Chapter 4

PM events and seasonal evolution

The EU Directive 1999/30/CE established limit values for particulate matter with aerodynamic diameter <10µm (PM10) which are progressively more restrictive from 2001 to 2010. After the 1st of January 2010, annual PM10 concentrations should not exceed 20µg/m^3, and the daily limit value of 50µg/m^3 should not be exceeded more than 7 days/year. This implementation will take place through an intermediate stage (2005) when the annual limit value and the number of permitted exceedances of the daily limit value will be 40µg/m^3 and 35 days/year, respectively. Although these PM10 limit values are established for the whole of Europe, in Southern regions a higher natural background PM10 load may be expected from the frequent occurrence of African dust outbreaks and from the local re-suspension of mineral soil particles (Querol et al., 1998b).

Levels and composition of PM10 in Europe is influenced by regional variations in: a) natural and anthropogenic particulate emissions, b) ambient conditions (e.g. insolation, temperature, humidity) and levels of reactive gases (e.g. OH or O_3) influencing the gas to particle conversion, and in c) regional atmospheric dynamics controlling transport and dispersion of pollutants. Many of these factors affect South and Central-Northern Europe in different ways. Thus, Central-Northern Europe, mostly characterised by a flat terrain, is mainly affected by the westerly winds that are frequently associated with eastward moving depressions, cold fronts and rain. These factors favour dilution of pollutants, air mass renovation and pollutant scavenging. In contrast, the Western-Central Mediterranean, surrounded by high coastal ranges (see Figure 4-1), frequently undergoes weak baric gradient conditions. Under this scenario, the intense heating of the land promotes the breeze circulation and the development of meso-scale processes, favouring the ageing of pollutants by restraining the air mass renovation (Regional events in accordance with Millán et al., 1997, 2000, Toll and Baldasano, 2000, Soriano et al., 1988, 2001, and Gangoiti et al., 2001). The lower frequency of Atlantic advections in the Western Mediterranean than in Northern Europe also accounts for a lower scavenging potential and the consequent longer residence time of suspended particulate matter in the atmosphere.

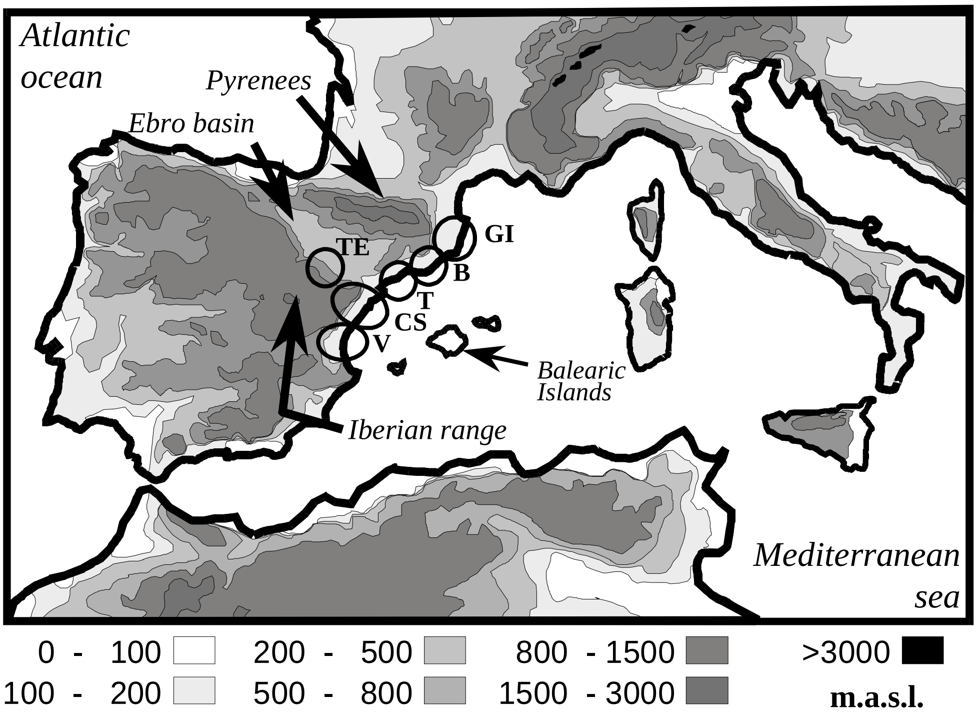


Figure 4-1. Location of the PM monitoring stations grouped by provinces (GI: Girona, B: Barcelona, T: Tarragona, CS: Castelló, V: Valencia, TE: Teruel).

4.1 Events affecting PM concentrations

Figure 4-2 and Table 4-1 show a statistical analysis of the TSP and PM10 concentrations recorded in selected monitoring stations located in the study area (Figure 4-1). TSP (or PM10 at MON rural site) data were available from 1996 to 2000 at most of the stations, but only 1997-2000 data were available at two sites, and TSP data were available for 1996-1999 and PM10 for 2000 at two sites.

As expected, mean levels of TSP showed an increasing trend from the rural, urban to industrial sites. At rural sites, mean TSP concentrations ranged between 14 and 20µg/m^3, and mean PM10 levels reached 17µg/m^3 at MON (rural site). The lowest mean TSP level was recorded at the COR rural-background site located at the top of the Catalan range. The annual mean concentrations showed a low variability, annual means in the ranges 17-18µgPM10/m^3 at MON, 12-16µgTSP/m^3 at COR, 19-21µgTSP/m^3 at MOR and 19-23µgTSP/m^3 at VIL.

Mean TSP levels were found in the 30-60µg/m^3 range for most urban sites, and in the 60-96µg/m^3 range for industrial sites. The annual TSP levels showed a much higher variability in the urban and industrial environments than at the rural sites. In some cases, the annual concentrations showed a markedly decreasing trend as a result of the PM emission abatement strategies, e.g. TSP levels decreased by 39, 25 and 29% from 1997 to 1999, respectively, at S.AN, IGU and G.VIA (see appendix 1). The Spanish standard for TSP was not exceeded at any of the stations in the study period.

Table 4-1. PM monitoring stations selected for the study with location information, PM data availability for 1996-2000 (DA in %) and PM (TSP or PM10) mean values. P=1996-2000, P1=1996-1999, P2=1997-2000, P3=2000. Alt = Altitude in meters above the sea level, Lat. = Latitude in degrees, Long. = Longitude in degrees. See location in Figure 4-1 (location code per provinces GI: Girona, B: Barcelona, CS: T: Tarragona, TE: Teruel, CS: Castelló, V: Valencia).

Station	Location	Alt., Lat., Long.,	DA(%),	PM (µg/m^3)
CORATXAR (COR), Rural background	CS	1235, 40.6ºN, 0.0º	89%	TSP^{P2} .= 14
MONAGREGA (MON), Rural background	TE	537, 40.9ºN, 0.2ºW	98%	$PM10^{P}$ = 17
MORELLA (MOR), Rural background	CS	1153, 40.6ºN, 0.0º	82%	TSP^{P} = 20
VILAFRANCA (VIL), Rural background	CS	1125, 39.7ºN, 0.1ºW	97%,	TSP^{P2} = 20
CUBELLES (CUB), Sub-urban coastal site	B	15, 41.2ºN, 1.2ºE	89%	TSP^{P} = 26
FORNELLS (FOR), Sub-urban, inland	GI	101, 41.9ºN, 2.8ºE	89%	TSP^{P} = 30
PENYETA (PEN), Sub-urban, coastal site	CS	93, 40.0ºN, 0.0º	90%	TSP^{P} = 33
L'HOSPITALET (L'HO), Urban canyon street	B	30, 41.4ºN, 2.1ºE	78%	TSP^{P1} = 46
			93%	$PM10^{P3}$= 49
SAGRERA (SAG), Urban canyon street	B	10, 41.4ºN, 2.2ºE	83%	TSP^{P1} = 46
IGUALADA (IGU), Urban-industrial, inland	B	320, 41.6ºN, 1.6ºE	83%	TSP^{P} = 52
MARTORELL (MAR), Urban-industrial, coastal valley	B	40, 41.5ºN, 1.9ºE	93%	TSP^{P} = 53
GRAN VIA (G:VIA), Urban, canyon street	V	11, 39.5ºN, 0.4ºW	96%	TSP^{P} = 59
GANDIA (GAN), Urban, canyon street	V	22, 38.9N, 0.2ºW	93%	TSP^{P2} = 77
ONDA (OND), Industrial background, coastal valley	CS	167, 39.9ºN, 0.2ºW	90%	TSP^{P2} = 57
BONAVISTA (BON), Industrial, coastal site	T	40, 41.0ºN, 1.1ºE	93%	TSP^{P} = 60
SAGUNTO (SAG), Industrial, coastal site	V	10, 39.6ºN, 0.2ºE	80%	TSP^{P} = 60
ERMITA (ERM) , Industrial, coastal site	CS	21, 39.9ºN, 0.0º	89%	TSP^{P} = 77
MONTCADA (MOT), Industrial, inland	B	33, 41.5ºN, 2.2ºE	82%	TSP^{P} = 79
S.ANDREU (S.AN), Industrial, coastal valley	B	33, 41.4ºN, 1.9ºE	80%	TSP^{P1} = 96
			93%	$PM10^{P3}$= 59

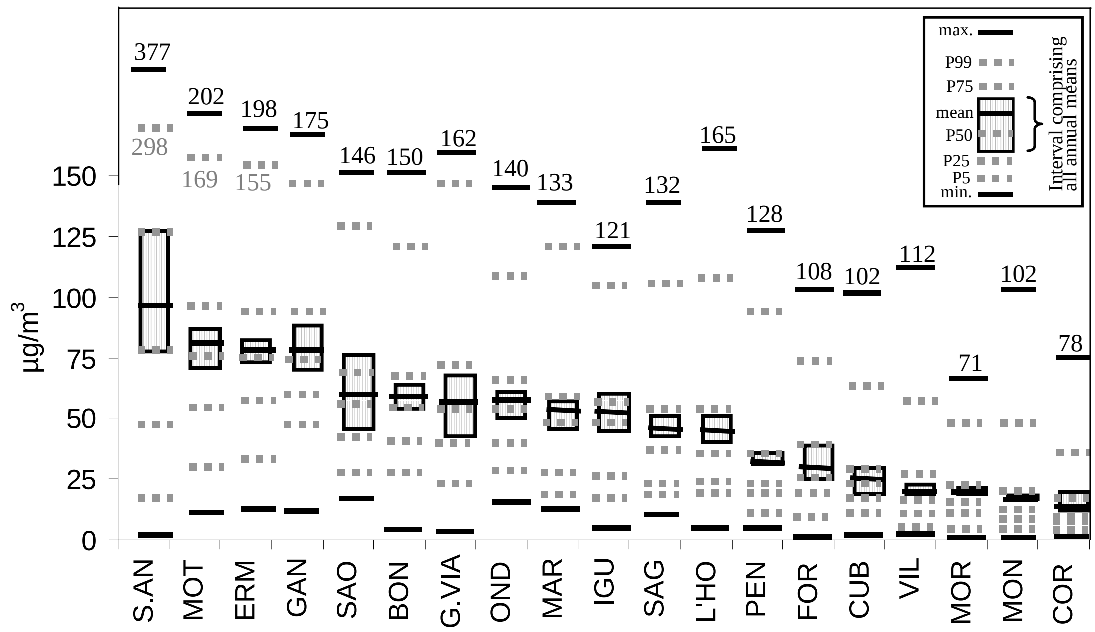


Figure 4-2. Statistic of TSP and PM10 (only at MON station, Table 4-1) concentrations in Eastern Spain.

