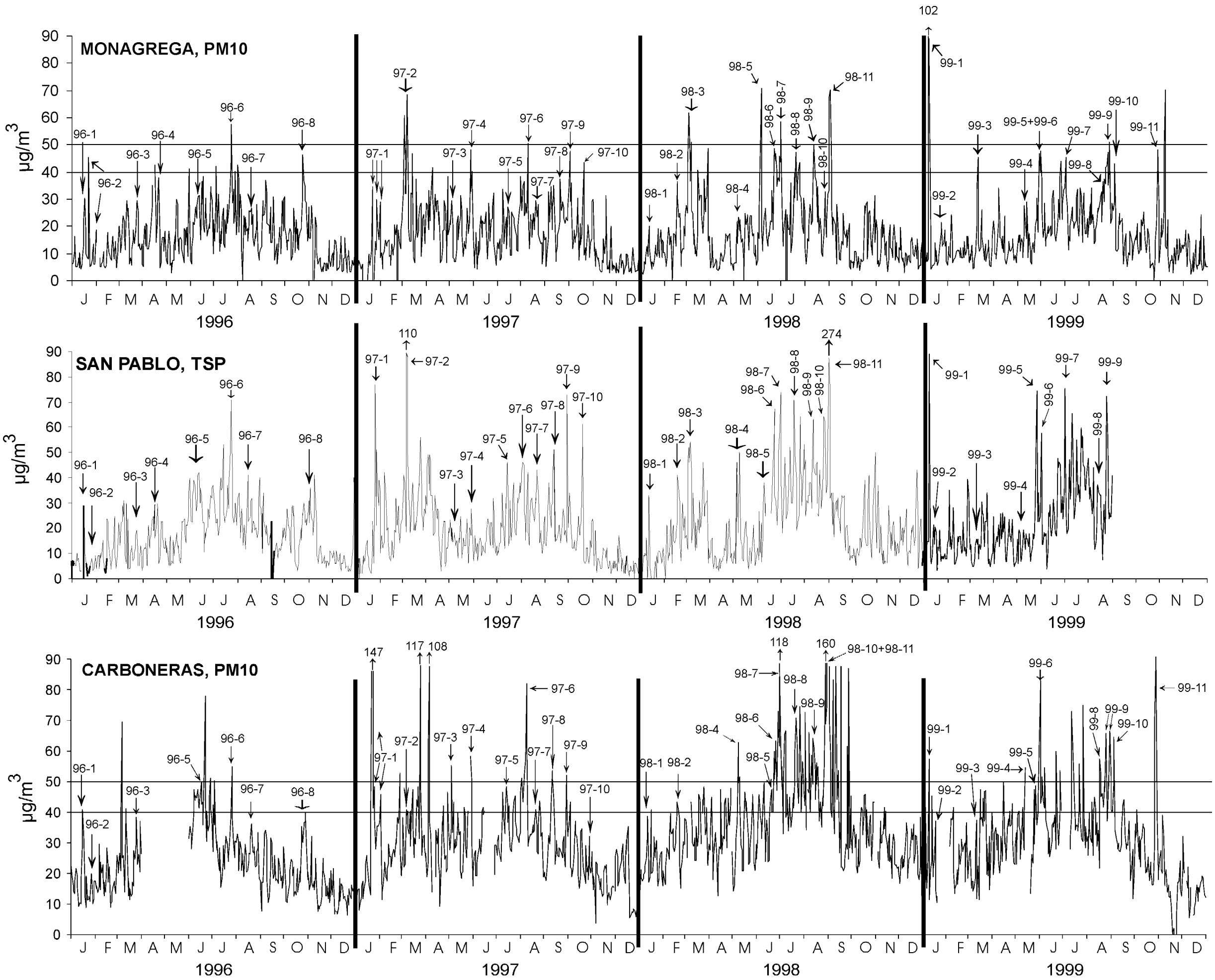

Fig. 1. 1996–1999 daily PM10 or TSP levels from the rural stations of Monagrega and Carboneras and the EMEP San Pablo station. Saharan dust events reported in Table 2 are highlighted in the particulate records.

the mixing of the lower troposphere levels with upper atmospheric masses proceeding from North Africa during these intrusions is enhanced during summer by the greater thickness of the mixing layer.

The influence of the Saharan air masses over the Iberian Peninsula under this synoptic pattern has been described by meteorological studies which highlight the strong influence of these hot and dry air masses on the weather conditions in Spain (Sánchez, 1993).

An example of high summer dust events occurred from 30 August to 2 September 1998 (Fig. 5). Owing to the location of the North African high over Algeria (a synoptic scenario similar to that shown in Fig. 2c), a dust intrusion approached Southeast Spain on 30th of August. Subsequently, it spread over Southwestern and Northeastern Spain on 31st August and 1st September, respectively. Daily PM10 means registered at rural stations reached $150\,\mu g\,m^{-3}$ in the South-eastern regions (CAR) and $70\,\mu g\,m^{-3}$ in the North-eastern regions (MON).