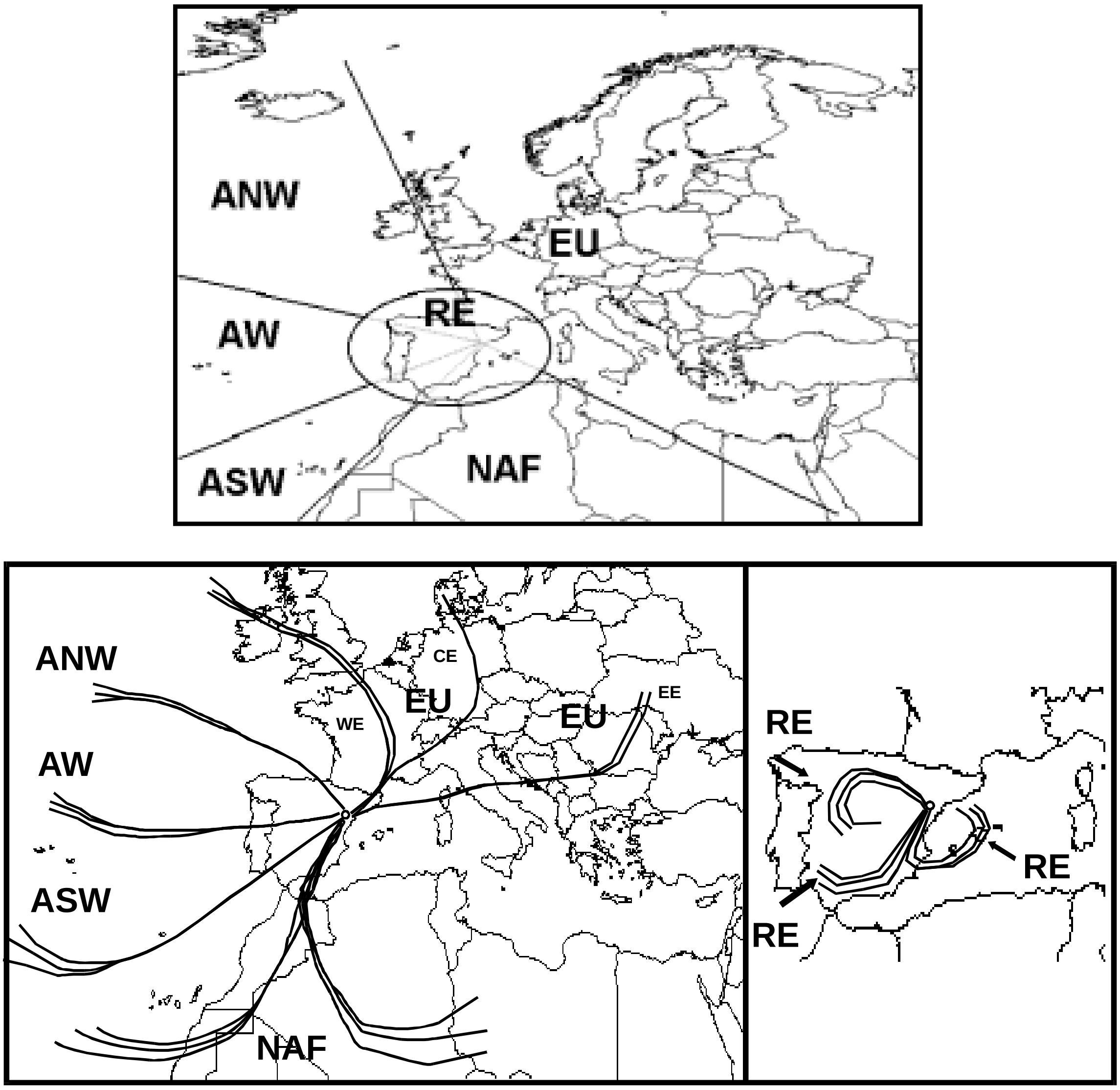


Figure 4-3. Upper: air masses source regions considered for the back-trajectories analysis (ANW: NW-Atlantic; AW: W-Atlantic; ASW: SW- Atlantic; NAF: North Africa; EU: Europe; RE: Regional). Below: Air back-trajectories associated with the typical transport patterns from each region.

Time series of daily TSP and PM10 (PM) concentrations from 1996 to 2000 were interpreted following the methodology described in chapter 3 - section 3.1. The main results are discussed below. Details on the PM events studied year per year (1996 to 2000) are presented in appendixes 2 to 4. Appendix 2 shows the result of the interpretation of the origin of the high and low PM episodes from 1996 to 2000.

The mean PM concentrations measured from 1996 to 2000 at most of the rural, urban and industrial sites in Eastern Spain for the Atlantic, Regional, European and African transport patterns (see Figure 4-3), and Local pollution episodes are plotted in Figure 4-4. Figure 4-5 shows the distribution of PM10 concentrations for each of the above episodes as measured at the MON rural site. The monthly number of days (1996 to 2000) with air masses arriving from

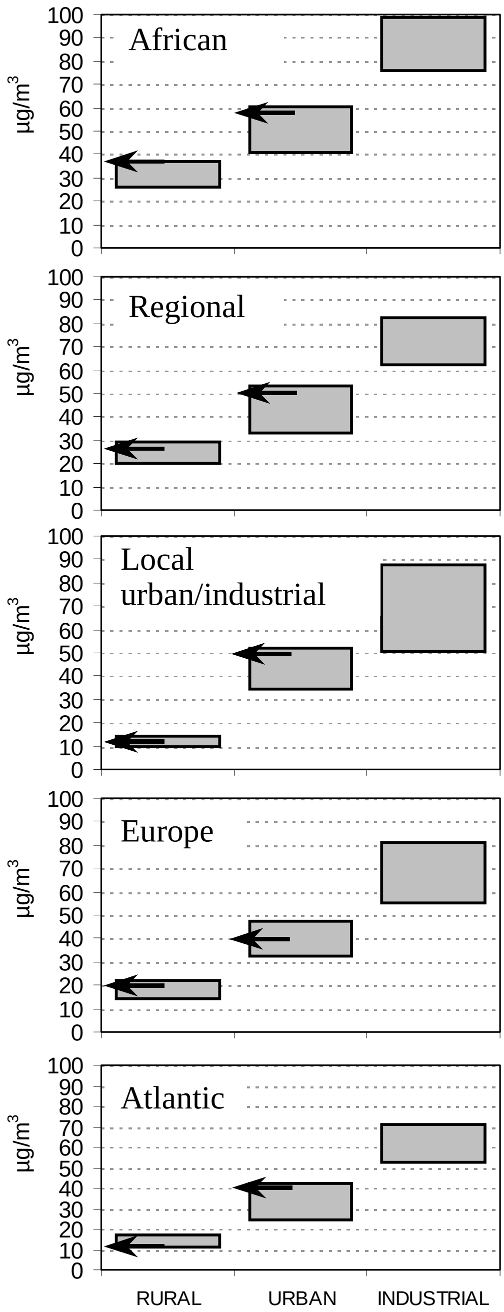


Figure 4-4

Figure 4-4. Range of mean TSP levels measured at rural (COR, MOR and VIL), urban (SAG, L'HO, PEN and FOR) and industrial (OND, BON and MOT) sites during African, Regional, Local urban/industrial, European and Atlantic. The arrows highlight the mean PM10 concentrations at one rural site (MON: 1996-2000) and at the urban site (L'HO 2000).

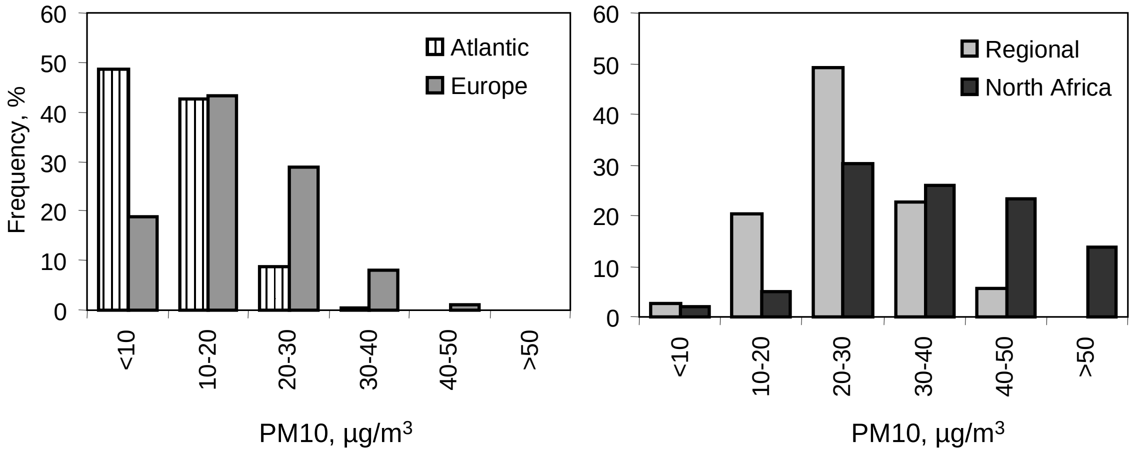


Figure 4-5. Frequency distribution of daily mean PM10 concentrations at the MON rural site (1996-2000, N= 1793) for each transport scenario: Atlantic (N=1036), European (N=354), Regional (N=257) and African (N=146).

different regions (Figure 4-3), and mean PM levels associated with each transport pattern is included in appendix 3.

4.1.1 Atlantic episodes

Low PM levels are measured at rural, urban and industrial sites during periods of intensive advections of Atlantic air masses. These Atlantic episodes over Eastern Spain take place under different meteorological scenarios, such as the North Atlantic anticyclone inducing intense entries of Atlantic air masses through the Gulf of Lion (e.g. Figure 4-6F, 4-7E, 4-8C), or Atlantic depressions developing to the North (e.g. Figure 4-6D) or West (e.g. Figure 4-7C) of the Iberian peninsula. Most of the intense Atlantic events are associated with cold fronts crossing Spain and rain. Since these episodes usually delimit high PM events, some of them will be described below together with examples of high PM episodes.

4.1.2 Regional episodes

During Regional episodes a simultaneous increase in PM levels takes place at the rural, urban and industrial sites. These episodes are developed under meteorological scenarios that are characterised by a lack of a significant air mass advection. Some typical patterns of these events are: a weak baric gradient over Eastern Spain and Western Mediterranean, short back-trajectories (over Eastern Spain, Western Mediterranean and/or the Iberian peninsula) and breeze circulation. During these episodes, the increase in PM levels at the rural sites is a consequence of the transport of pollutants from the urban/industrial areas. The frequent simultaneous increase in temperature and ozone levels with PM concentrations also indicates a regional origin of these events due to Millán et al. (1997, 2000) attributed the high ozone episodes in this region to thermally activated re-circulation processes of polluted air masses over Eastern Spain.

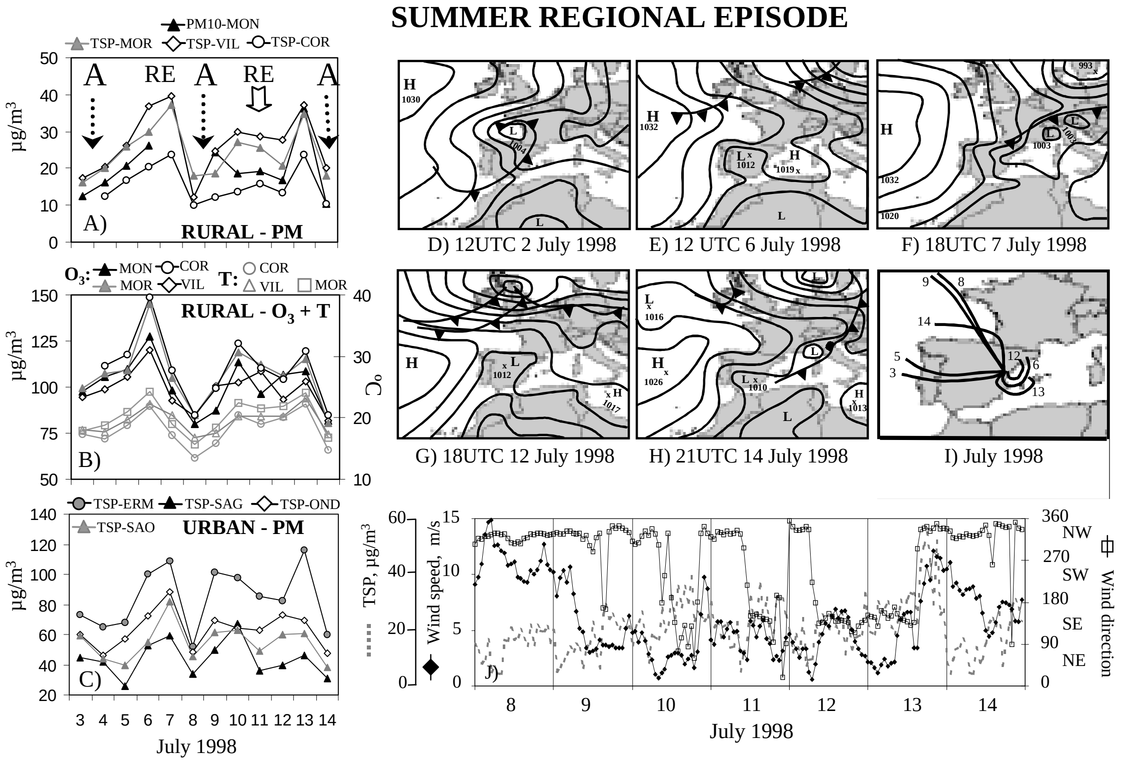


Figure 4-6. Example of a summertime Regional episode from July 1998. Left: Daily mean PM levels at rural and urban/industrial sites, and daily mean O_3 and T values at rural sites. A, Atlantic episode, RE, Regional episode. Top right: Synoptic charts of pressure (mb) at sea level during Atlantic (D, F, H) and Regional (E, G) episodes, and isentropic back-trajectories (I). Bottom right: Hourly values of wind direction and velocity, and TSP levels at the MOR rural site (J).

Under weak baric gradients over Eastern Spain and the Western Mediterranean, the heating of mountains and valley slopes induces the breeze circulation at coastal and mountain sites. The breeze favours the inland transport of pollutants (channelled into the valleys) from the eastern coast (where the main urban/industrial areas are located) to the inland rural sites during daylight, and the seaward return of the pollutants at night. Moreover, Millán et al. (1997, 2000) and Gangoiti et al.(2001) argue that polluted air masses experience vertical re-circulations in this area due to 1) the inland up-slope winds during daylight in the coastal ranges, 2) the dominant westerly winds over the coastal ranges, 3) the subsidence over the Western Mediterranean sea, and to 4) the inland entry of the breeze during the following days. The persistence of this breeze circulation setting for relatively long periods (up to 2 weeks were observed in summer) results in the accumulation of the airborne particulates on a regional scale because of a scarce renovation of air masses.