A significant and common feature of a number of summer dust outbreaks (96-5, 98-6, 98-9, 99-8 and 99-9) caused by the North African anticyclone is the slow reduction in the particulate levels after the Saharan event when compared with the winter events. This is probably due to the following factors:

1. The larger extension of the high-particulate air mass in contrast to the plume-like morphology of the winter intrusions.
2. The high convective dynamics account for a high resuspension of dust and for a slow renovation of the air masses (Millán et al., 1997)
3. The low atmospheric scavenging potential due to the low rainfall.

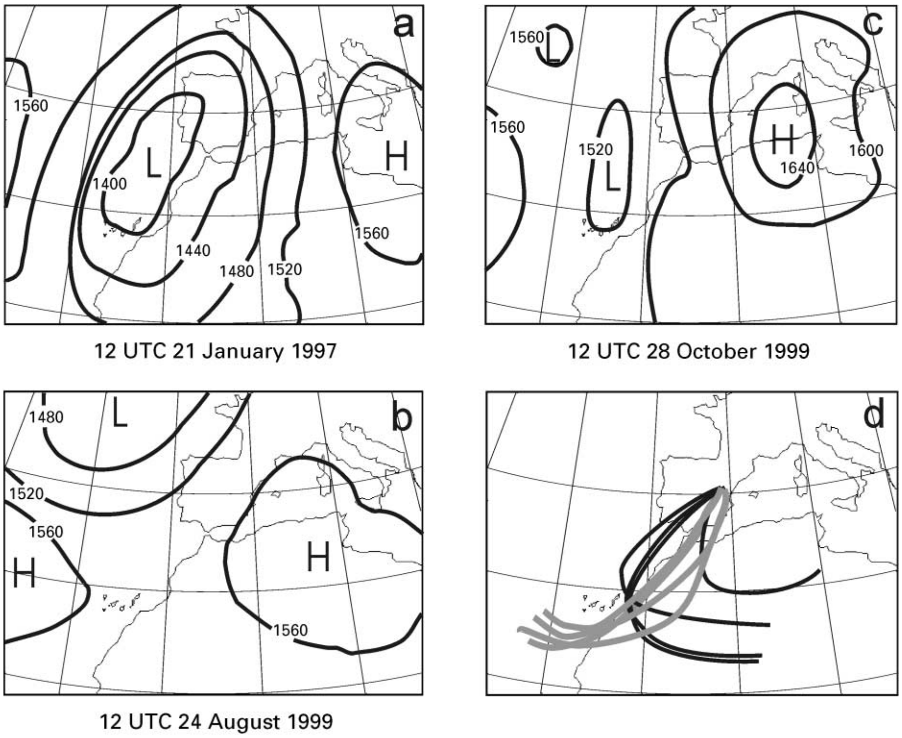

Fig. 2. Synoptic pressure systems (altitude of the 850 hPa level in meters) inducing the transport of the Saharan dust to the Iberian Peninsula. (a) Low pressures over West and/or Southwest of Portugal. (b) High pressure over East or Southeast of Iberian Peninsula. (c) The combination of both low and high pressure systems. (d) Back-trajectories to the Monagrega station induced by these synoptic systems: bold lines for anticyclonic pathways (25 July 1996, 27 August 1998, 31 August 1998 and 13 May 1999) and grey lines for the cyclonic pathways (22 January 1996, 10 August 1997, 10 March 1999, 27 October 1999).

By contrast, the summer dust events caused by Southwestern depressions or ending with an injection of Atlantic air masses show a sharp reduction in the particulate levels as in the case of the winter events.

3.2. Comparison with other measurements in the Mediterranean basin

The magnitude of the daily PM10 and TSP levels recorded for the Saharan events in this study is in agreement with the findings of earlier studies in the Mediterranean basin. Thus, Molinaroli et al. (1993) recorded average levels of 98 μg TSP m^{-3} in Sardinia in March 1991 during Sahara events, whereas Northern and Western airflows gave rise to TSP concentrations between 11 and 32 $\mu g\,m^{-3}$. In August 1984, Lefèvre et al. (1986) in Sicily recorded average daily TSP levels of 92 $\mu g\,m^{-3}$ during a Saharan event and 33 $\mu g\,m^{-3}$ for dominant Northern flows. Correggiari et al. (1989) measured 72 μg TSP m^{-3} on board ship South of 40°N latitude during a Saharan episode on February 1983. Chester et al. (1984) recorded 100 μg TSP m^{-3} in the Central Tyrrehenian in October 1979, whereas they measured 11 μgTSP m^{-3} under Eastern flow conditions. Guerzoni et al. (1989) recorded 5 μgTSP m^{-3} in the southern Thyrrhenian on board ship under North-western airflows.