The Regional events occur at a rate of 12 major events per year, with a mean duration of 4-6 days. Although these episodes occur mainly from March to October, summer events are more frequent, have a longer duration, and reach higher PM levels (20-45µg/m^3 in summer) at rural sites. From June to September most of the Regional episodes occur under the typical

SPRING REGIONAL EPISODE

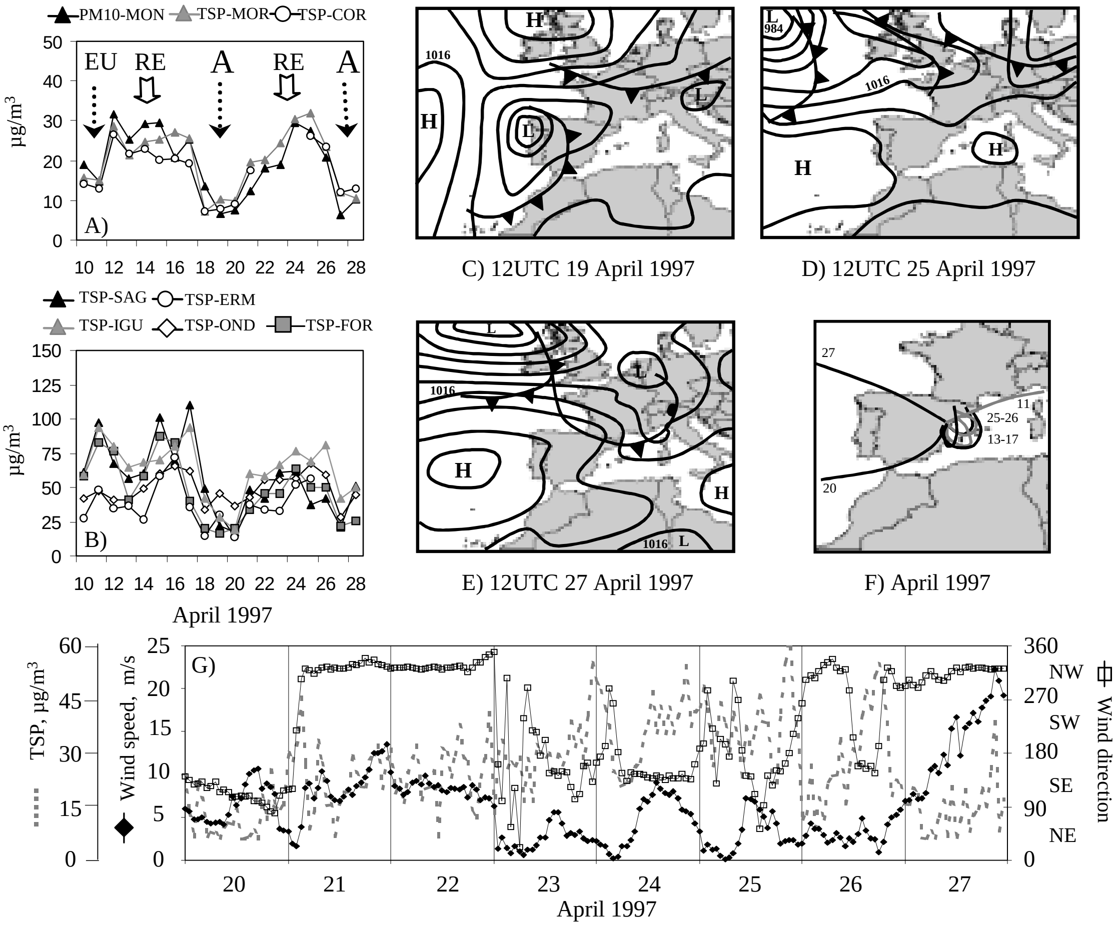


Figure 4-7. Example of a spring Regional episode in April 1997. Top left: Daily mean PM levels at rural and urban/industrial sites. EU, European episode, A, Atlantic episode, RE, Regional episode. Top right: Synoptic charts of pressure (mb) at sea level during Atlantic (C, E) and Regional (D) episodes, and isentropic back-trajectories (F). Bottom: Hourly values of wind direction and velocity, and TSP levels at the MOR rural site (J).

summer-time meteorological scenario: the North Atlantic anticyclone affecting Western Europe and a weak baric gradient over Western Mediterranean, frequently associated with the Iberian thermal low development.

Figure 4-6 shows a typical summer Regional episode in the periods July 6-7 and July 10-13 1998. From July 5 to 7 and 10 to 13 an increase in the temperature and in PM and ozone concentrations was recorded. In these two periods the typical summer meteorological scenario associated with an intense breeze circulation prevailed (see synoptic meteorology in Figure 4-6E and G and wind speed and direction at the MOR station in Figure 4-6J). Cool Atlantic air masses were advected towards the Western Mediterranean (Figure 4-6F) in the periods July 2-3, 7-9 and 14 (Figure 4-6D, F, H and I) resulting a drop in temperature, and in PM and ozone concentrations (Figure 4-6A, B and C).

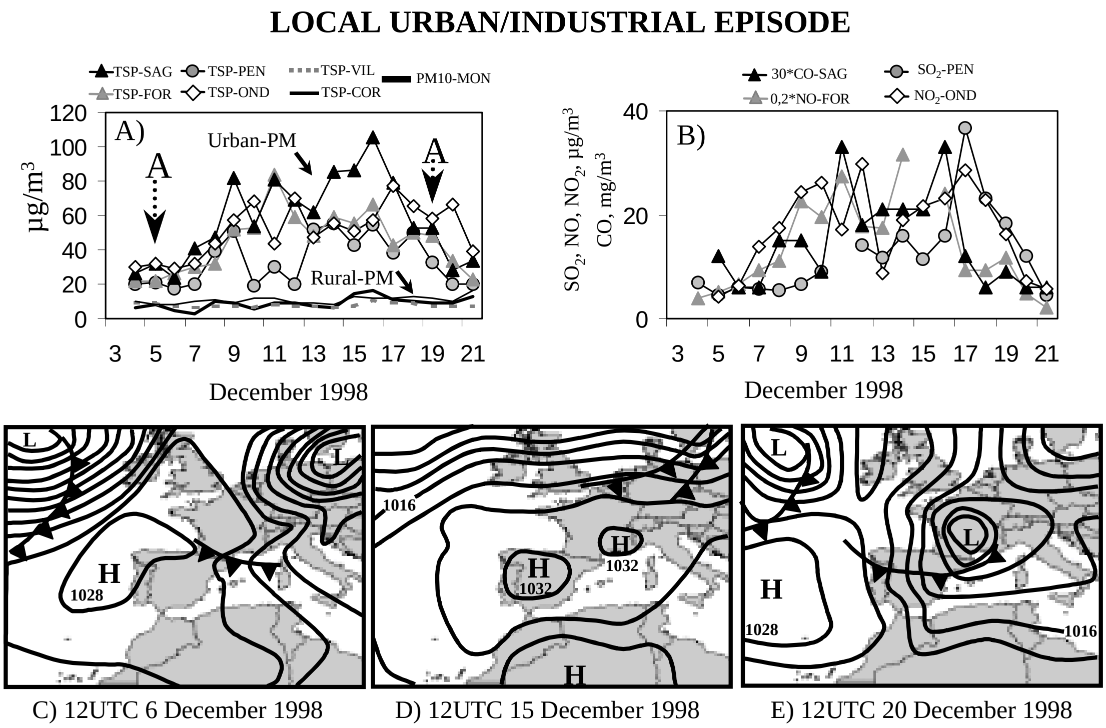


Figure 4-8. Example of a Local urban/industrial pollution episode from December 1998. Top left: Daily PM levels at urban/industrial and rural sites. A: Atlantic episode, Local U/I: Local urban/industrial pollution episode. Top right: Daily levels of gaseous pollutants at urban and industrial sites. Bottom: Synoptic charts of pressure (mb) at sea level during Atlantic (C, E) and Local urban/industrial (D) episodes.

In the semi-warm periods (March to May and October) the intense Regional episodes occur under a similar summer meteorological scenario but in most cases the Iberian thermal low is not developed. PM concentrations ranging from 20 to 30µg/m^3 are usually recorded during the Regional spring/autumn episodes at rural sites. Figure 4-7 shows examples of successive Regional and Atlantic episodes for April 1997. After the Regional event from April 12-17, an Atlantic episode (Figure 4-7A and C) induced low PM concentrations. From April 22 to 26, weak baric gradient conditions over the Iberian peninsula favoured the breeze circulation (Figure 4-7D and G), and a Regional episode was again developed. On April 27 cool Atlantic air masses reached the Western Mediterranean, diminishing PM concentrations at rural (Figure 4-7A) and urban/sites (Figure 4-7B).

4.1.3 Local urban/industrial pollution episodes

During Local urban/industrial pollution episodes the difference between urban and rural PM10 events reached maximum values. These events are associated with high PM levels at urban and industrial sites and simultaneous low levels at rural sites, as a consequence of the predominance of locally emitted PM over the external inputs. These events mainly occur from November to February.

Most of these Local urban/industrial events occur when the North Atlantic anticyclone expands or is present over the Iberian peninsula. Some of these events may also occur under weak gradient conditions over Eastern Spain and the Western Mediterranean.

The marked difference between the PM levels measured at the urban/industrial and rural sites is a consequence of the inhibition of the dispersive mechanisms in the cold season, most of which are thermally activated. In Eastern Spain most of the large urban and industrial settlements are located on the coastal flat, between the shore and the coastal ranges (e.g. Barcelona, Tarragona, Castelló or Valencia in Figure 4-1). Thus, in the warm season the breeze circulation favours the dispersion of pollutants and the transport towards rural sites (Baldasano et al., 1994, Millán et al., 1997), but not in the cold seasons when these thermal circulations reach the minimum development. Furthermore, in the cold season, the thinning of the continental mixing layer favours the concentration of pollutants in inland urban environments. The less intense heating of the land in winter and the frequent occurrence of temperature inversion layers near the ground inhibit the vertical dilution of pollutants. Figure 4-8 shows an example of a Local urban/industrial episode from December 9-19 1998. During this episode, high concentrations of TSP and gaseous pollutants (CO, NO, NO_2, CO) were recorded at the urban and industrial sites under a typical winter anticyclonic scenario (Figure 4-8). Most of the urban/industrial sites recorded TSP levels in the range of 40-90µg/m^3, whereas PM concentrations <15µg/m^3 were recorded at rural sites. Before and after this event, low PM levels were attributed to the occurrence of Atlantic episodes (Figure 4-8).

4.1.4 African dust episodes

African air mass transport over Eastern Spain is usually associated with very high PM concentrations at all the monitoring stations (Figures 4-4 and 4-5). At the rural sites the highest PM concentrations are recorded during these African episodes. At COR (1235 m.a.s.l. in the Catalan range) and MON (537 m.a.s.l. in the Ebro basin) daily mean concentrations >45µgTSP/m^3 and >50µgPM10/m^3 were recorded for 3-4 and for 1-7 days per year, respectively, during these episodes. African dust outbreaks over Eastern Spain were observed under meteorological scenarios characterised by a depression located to the West or Southwest of Portugal (most frequently in the January-June period) and/or an anticyclone located to the East or Southeast of the Iberian peninsula (most frequently in July-August, and some years in March). Detailed analysis on the African dust transport patterns and the dates of the most important African dust events are provided further in section 4.2. Approximately 10 African events per year with a mean duration of 3 days were recorded in 1996-2000. These episodes frequently occur from January to September, and in some years in October, with a higher frequency in summertime. Heavy episodes occurred in March in 3 of the 5 years studied. Important African events were not recorded in November and December during the study period.

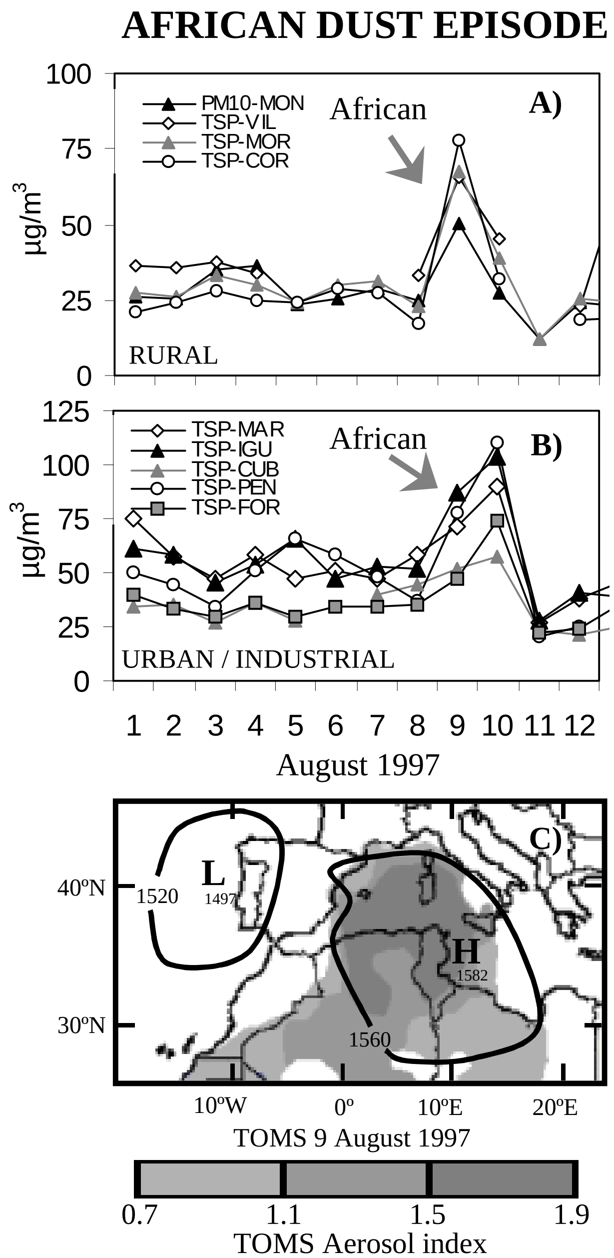


Figure 4-9. Example of an African dust episode from August 1997. A and B) Daily PM levels at rural and urban/industrial sites. C) TOMS map and 850hPa synoptic chart (altitude in m.a.s.l. of the 850 mb level at 12UTC) for 9 August 1997.