The high mineral load in ambient air PM10 levels during the Saharan dust outbreaks was quantified for the event occurring from 23 to 28 August 1999 (Fig. 6). PM10 sampling was simultaneously carried out at urban stations in Madrid and L'Hospitalet-Barcelona (MAD and L'HO), at Onda industrial (OND) and Monagrega rural (MON) sites from 25 to 26 August. Synoptic meteorological mechanism inducing the transport of Saharan dust to Iberian Peninsula during this event is shown in Fig. 2b. Fig. 6 shows the dust load over the study area as well as the results of the PM10 source apportionment analysis. The first interesting result is that minor differences in the PM10 levels were obtained at rural, industrial and urban sites. The chemical source apportionment showed a large mineral load (crustal end member), which ranged from 15 to 20 $\mu g\,m^{-3}$ on bulk PM10 levels close to 45 $\mu g\,m^{-3}$ at OND and L'HO, and from 30 to 35 $\mu g\,m^{-3}$ for bulk PM10 levels close to 50 $\mu g\,m^{-3}$ at MON and MAD. Similar studies by Molinaroli et al.

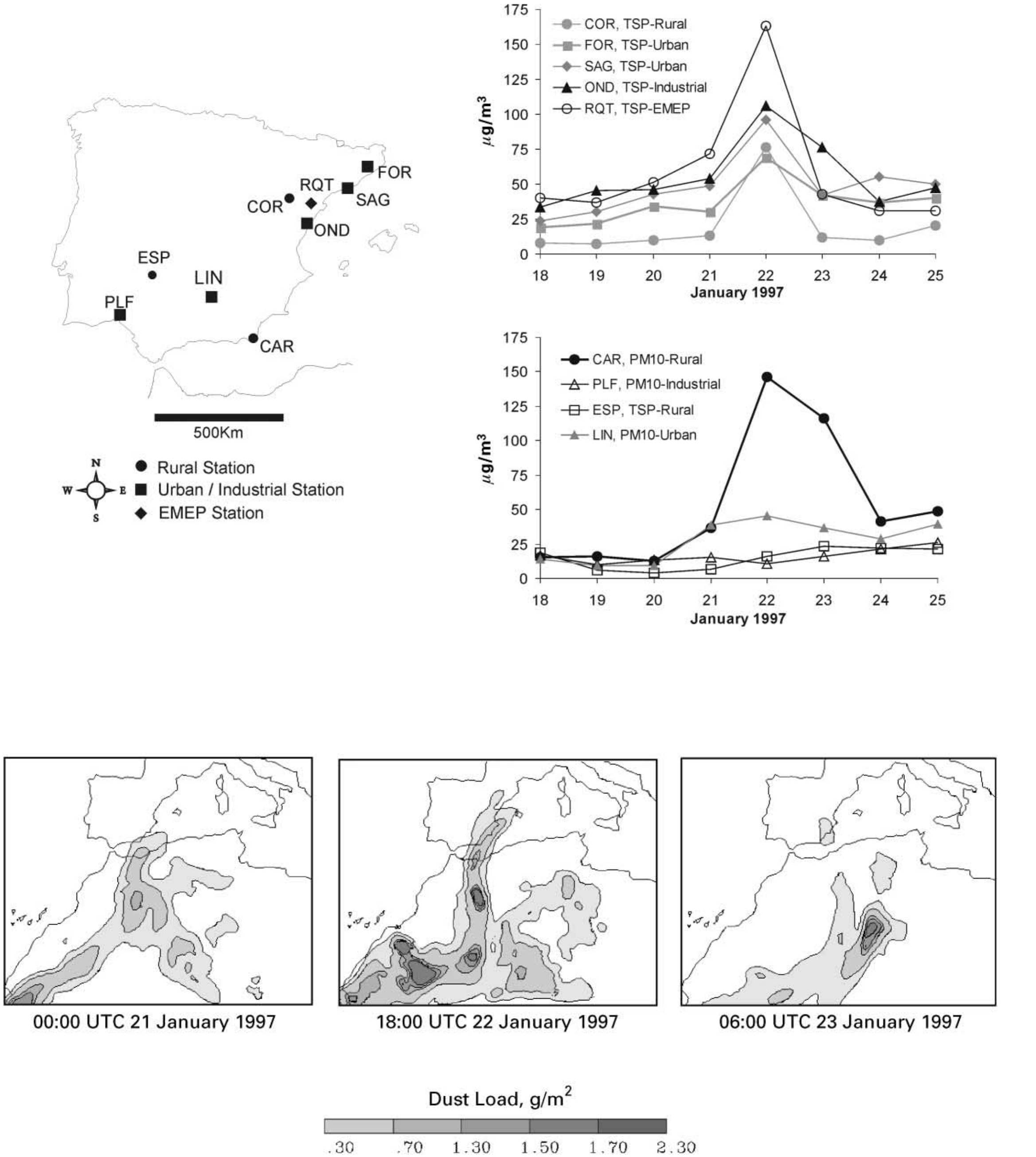

Fig. 3. Daily PM10 and TSP levels recorded at selected monitoring stations during the Saharan event from 22 to 23 January 1997 and evolution of the dust load (vertically integrated dust concentration in $g\,m^{-2}$) from the SKIRON model.

(1993) in Sardinia obtained a mineral load concentration of 39–50 $\mu g\,m^{-3}$ for bulk TSP levels of 80–100 $\mu g\,m^{-3}$. The higher dust load obtained by Molinaroli et al. (1993) is due to the TSP sampling which includes a higher proportion of mineral load than PM10. In addition to the mineral load, the Saharan dust may also have an anthropogenic fraction (mainly aerosols from biomass combustion and nitrates, for details see Savoie et al., 1992; Díaz and Miranda, 1997; Avila et al., 1998) which may have been emitted in Northern Africa (Rúa et al., 1998) or re-circulated from Europe (Kallos et al., 1998).

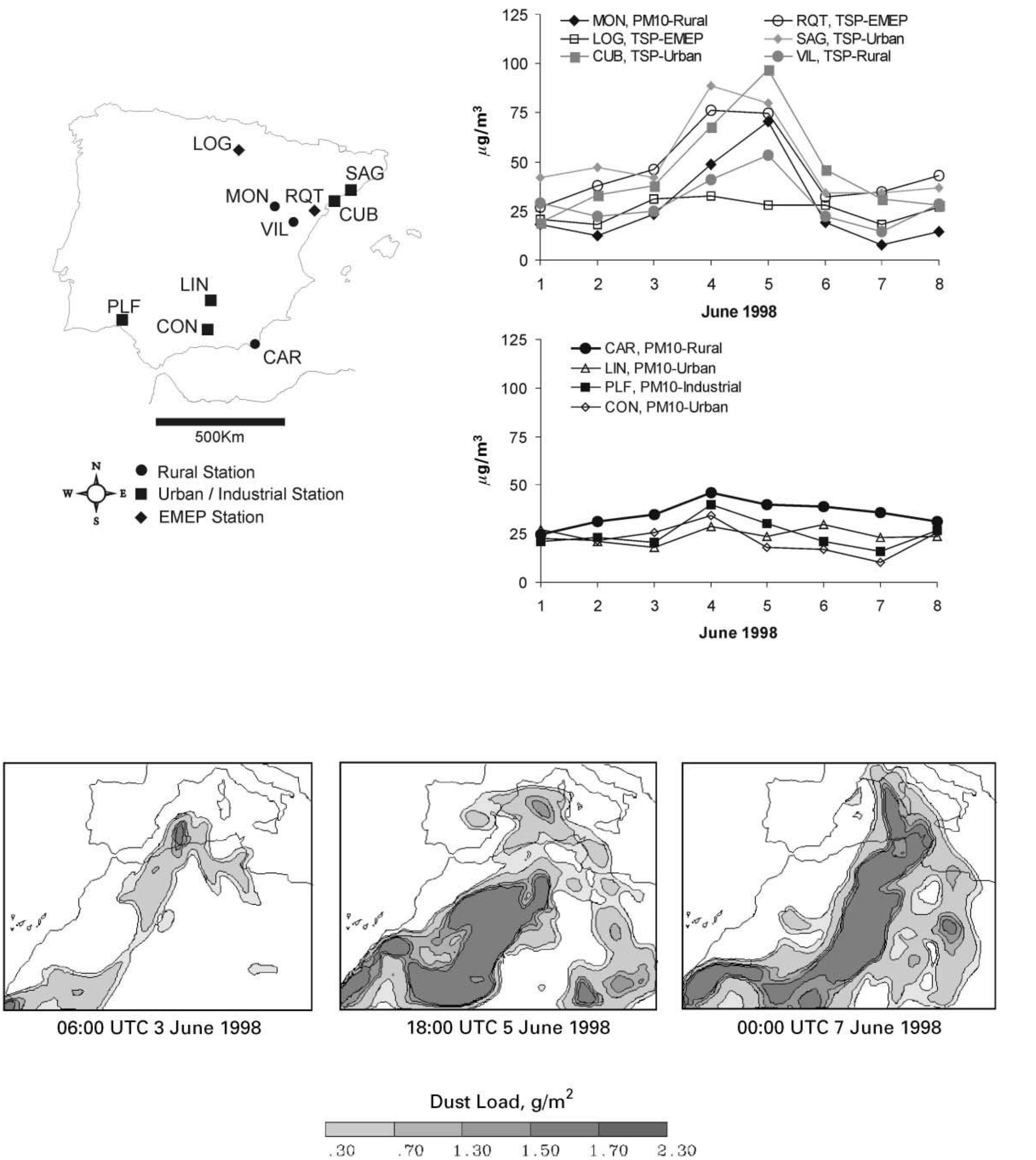

Fig. 4. Daily PM10 and TSP levels recorded at selected monitoring stations during the Saharan event from 4 to 5 June 1998 and evolution of the dust load (vertically integrated dust concentration in $g\,m^{-2}$) from the SKIRON model.