Figure 4-9 shows an example of an African dust outbreak from August 9-10 1997 caused by the simultaneous presence of a depression to the west of Portugal and an anticyclone over Algeria. This short event (2 days) is selected owing to the extremely high TSP concentration recorded at the COR rural-background site: 78µg/m^3 daily mean. At the MON rural site the daily PM10 concentrations increased from 25µg/m^3 the day before the event to 50µg/m^3 during the event.

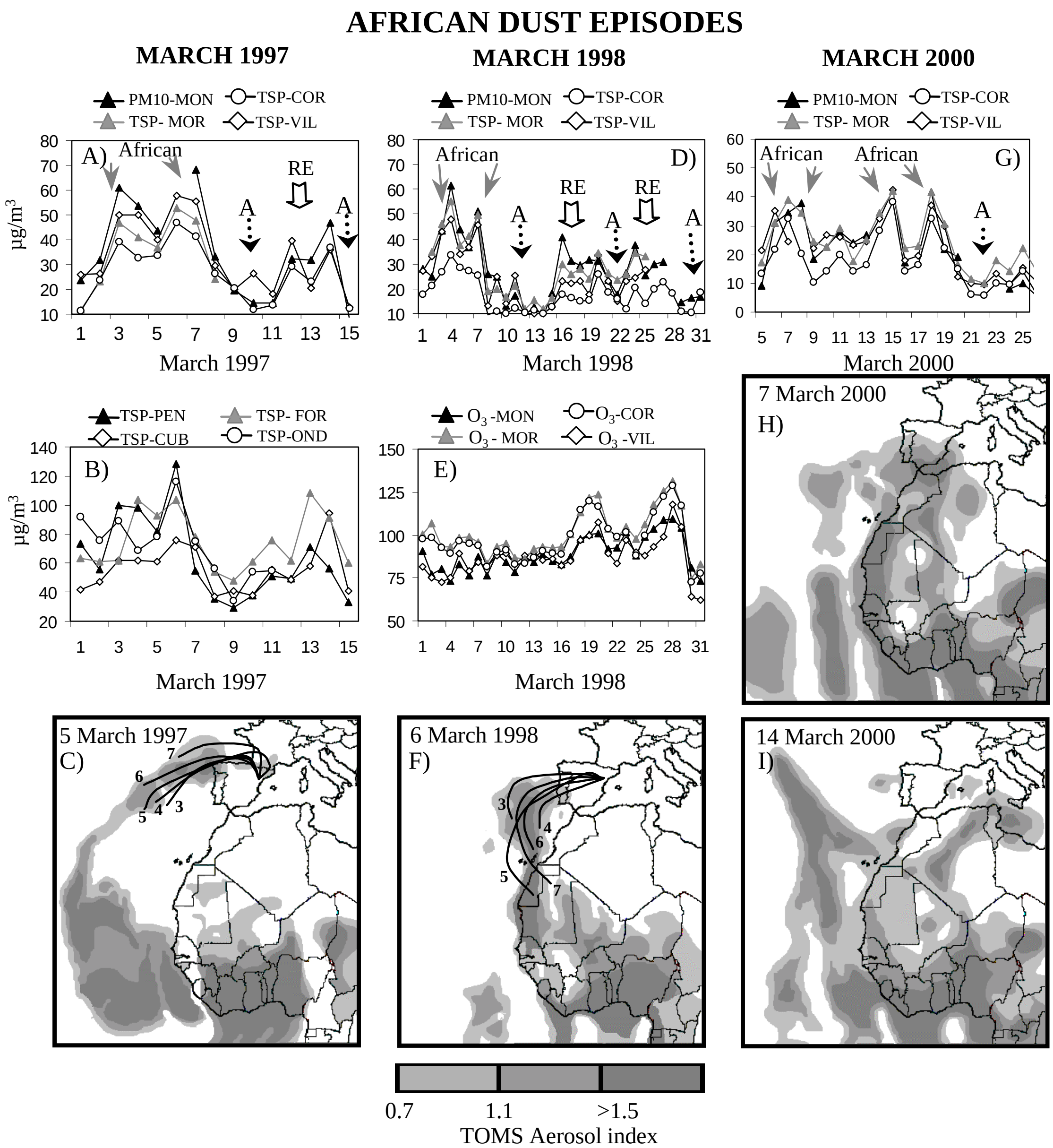
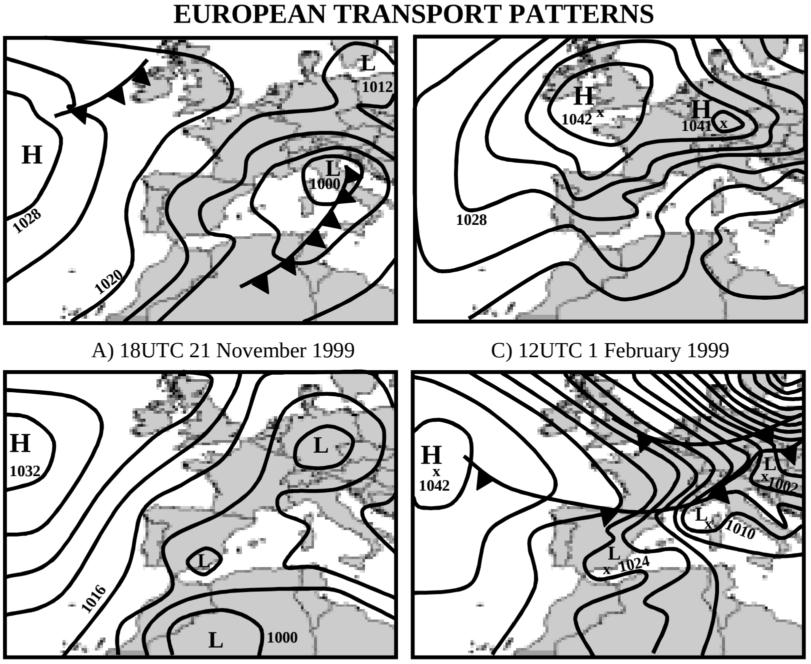


Figure 4-10. March African dust episodes for 1997, 1998 and 2000. Daily PM levels at rural (A, D and G) and urban/industrial (B) sites. Daily O_3 levels at rural sites (E). TOMS maps for March 5 1997 (C), March 6 1998 (F) and March 7 and 14 2000 (H and I). Overlapped over TOMS maps are plotted back-trajectories for March 3-7 1997 (C) and March 3-7 1998 (F).

Figure 4-10 shows the Sahel dust outbreaks in March 1997, 1998 and 2000. These events have one factor in common: the African dust is transported from the Sahel region to the Iberian peninsula via the Atlantic ocean. The episodes in 1997 and 1998 were caused by an anticyclone which spread over North Africa and the Eastern Iberian peninsula. A similar scenario was described by Schwikowski al. (1995) for an African dust event that reached the Alps via the Atlantic in March 1990. The southerly winds giving rise to the African dust event from March 6-8 2000 was caused by the NW Africa anticyclone and a cyclone over the Canary Islands.



A) 18UTC 21 November 1999 C) 12UTC 1 February 1999

B) 12UTC 1 May 1998 D) 18UTC 5 February 1999

Figure 4-11. Meteorological scenarios favourable for the transport of air masses from mainland Europe.

The northward movement of the cyclone led to the events from March 14-15 and 18-19 2000.

These intensive African dust events identified in March in three of the five years studied agree with the results of previous studies. The period February to April, when the African dust is mainly generated in the Sahel region, has been identified as a period of intensive dust outbreaks over the NW coast of Africa and the Eastern North Atlantic (Swap et al., 1996). Herman et al. (1997) reported a dust event in March 1988 similar to that shown in Figures 4-10C and 4-10F.

African and Regional episodes account for the first and second highest PM events at the rural sites, respectively. PM episodes are usually associated with ozone peak levels in Regional events but not in African episodes (see Figure 4-10E). Even slight decreases in ozone concentrations may be recorded during intensive African dust outbreaks.

4.1.5 European episodes

The transport of particulate pollutants from mainland Europe to Eastern Spain was considered a priori as a potential scenario for high PM episodes, as reported for other EU regions, such as the UK (Stedman, 1997) or Finland (Pakkanen et al., 2001). However, as Figures 4-4 and 4-5 show, European transport patterns present the second lowest mean PM concentrations. This is probably due to the fact that some of the meteorological scenarios inducing the transport of air masses from mainland Europe to Eastern Spain are associated with wet/rainy weather situations and strong winds which favour pollutant dilution and scavenging during air mass transport. A depression developing over Italy (Figure 4-11A) or mainland Europe (Figure 4-11B) induces the advection of air masses from Central Europe to Eastern Spain, and rain, favouring air mass scavenging. For example, episodes of advections of air masses from mainland Europe caused by cyclonic circulation occurred in November 20-23 1999 (Figure 4-11A) and May 1-5 1998 (Figure 4-11B). During these dates low PM levels were recorded at the rural stations (around 9 and 15 µgPM10/m^3 during the November 1999 and May 1998 episodes at MON, respectively, and around 9 and 12 µgTSP/m^3 at COR during these episodes).

Advections of air from mainland Europe may also be associated with strong winds and cold fronts (Artic air masses reaching the Mediterranean), favouring the cleaning of the atmosphere (Figure 4-11D). One of these events occurred in February 1-5 1999, when low PM levels at the rural sites were recorded (around 10µgPM10/m^3 at MON and 11µgTSP/m^3 at COR during the episode). Finally, air masses from Eastern Europe and Northern Italy may also be advected under dry weather conditions, such as an anticyclone over Central Europe (Figure 4-11C). In some of these events PM levels at rural sites often reached up to 20-25µg/m^3 of PM10 and TSP. Although the transport of particulate pollutants from mainland Europe cannot be denied during these episodes, a predominant transport of PM from the urbanised eastern coast of Spain is expected to contribute to PM10 levels at the rural sites because of the northeasterly winds.

4.2 Study of the African dust transport patterns by simulations in the SKIRON forecast system

The transport patterns of African dust to Eastern Spain were studied by simulations in the SKIRON forecast system following the methodology described in detail in the section 3.1.2 of chapter 3. Table 4-2 shows the location of the monitoring stations used for this study. In addition to the monitoring stations in Eastern Spain (in the autonomous regions of Catalonia, Valencia and Aragón), stations located in the Southern and Central areas of the Iberian peninsula were also used to support the interpretations. The African dust events in 2000 are not included in this study because the simulations were performed between October and December 1999.

4.2.1. African dust events and synoptic meteorological contexts

The African dust events in the study area occur when air masses over the North African desert move northward over the Iberian peninsula. This results in an increase in PM10 and TSP levels at the air quality stations due the high mineral load of the African air masses. Table 4-3 summarises the particulate events with a major African origin detected in the period 1996 – 1999. The interpretation of the meteorological mechanisms giving rise to each African intrusion is also included in this table. Figure 4-12 shows the daily PM10 and TSP levels recorded at the

Table 4-2. TSP and PM10 monitoring stations used in the present study for comparison with SKIRON simulations . See location in Figures 4-14 to 4-17

Station	Parameter	Location	Environment
MONAGREGA, MON	PM10	40.5ºN, 0.2ºW, 600 m.a.s.l.	Rural
CARBONERAS, CAR	PM10	37.0ºN, 1.9ºW, 11m.a.s.l.	Rural
CORATXAR, COR	TSP	40.4ºN, 0.0º, 1235m.a.s.l.	Rural
VILAFRANCA, VIL	TSP	39.7ºN, 0.1ºW, 1125m.a.s.l.	Rural
ESPIEL, ESP	TSP	38.0ºN, 5.0ºW, 520m.a.s.l.	Rural
SAN PABLO, SPA	TSP	39.2ºN, 4.2ºW, 917m.a.s.l.	Rural, EMEP
LOGROÑO, LOG	TSP	42.4ºN, 2.5ºW, 503m.a.s.l.	Rural, EMEP
CAMPISÁBALO, CAM	TSP	41.2ºN, 3.0ºW, 1405m.a.s.l.	Rural, EMEP
ROQUETAS, ROQ	TSP	40.5ºN, 0.0ºW, 46m.a.s.l.	Rural, EMEP
FORNELLS, FOR	TSP	41.6ºN, 2.6ºE, 101m.a.s.l.	Urban
SAGRERA, SAG	TSP	41.2ºN, 2.1ºE, 60m.a.s.l.	Urban, Barcelona city
L'HOSPITALET, L'HO	PM10	41.2ºN, 2.0ºE, 70m.a.s.l.	Urban, Barcelona city
ONDA, OND	TSP	39.8ºN, 0.1ºW, 193m.a.s.l.	Industrial
LINARES, LIN	PM10	38.0ºN, 3.0ºW, 280m.a.s.l.	Urban, Linares city
PALOS DE LA FRONTERA, PLF	PM10	37.1ºN, 6.5ºW, 24m.a.s.l.	Industrial
CONSTITUCIÓN, CON	PM10	37.1ºN, 3.4ºW, 688m.a.s.l.	Urban, Granada city
MADRID, MAD	PM10	40.2ºN, 3.4ºW, 650m.a.s.l.	Urban, Madrid city

rural stations of MONAGREGA (NE Spain, MON in Figure 4-17), CARBONERAS (SE Spain, CAR in Figure 4-17), and EMEP SAN PABLO station (SW Spain, SPA in Figure 4-17), where the African events are highlighted. For the sake of brevity, only some examples of African events will be discussed.