During September and October, Saharan intrusions over Iberian Peninsula were mostly induced by low-pressure systems (97-8, 97-9 and 97-10, all cases in 1997) or by the simultaneous occurrence of a western/south-western depression and eastern anticyclone (99-10 and 99-11).

Most of the Saharan intrusions coupled with important rain events are probably not considered here given that these events have no significant impact on ambient air particulate levels, and consequently have not been identified in the PM10/TSP time series. As for the seasonal distribution of the events having major impacts on

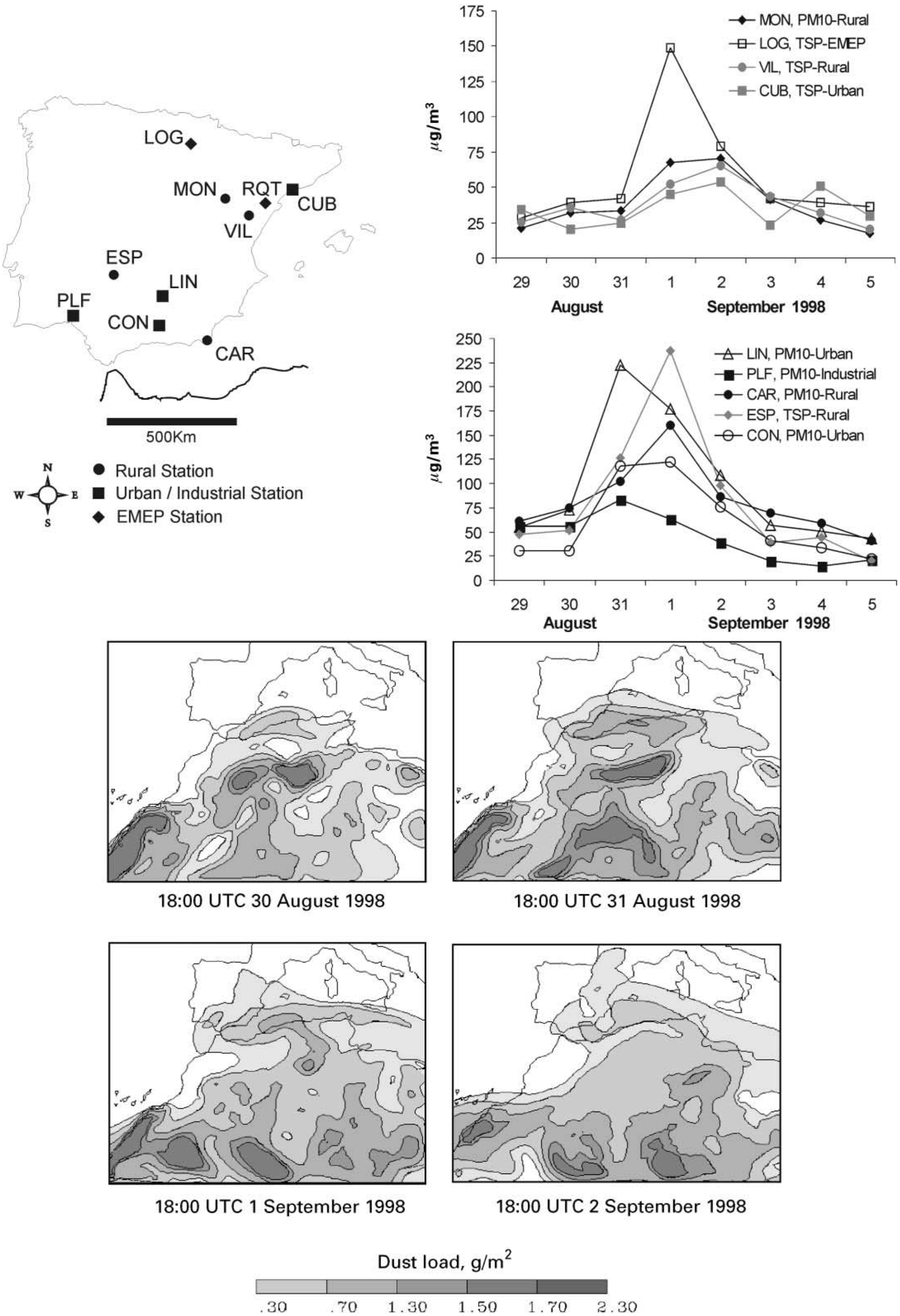

Fig. 5. Daily PM10 and TSP levels recorded at selected monitoring stations during the Saharan event from 30 August to 2 September 1998 and evolution of the dust load (vertically integrated dust concentration in $g\,m^{-2}$) from the SKIRON model.