African air masses reach the study area when the synoptic situation is governed by depressions located to the West or Southwest of the Iberian peninsula (coast of Portugal or in the region between Cape Saint Vincent, the Straits of Gibraltar and the Canary Islands, Figure 4-13a) or when the North African anticyclone (usually placed between Algeria and Egypt at the 850hPa levels) shifts to the East or Southeast of the Iberian peninsula (Figure 4-13b). The combination of both cyclone and anticyclone systems has also been reported as a scenario favouring the dust transport from North Africa towards Spain (Figure 4-13c). Air back trajectories of these transport scenarios are shown in Figure 4-13d.

From January to June most of the African 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 4-12 and Figure 4-12) or between Cape Saint Vincent, 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 African 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 µgPM10/m^3 at MON and CAR stations on average for all the events, whereas maximum averaged levels reached 41 and 57 µgPM10/m^3,

Table 4-3. PM10 and TSP peak events induced by African 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 Cape Saint Vincent, the Canaries and Gibraltar (type L(C)), high pressure at the South or Southeast of the Iberian peninsula (H) or by the simultaneous presence of a Western or South-western depression and an Eastern anticyclone (type L+H).

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

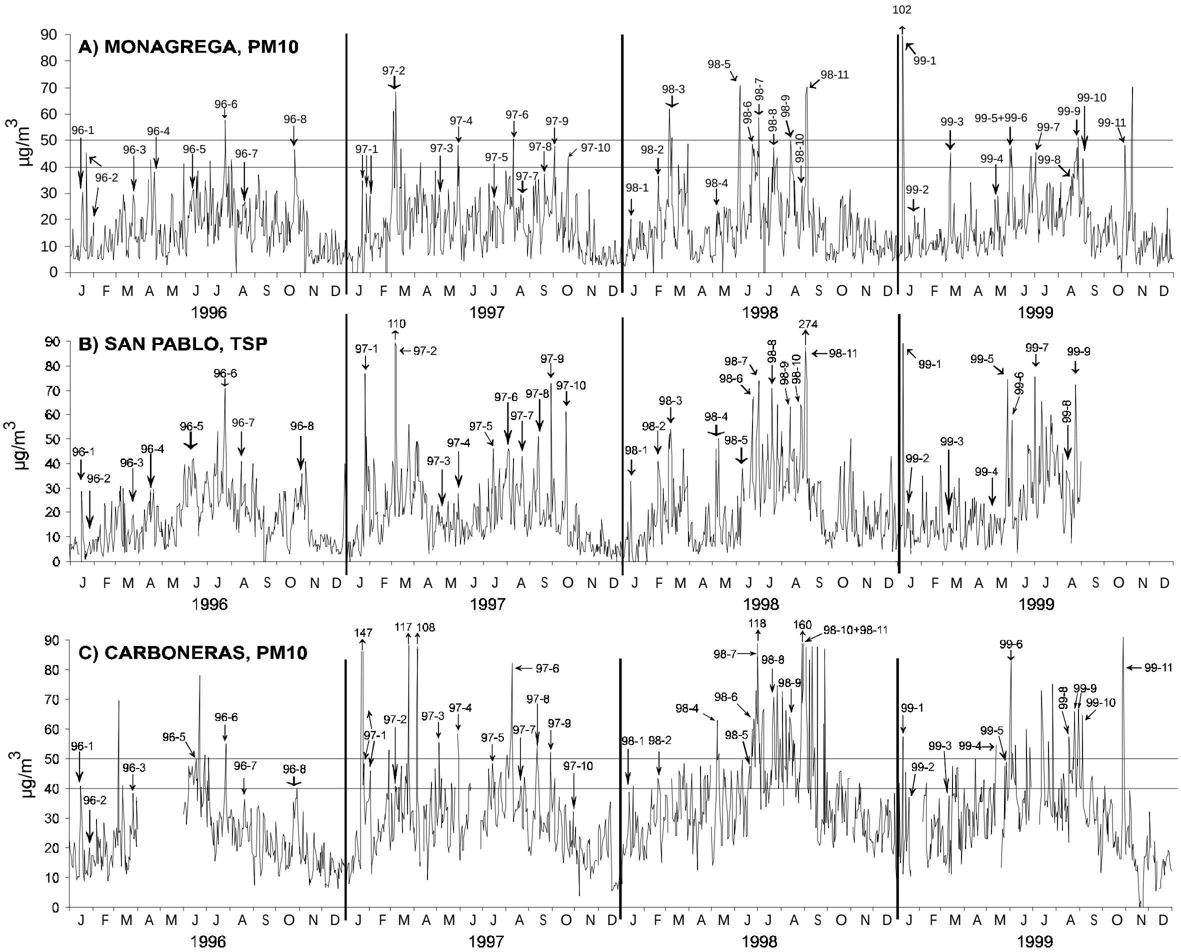


Figure 4-12. 1996 to 1999 daily PM10 or TSP levels from the rural stations of MONAGREGA and CARBONERAS and the EMEP SAN PABLO station. African dust events reported in Table 2 are highlighted in the particulate records.

respectively. Low PM10 concentrations before the African 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 African intrusion under these scenarios. Furthermore, the rapid decrease in particulate levels recorded at the end of the events are due to rainfall scavenging. This is the origin of the well known African red rains in Spain (Avila et al., 1997 and 1998).

The event of January 20 to 23 1997 is a typical case of African dust transport affecting Eastern Spain (Figure 4-14). On January 19 the depression located over the San 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 January 20 the strong pressure differences between the Atlantic and the Western Mediterranean resulted in a persistent northward flow until January 22 (Figure 4-13a). Thus, an African 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 South-western stations, outside of the influence of the plume.

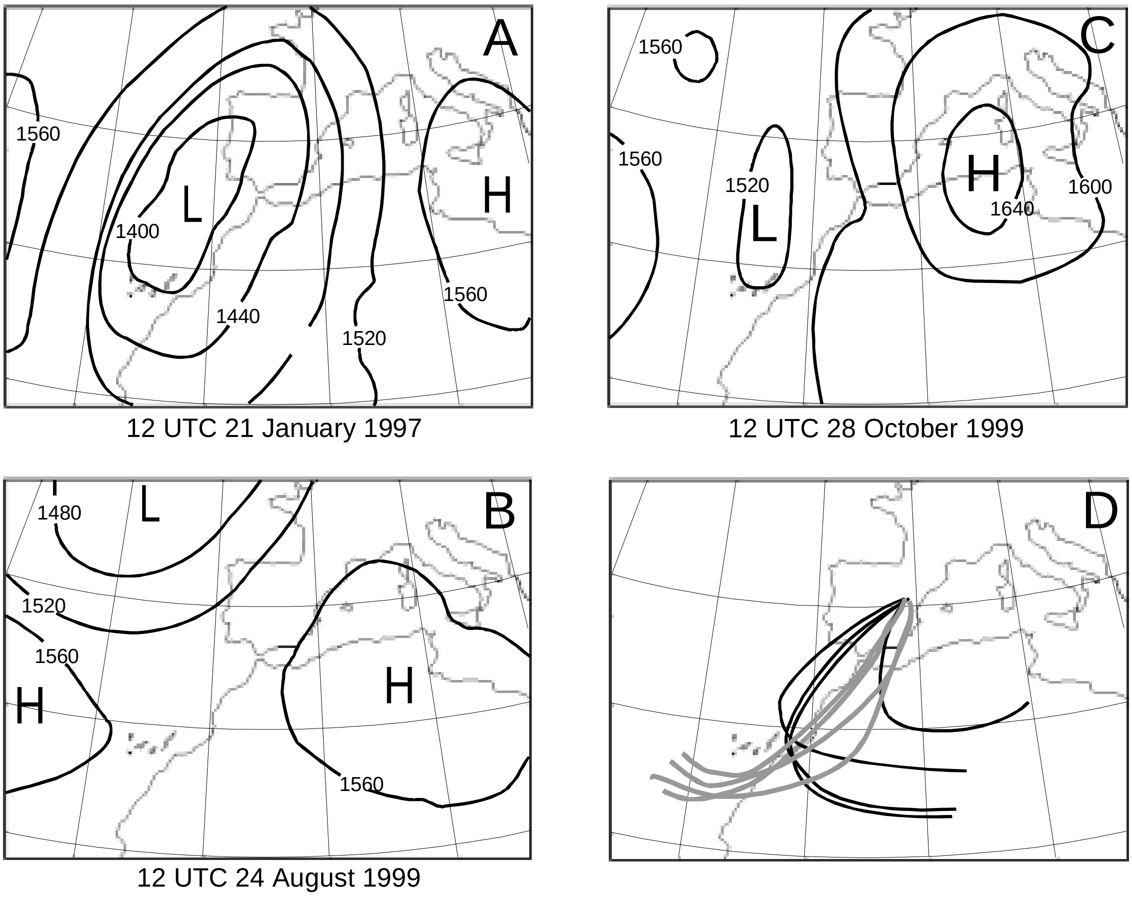
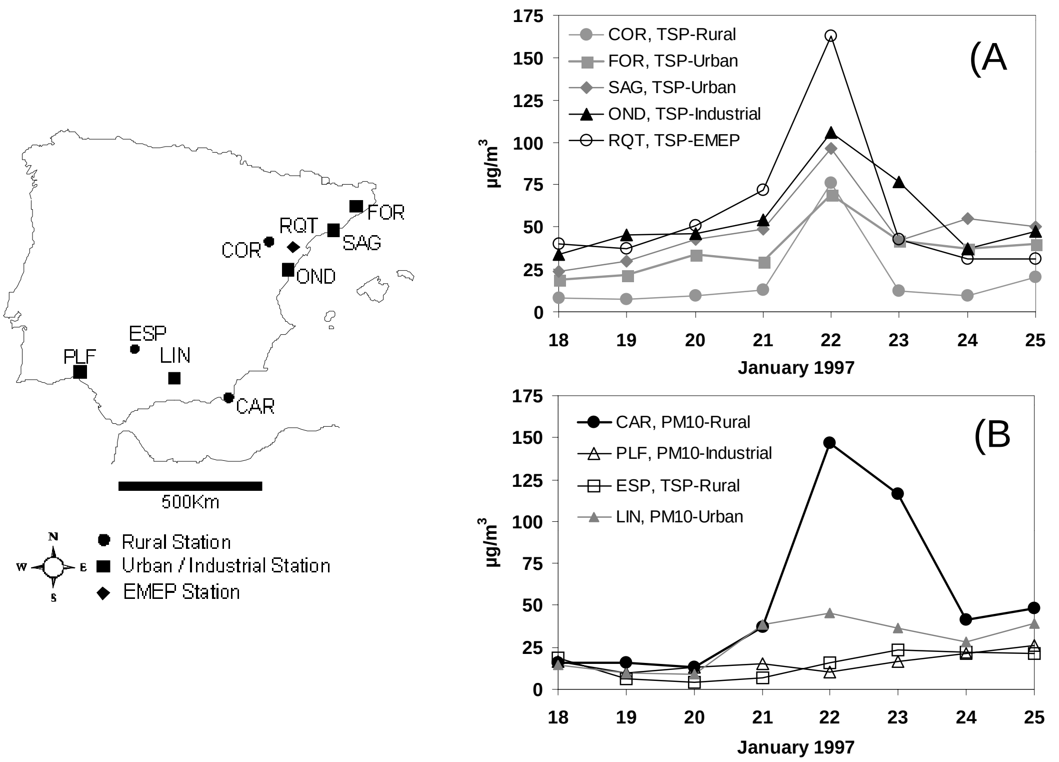
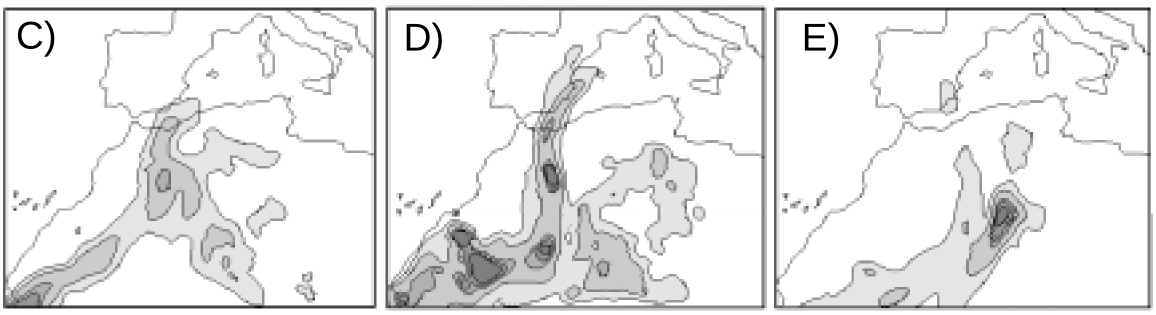


Figure 4-13. Synoptic pressure systems (850hPa) inducing the transport of African dust to the Iberian peninsula. A) Low pressures over West and/or Southwest of Portugal. B) High pressure over East or Southeast of the Iberian peninsula. C) The combination of both low and high pressure systems. D) Back trajectories to the MONAGREGA station induced by these synoptic systems: Bold lines for anticyclonic pathways (25-Jul-96, 27-Aug-98, 31-Aug-98 and 13-May-99) and grey lines for the cyclonic pathways (22-Jan-96, 10-Aug-97, 10-March-99, 27-Oct-99).