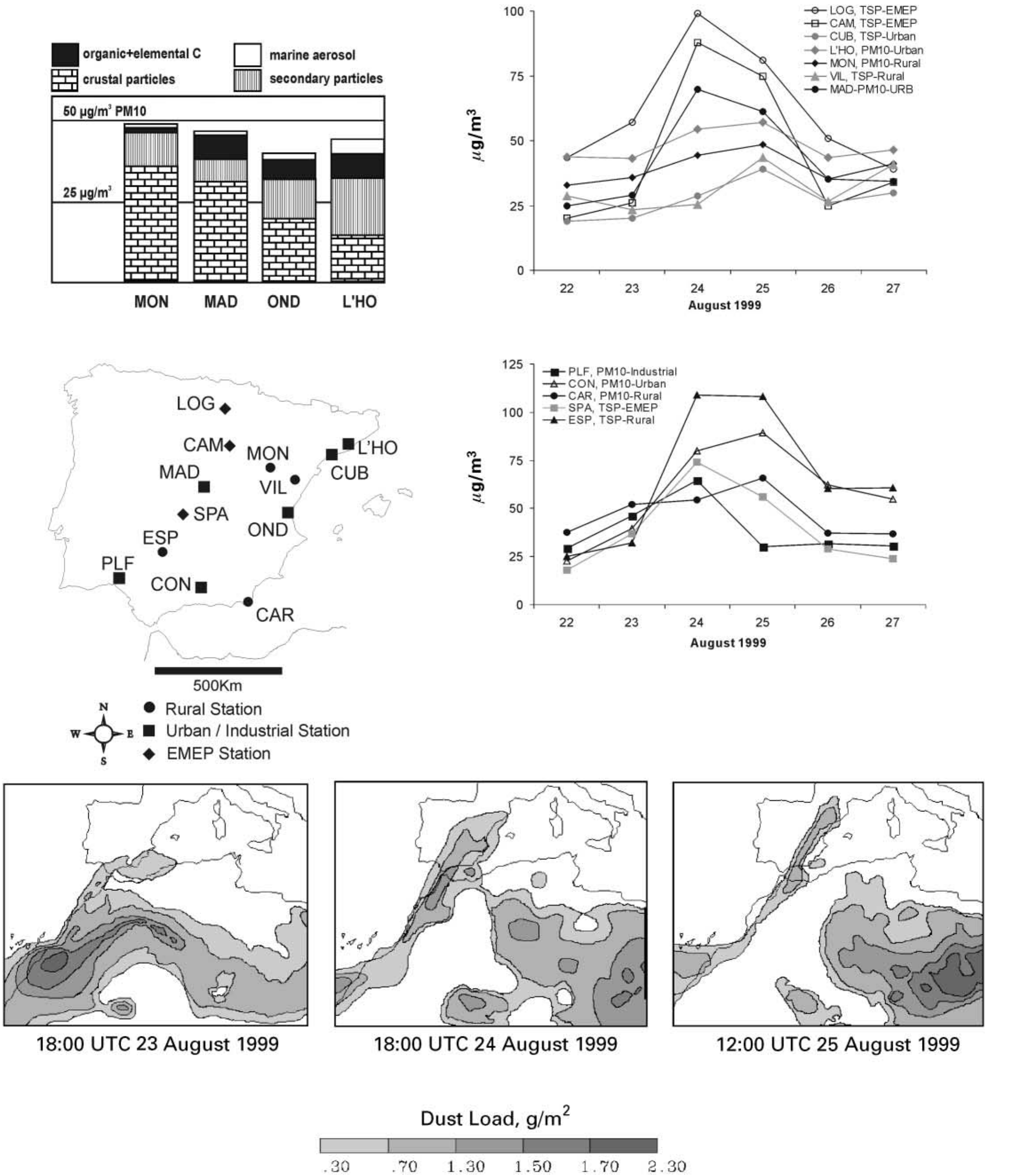

Fig. 6. Daily PM10 and TSP levels recorded at selected monitoring stations during the Saharan event from 23 to 28 August 1999 and evolution of the dust load (vertically integrated dust concentration in $g\,m^{-2}$) from the SKIRON model. The results of the PM10 source apportionment analysis obtained simultaneously at the urban stations of Madrid and Barcelona (L'Hospitalet), the industrial station of Onda and the rural station of Monagrega in the 24 h sampling from 25 to 26 August 1999 are included.

the records of particulate levels, most of the Saharan dust outbreaks (63%) reaching the Iberian Peninsula occurred in the period May–September. January, May, June, August and October exhibited the highest probabilities for dust outbreaks during the study period. The number of events range from 8 to 11 per year, each event lasting from 2 to 7 days (Table 2), with a mean duration of 3–4 days. Consequently, the number of days per year affected by particulate matter input from the Sahara ranges from 38 to 53 days for the study period (Table 2).