The subsequent particulate scavenging by rainfall resulted in a sharp reduction in the particulate levels on January 23 and 24 in North-eastern and South-eastern Spain, respectively. PM10 daily levels, which exceeded the EU daily limit value by a factor>2, were recorded at the CAR rural station during this event (147µgPM10/m^3 and 116µgPM10/m^3 on January 22 and 23 respectively).

Another interesting example showing the influence of the African plume on the daily PM10 and TSP levels occurred on June 4 and 5 1998 (Figure 4-15). 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 June 4 and 5. Daily levels of 49 and 71 µgPM10/m^3 were recorded at the rural MON station during this event. As described for the January 1997 event, a sharp decrease in particulate levels was induced by rainfalls on June 6 and 7 and by the eastward displacement of the African





00:00 UTC January 21 1997 | 18:00 UTC January 22 1997 | 06:00 UTC January 23 1997

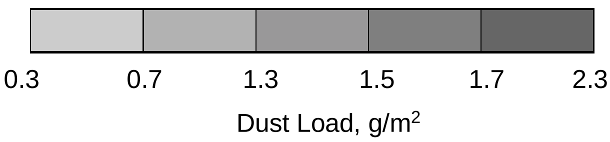


Figure 4-14. Daily PM10 and TSP levels recorded at selected monitoring stations during the African event from 22 to 23 January 1997 and evolution of the dust load (vertically integrated dust concentration in g/m^2) from the SKIRON system.

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 the land) and by the intensification of the North African anticyclone (in the 850hPa level; Font, 1956). The thermal convective activity over the African 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 South-westward as far as the Caribbean Islands (Prospero and Ness, 1986). The African 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 4-12). The strong convective activity under the Iberian thermal low conditions (Millán et al., 1997) lead to the abatement of the African air masses over the Iberian peninsula. Dayan and Miller (1989) demonstrated that the mixing of the lower troposphere levels with upper atmospheric masses from North Africa during these intrusions is enhanced in summer by the greater thickness of the mixing layer.

An example of high summer dust events occurred from August 30 to September 2, 1998 (Figure 4-16). Owing to the location of the North African high over Algeria (a synoptic scenario similar to that shown in Figure 4-13c), a dust intrusion approached Southeast Spain on August 30. Subsequently, it spread over South-western and North-eastern Spain on August 31 and September 1, respectively. Daily PM10 means registered at rural stations reached 150µg/m^3 in the South-eastern regions (CAR) and 70 µg/m^3 in the North-eastern regions (MON).

Figure 4-17 shows another example of a typical summer African dust event in August 1999 under a meteorological scenario similar to that in Figure 4-13B.

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 African 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 re-suspension 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.

By contrast, the summer dust events caused by South-western depressions or ending with an advection of Atlantic air masses present a sharp decrease in the particulate levels as in the case of the winter events.

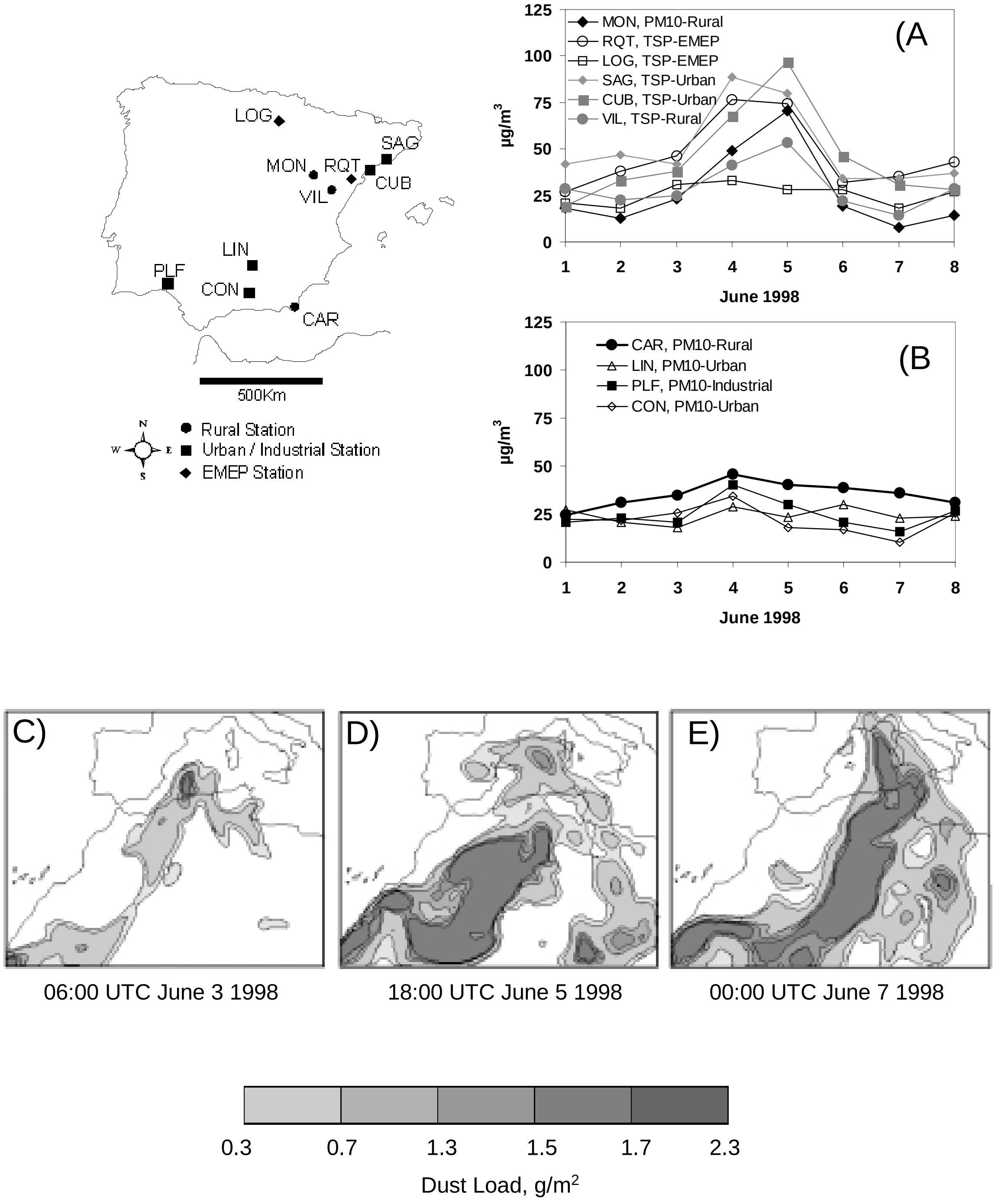


Figure 4-15. Daily PM10 and TSP levels recorded at selected monitoring stations during the African event from 4 to 5 June 1998 and evolution of the dust load (vertically integrated dust concentration in g/m^2) from the SKIRON system.

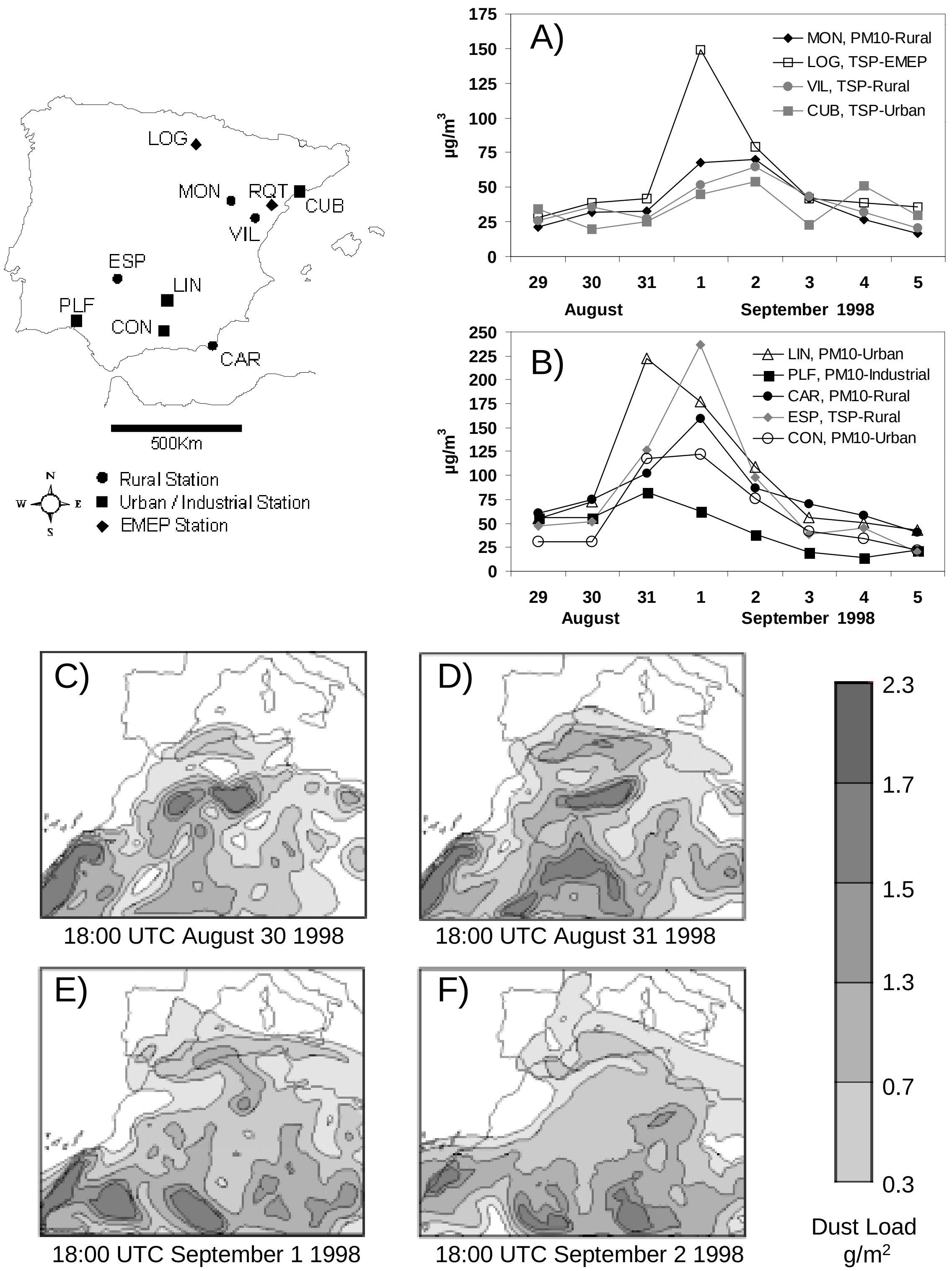


Figure 4-16. Daily PM10 and TSP levels recorded at selected monitoring stations during the African event from August 30 to September 2 1998 and evolution of the dust load (vertically integrated dust concentration in g/m^2) from the SKIRON system.

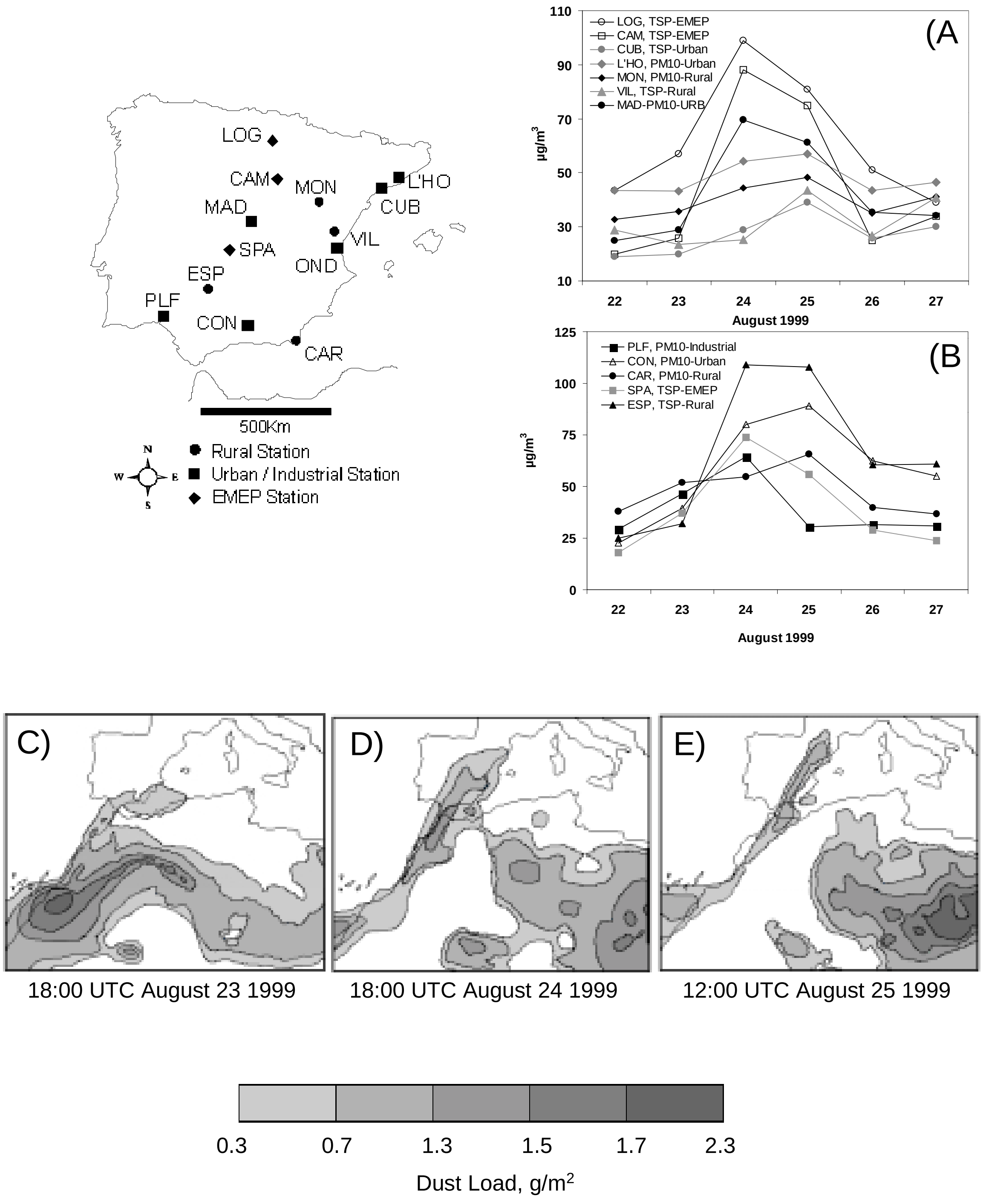


Figure 4-17. Daily PM10 and TSP levels recorded at selected monitoring stations during the African event from 23 to 28 August 1999 and evolution of the dust load (vertically integrated dust concentration in g/m^2) from the SKIRON model.