Table 3
Mean annual daily PM10 ($\mu g\,m^{-3}$), total number of days exceeding the new EU daily limit value ($50\,\mu g PM10/m^3$, $N > 50$), and exceedances produced during Sahara events

	Annual mean	$N > 50$	
		Total	Sahara
Monagrega (rural)			
1996	17	1	1
1997	18	4	4
1998	18	7	7
1999	17	4	3
Carboneras (rural)			
1996	23[a]	6[a]	1
1997	29	18	10
1998	37	48	23
1999	30	22	14
Palos de la F. (industrial)			
1996	30	30	9
1997	32	32	11
1998	35	67	11
1999	32	35	9
Linares (urban)			
1996	28	31	7
1997	34	56	17
1998	38	86	26
1999	35	66	27

[a]No data available during April and May 1996.

3.3. PM10 levels and implications for the new European standards

Table 3 reports the PM10 annual mean daily values for 1996–1999, the number of daily exceedances of the EU limit value registered in selected monitoring stations (future threshold value of the new EU standards) and the number of exceedances registered during the Saharan events. At the Monagrega rural station (MON) the PM10 annual mean (17–18 μg PM10 m^{-3}) and the number of exceedances (4–7 days per year) of the limit value are slightly lower than those of the European Directive (20 μg PM10 m^{-3}, and 7 days per year, respectively). Most of these exceedances may be attributed to Saharan events since only one exceedance occurred outside the Saharan dust outbreaks and was probably related to farming activities in the vicinity. Most of the days with daily means $> 45\,\mu g\,PM10\,m^{-3}$ were also recorded during Saharan events (from 4 to 13 days per year).

At the Carboneras rural station (CAR) both the PM10 annual mean (29–37 μg PM10 m^{-3}) and the number of exceedances (18–48 days per year) of the limit of the European Directive were surpassed. Approximately 55% of the daily exceedances of the EU limit value for PM10 occurred during Saharan episodes, 10, 23 and 14 exceedances for 1997, 1998 and 1999, respectively. The 1996 PM10 time series is excluded from this evaluation owing to the absence of records for the period April and May. As for Carboneras, in the Linares urban station (LIN) both the PM10 annual mean (28–38 μg PM10 m^{-3}) and the number of exceedances (31–86 days per year) of the limit of the European Directive were surpassed. Approximately 30% of the daily exceedances of the EU limit value for PM10 occurred during Saharan episodes, 7, 17, 26 and 27 exceedances for 1996, 1997, 1998 and 1999, respectively. The evaluation of the records of particulate levels obtained at the industrial station of Palos de la Frontera (PLF) yielded results that were very similar to those at CAR and LIN. Thus, daily PM10 means ranged from 30 to 35 $\mu g\,m^{-3}$ and daily exceedances from 30 to 67 times per year. Approximately 23% of the exceedances (9–11 times per year) occurred during Saharan events.

The lower number of Saharan-induced exceedances of the PM10 limit value recorded at the MON rural station (Northeastern Spain) when compared with the CAR rural station (southern Spain) could be attributed to the proximity of the last station to the African continent. Moreover, the homogeneity of the daily annual PM10 values and the number of daily exceedances obtained at the rural (CAR), urban (LIN) and industrial (PLF) stations in Southern Spain demonstrate the significance of the natural dust load in this region.

The seasonal distribution of the exceedances of the daily PM10 EU limit value may provide useful information on the possible origin of non-Saharan exceedances. As shown in Fig. 1 the exceedances recorded at the CAR rural station occurred mainly in summer. At the stations with a higher anthropogenic influence, such as the LIN (urban) and the PLF (industrial) stations, the seasonal pattern of the exceedances also shows a high summer frequency but in addition a winter mode is observed (Fig. 7). The higher summer PM10 levels contrasts with the typical seasonal pattern of urban and industrial environments in Central and Northern Europe (Monn et al., 1995). In these Northern areas, higher particulate levels are registered in winter due to enhanced anthropogenic emissions coupled with lower vertical dispersion potential (stagnant atmospheric conditions induced by inversions and weak convective activity, Monn et al., 1995). The second-order winter mode of exceedances of the EU limit value recorded at the urban and industrial stations of LIN and PLF (Fig. 7) probably occurs in this classic pollution scenario. However, the first-order summer mode of exceedances recorded in all southern stations cannot be attributed to these two factors given the high dispersive conditions resulting from the intensive summer convective dynamics of the study area. In addition to the aforementioned higher frequency of Saharan events in June–August, the high PM10 summer levels are