During September and October, African 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).

The African intrusions coupled with heavy rains are 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 and TSP time series. As for the seasonal distribution of the events having major impacts on the records of particulate levels, most of the African dust outbreaks (63%) reaching the Iberian peninsula occurred in the period May to 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 4-3), with a mean duration of 3 to 4 days. Consequently, the number of days per year affected by particulate matter input from Africa ranges from 38 to 53 days for the study period (Table 4-3).

Finally, links between the African dust transport patterns to Eastern Spain and the African dust source regions should be taken into account. In winter and spring, African dust is mostly produced in the Sahel (or sub-Saharan region; between the 10ºN and 20ºN latitudes) region, whereas in summer the Saharan region (between the 15ºN and 25ºN latitudes) is the most important source of African dust (Swap et al., 1996; Herman et al., 1997). This northward displacement in the source region of dust from winter to summer is associated with a displacement in the intertropical convergence zone (Dubief, 1979). Moreover, the hottest and driest North African region is the Sahel in winter and spring and the Sahara region in summer. Although only a few examples are discussed here, this description agrees with the results of this study. In the African dust outbreaks in January 1997 (Figure 4-17) and March 1997, 1998 and 2000 (Figure 4-10), the plume of dust provides evidence of long range transport from the Sahel, whereas in the African events in August 1997 (Figure 4-9), 1998 (Figure 4-16) and 1999 (Figure 4-17) the dust outbreaks over the Iberian peninsula are transported from the Saharan region.

4.2.2 Comparison with other measurements in the Mediterranean basin

The magnitude of the daily PM10 and TSP levels recorded for the African 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 98µgTSP/m^3 in Sardinia in March 1991 during African events, whereas Northern and Western airflows gave rise to TSP concentrations between 11 and 32µg/m^3. In August 1984, Lefèvre et al. (1986) in Sicily recorded average daily TSP levels of 92µg/m^3 during an African event and 33µg/m^3 for dominant Northern flows. Correggiari et al. (1989) measured 72µgTSP/m^3 aboard ship South of 40ºN latitude during an African episode on February 1983. Chester et al. (1984) recorded 100µgTSP/m^3 in the Central Tyrrehenian in October 1979, whereas they measured 11µgTSP/m^3 under Eastern flow conditions. Guerzoni et al. (1989) recorded 5µgTSP/m^3 in the southern Thyrrhenian aboard ship under North-western airflows.

4.3 Study of the summer Regional episodes

A number of studies have reported that in rural environments in the Mediterranean airborne particulate concentrations undergo a seasonal cycle characterised by a summer maximum (Bergametti et al., 1989; Kubilay and Sadam, 1995; Querol et al., 1998a, 1998b). To the best of our knowledge, this seasonal cycle has not been reported at rural sites in Central and Northern Europe (e.g. Monn et al., 1995; Turnbull and Harrison, 2000; Röösli et al., 2001). This section is focused on the influence of the typical summer meteorology on the levels of PM10 (particulate matter ≤10µm) and TSP (total suspended particles) in rural areas in Eastern Spain. To this end, the temporal variations in PM10 and TSP concentrations at rural sites were studied by means of meteorological analysis and TOMS-spectrometer satellite observations for detecting African dust outbreaks. Given that preliminary results based on interpretations of 1996-2000 PM10 and TSP time series showed that every summer the PM10 and TSP concentrations are influenced by the same meteorological processes, only data from the summer 2000 are presented. Special attention is paid to these summer Regional episodes given that they induce high PM background levels at rural sites.

4.3.1 PM levels at rural sites in summer

This study is based on PM (particulate matter: PM10 and TSP) measurements performed at rural stations in Eastern Spain. This region is characterised by an abrupt orography (Figure 4-18), constituted by the Iberian range (NW-SE) and the Catalan coastal range (NE-SW). The latter is crossed by deep valleys that reach the flat coastal area. Situated between these two ranges is the Ebro basin (Figure 4-18), which is characterised by a semi-arid soil. The ranges are mainly covered by typical Mediterranean and coniferous forests. The main urban and industrial settlements in Eastern Spain are located along the coastal plain (Figure 4-18).

Levels of PM10, TSP and gaseous pollutants (NOx, SO_2 and O_3) are continuously measured at the rural stations belonging to the Valencia Autonomous Government and ENDESA (Empresa Nacional de Electricidad, S.A.) air quality networks. Levels of TSP are determined by means of automatic beta radiation attenuation monitors (Dasibi), whereas levels of PM10 are measured using automatic TEOM (Rupprecht and Patashnick) or GRIMM laser spectrometer monitors depending on the station (Table 4-4). Gaseous pollutants are monitored by standard methods. The rural stations (Table 4-4) are located in different micro-geographical settings in the Ebro basin (MONAGREGA), the top of the range close to the coast (VILAFRANCA, MORELLA and CORATXAR) and the bottom of a valley (SORITA). It should also be pointed out

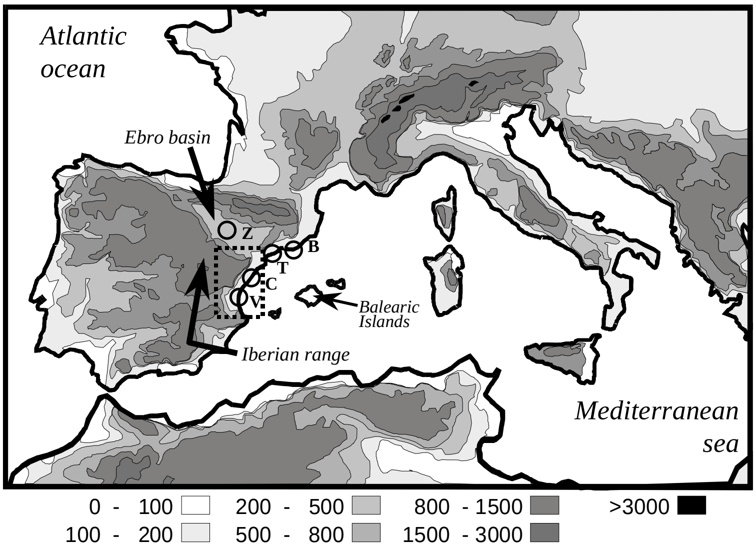


Figure 4-18. Topography of the Western Mediterranean (in meters above sea level). The dotted square highlights the region where the rural monitoring stations referred to in Table 1 are located. The locations of the main urban and industrial settlements in Eastern Spain are also indicated. B: Barcelona, T: Tarragona, C: Castelló, V: Valencia, Z: Zaragoza.

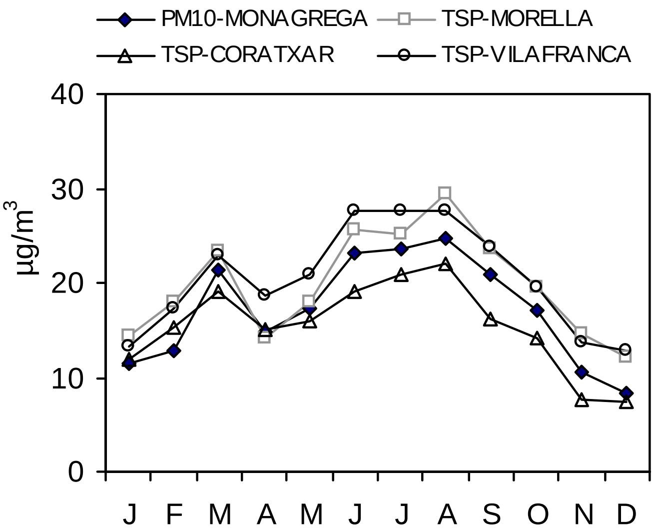


Figure 4-19. Monthly mean concentrations (1996-2000) of PM10 and TSP at rural sites in Eastern Spain.

Table 4-4. TSP and PM10 rural monitoring stations. See location in Figure 4-18. AM = annual mean in 2000; %R= annual percentage of available data in 2000.

Station	PM	Equip.	AM	%R	Location	Altitude	Observations
MONAGREGA	PM10	TEOM	17µg/m^3	99%	40.5ºN,0.2ºW	600m.a.s.l.	Flat Ebro basin
SORITA	PM10	GRIMM	12µg/m^3	77%	40.7ºN, 0.2ºW	640m.a.s.l.	Into a valley
MORELLA	TSP	Beta	21µg/m^3	92%	40.6ºN, 0.1ºW	1153m.a.s.l.	Top of the range
CORATXAR	TSP	Beta	15µg/m^3	92%	40.4ºN,0.0º	1235m.a.s.l	Top of the range
VILAFRANCA	TSP	Beta	20µg/m^3	98%	39.7ºN,0.1ºW	1125m.a.s.l	Into a valley

that some of these sites are located at a high altitude (>1000 m.a.s.l.), and most of them can be considered as representing the regional background environment.

In the study area the seasonal evolution of PM is characterised by the typical summer maximum of the rural Mediterranean environments (Figure 4-19). Figure 4-20 shows the daily PM concentrations from early summer to early autumn 2000 (July to October). Note the high degree of correlation between the PM concentrations recorded at the different rural sites. Moreover, the range of PM10 and TSP concentrations is very narrow (Figure 4-20). Consequently, these summer TSP concentrations are considered to be representative of the regional PM10 levels in the rural environment in Eastern Spain. These regional PM10 concentrations measured at rural sites in 2000 (12-17µg/m^3 PM10 annual mean, Table 4-4) are relatively high when compared with the 2010 EU limit values for PM10 (annual mean ≤20µg/m^3 and do not exceed the daily concentrations of 50µg/m^3 on more than 7 days/year, EU Directive 1999/30/CE). Note that in the period from July to September, daily PM10 concentrations exceeding 40µg/m^3 and 50µg/m^3 were recorded for 11 and 3 days, respectively, at the MONAGREGA rural site.

The above seasonal pattern (Figure 4-19) contrasts with the seasonal evolution of PM reported for rural sites in Central and Northern Europe. In the Swiss Alps, Monn et al. (1995) reported an autumn-winter maximum (weekly PM levels from 20 to 60µg/m^3 in autumn-winter and from 10 to 30µg/m^3 in summer). In the study area, during the year 2000 the weekly PM levels ranged from 5 to 15µg/m^3 in autumn and winter (March is not included) and from 20 to 40 µg/m^3 in summer months and March. At rural sites in Switzerland, the monthly PM10 concentrations only exceeded the level of 20µg/m^3 in November (Röösli et al., 2001) during the period April 1997- May 1998. In contrast, in Eastern Spain the PM10 monthly concentration of 20µg/m^3 is exceeded throughout the summer (Figure 4-19). Thus, this level was surpassed during 4 months in 2000 at the MONAGREGA rural site. Turnbull and Harrison (2000) did not report a significant PM10 seasonal cycle at rural sites in the United Kingdom.

4.3.2 PM levels and synoptic meteorological scenarios

The different periods of high and low daily PM concentrations detected from early summer to early autumn 2000 were classified as a function of the synoptic meteorological scenarios (Figure 4-20 and Table 4-5) and origin of the air masses.