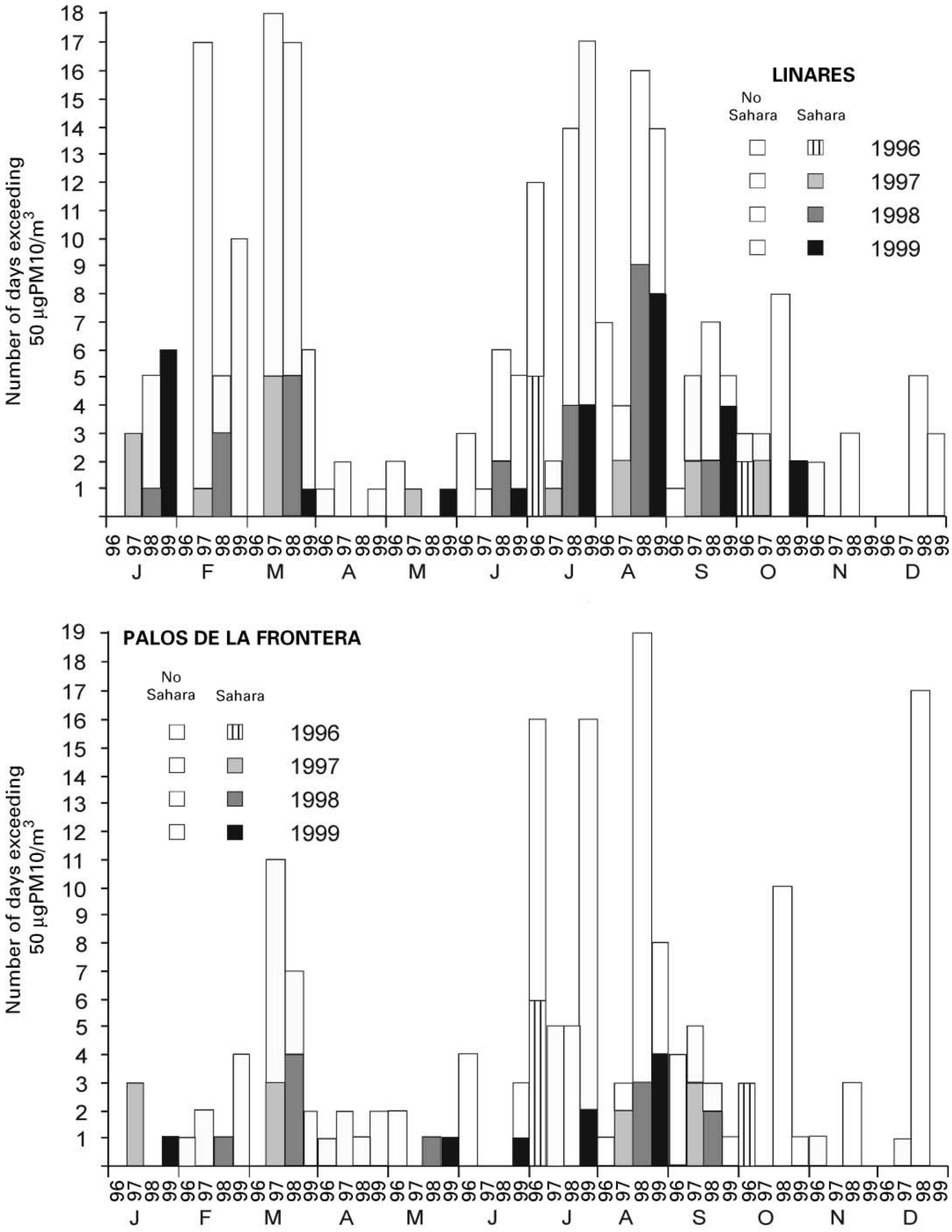

Fig. 7. Monthly variation of the number of days exceeding the daily limit value for PM10 ($50\,\mu g\,m^{-3}$) of the new EU Directive (1999/30/CE) at an urban (LIN) and an industrial (PLF) station from 1996 to 1999.

probably related to: (1) high photochemical conversion rate of gaseous pollutants to secondary aerosols (such as nitrates and sulphates), (2) high mineral dust load from soils induced by intense atmospheric convective dynamics and anthropogenic induced re-suspension, (3) lower rainfall rate which reduces the particulate scavenging potential and (4) an atmospheric particulate reservoir effect caused by a scarce renovation of the atmospheric masses in the Western Mediterranean basin (Millán et al., 1997).

4. Conclusions

The results of this study demonstrate that the anthropogenic contribution to atmospheric particulate matter in Southern and Northern European regions cannot be monitored with one criterion given the higher natural input in the Mediterranean countries. This higher natural load accounts for the different chemical composition and for the contrasting seasonal evolution of particulate levels, which must be taken into account when monitoring ambient air PM10 levels. Analogous differentiation in the source apportionment of atmospheric particulate matter has been highlighted in United States. In Western US, PM10 shows on average 36% of mineral load, whereas in Eastern US only 19% of PM10 is supplied by this particulate source (US-EPA, 1996). In certain Western cities, dust load reaches 80% of bulk PM10 (Gertler et al., 1995).

Current research on the PM10/PM2.5 monitoring in Spain shows that monitoring of PM2.5 instead of PM10 may avoid the interference of crustal particles without a major reduction in the secondary anthropogenic load, with the exception of nitrate. However, the PM2.5 measurement is not an adequate methodology for monitoring the impact of primary particulate emissions (such as ceramic, mining or cement emissions) on air quality since major ambient air particles derived from these emissions are mainly in the range of 2.5–10 μm (Querol et al., 2001).

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