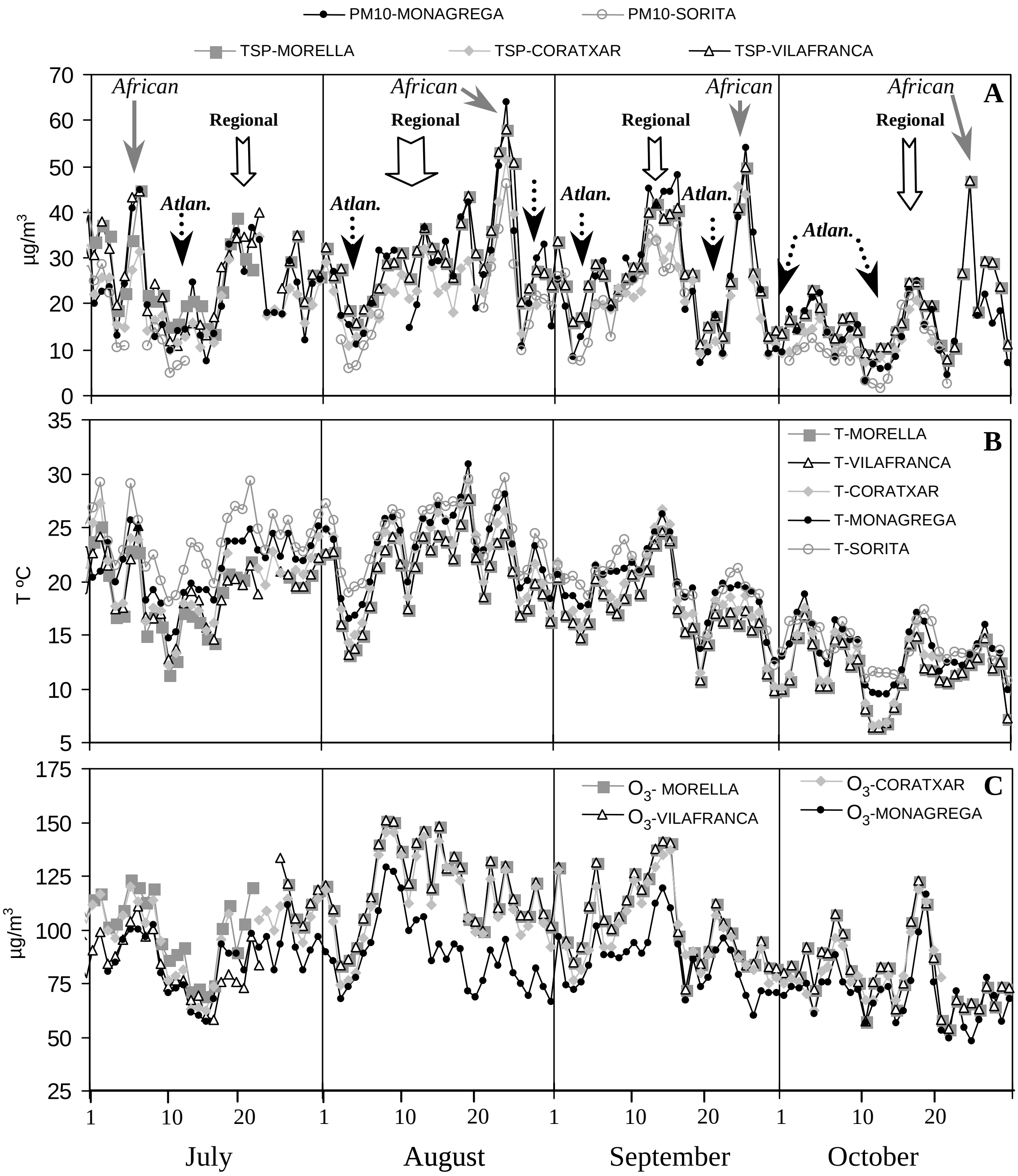


Figure 4-20. Daily mean values of (A) PM10 and TSP concentrations, (B) temperature and (C) O_3 concentrations in selected rural stations from July to October 2000. The arrows highlight the periods in which the main African (grey arrow), Regional (white arrow) and Atlantic (black dotted arrow) episodes occurred

Table 4-5. PM episodes recorded in summer 2000. Meteorological pressure patterns, local wind direction, air mass origin deduced from the back trajectory analysis, and mean PM10 (in μg/m^3) concentrations at the MONAGREGA station during the episodes (ND: No data). Pressure patterns: L indicates a depression located over the Atlantic at mid-latitudes L(A), in front of Portugal L(P), or over the Straits of Gibraltar L(G). H indicates anticyclone over North Africa H(NAF), the Atlantic ocean H(A), or the Atlantic and Western Europe H(A+WE).ITL: Iberian Thermal Low, WGC: Weak gradients conditions, IB: Iberian peninsula, WM: Western Mediterranean. Local winds (direction) - S: south, SE: Southeast, N: North, NW: Northwest. WE: Western Europe.

Dates	***Pressure pattern***	***Local Wind***	***Air mass origin***	**PM10**
July 1-3	WGC(WM)+ITL	Breeze	*Regional* (SE-IB)	22
July 4-5	L(A)	S-advection	*W-Atlantic*	19
July 6-7	H(NAF)	S-advection	*Africa*	43
July 8-17	H(A)+L(A)	NW-advection	*NW-Atlantic*	15
July 18-23	WGC(WM)+ITL+H(A+WE)	Breeze / SE	*Regional* (WM)	31
July 24 – Aug 1	WGC(WM)+ITL+H(A+WE)/L(A)	Breeze / NW	*Regional / Atlantic*	24
Aug. 2-7	H(A+WE)	N/NW-advection	*NW-Atlantic*	20
Aug. 8-23	WGC(WM)+ITL+H(A+WE)	Breeze/SE	*Regional* (WM+SE-IB)	28
Aug. 24-26	L(P)+H(NAF)	S-advection	*Africa*	50
Aug. 27 –Sept. 1	H(A+WE) / L(A)	NW/S-advection	*NW / W-Atlantic*	22
Sept. 2- 10	H(A+WE) / L(A)	NW/N-advection	*NW-Atlantic +WE*	22
Sept. 12-17	WGC+ ITL	Breeze	*Regional* (WM)	42
Sept. 18-24	L(A)	S-advection	*W / SW-Atlantic*	16
Sept. 25-27	H(NAF)	S-advection	*Africa*	43
Sept. 28-30	L(A)	NW-advection	*NW-Atlantic*	14
Oct. 1-6	H(A+WE)+L(A)	NW-advection	*NW-Atlantic*	19
Oct. 7-16	H(A) / L(A)	NW-advection	*NW-Atlantic*	10
Oct. 17-20	WGC+ H(IB)	Breeze / SE	*Regional* (WM / W-IB)	21
Oct. 21-24	L(G)	S-advection	*Africa + intensive rains*	11
Oct. 25-26	L(G)	S- advection	*Africa*	ND
Oct. 26-30	H(A+WE)	N / S	*Regional*	14

July-2000. Weak gradient conditions over the Western Mediterranean and the Iberian thermal low (ITL) development constituted the prevailing scenario from June 26 to July 3 (Figure 4-22A). On July 4 an Atlantic cold front crossed Eastern Spain, causing a drop in temperature (T) and in the PM and O_3 concentrations. On July 6 and 7 a peak in the PM concentrations was reported. The TOMS satellite observations showed a plume of African dust (Figure 4-21) over Eastern Spain and the Western Mediterranean. Subsequently, abrupt entries of Atlantic air masses in the Western Mediterranean basin persisted up to July 17 (Figure 4-22B), leading to a fall in T and in the PM and O_3 concentrations. Owing to the fact that several cold fronts reached the Western Mediterranean during this period (on July 11, 15 and 16), the temperature decreased reaching the minimum values for this month (~12ºC). The ozone and PM concentrations also fell reaching the minimum values for this month: O_3<60μg/m^3 and PM<15μg/m^3. These are very low PM and O_3 (Millán et al., 2000) levels when compared with those that are typical of this region in summer (Figure 4-20). From July 18 to 23 the "typical summer" scenario prevailed. This was characterised (Figure 4-22C) by weak gradient conditions

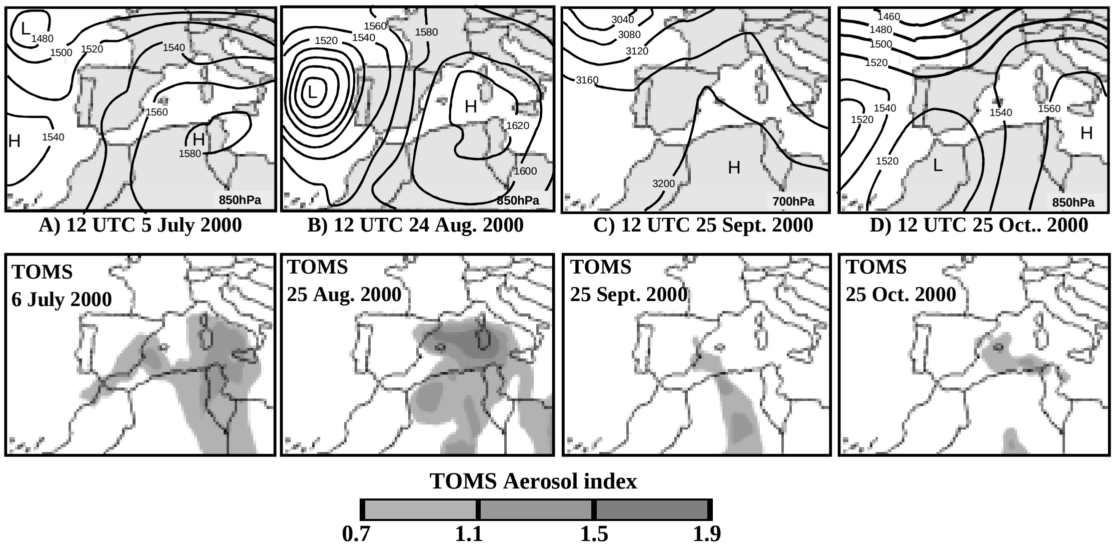


Figure 4-21. Above: TOMS UV-radiation absorbing aerosol index maps during the African dust outbreaks between July and October 2000. Below: synoptic charts (altitude in meters above sea level of the pressure level of 850hPa, or 700hPa for the event at the end of September) showing the meteorological patterns causing the transport of African dust.

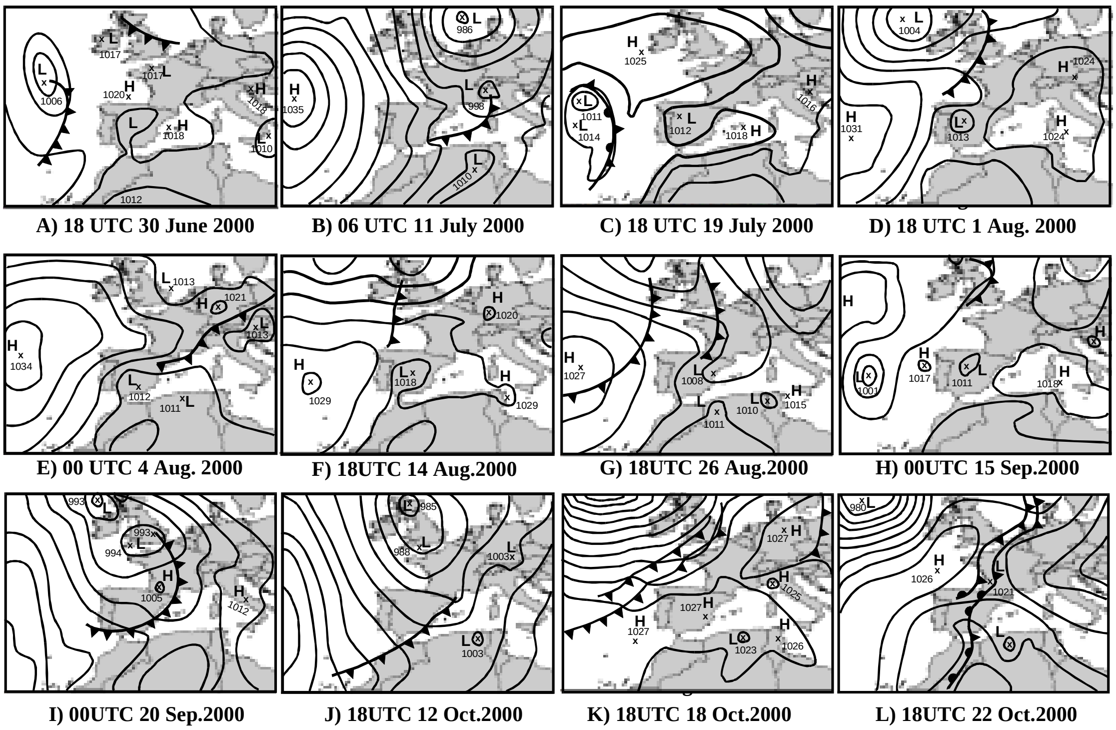


Figure 4-22. UK Meteorological Office synoptic charts of pressure (mb) at sea level.

over the Western Mediterranean, the ITL and the North Atlantic anticyclone affecting northwest Europe. In this period an increase in T and in the PM and O_3 concentrations was again reported. These weak gradient conditions over the Western Mediterranean favour the intense breeze circulation (Gangoiti et al., 2001). In a 3 day period (July 17 to 19) the PM and O_3 concentrations increased from 14 to 32µg/m^3 and from 69 to 91µg/m^3, respectively (as daily values at the rural monitoring stations). From July 24 to August 1 the typical summer scenario (Figure 4-22D) alternated with smooth advections of Atlantic air masses brought about by the displacement of an Atlantic depression towards the Iberian peninsula.

August-2000. From August 2 to 7 cool Atlantic air masses entered the Western Mediterranean basin via the Gulf of Lions (Figure 4-22E) and reached Eastern Spain, diminishing the T and the PM and O_3 concentrations down to values <15ºC for T, <15µg/m^3 for PM and <80µg/m^3 for O_3 at most sites. From August 8 to 23 the summer meteorological scenario favouring the breeze circulation was again dominant (Figure 4-22F), with an increase in the T and the PM and O_3 levels. During this period the PM and O_3 concentrations ranged between 20 and 40µg/m^3 and between 100 and 150µg/m^3, respectively. These ozone levels are in the range of the high ozone episodes typical of this region in summer (Millán et al., 2000). From August 24 to 26 an African dust outbreak over the Western Mediterranean and Eastern Spain (Figure 4-21) resulted in a peak PM event which coincided with a decrease in the O_3 levels. The PM10 concentrations during the African event exceeded the forthcoming EU PM10 daily limit value of 50µg/m^3 at the MONAGREGA station. Subsequent abrupt entries of Atlantic air masses (Figure 4-22G) brought about a marked decrease in the PM10 levels from 64 to 11µg/m^3 at the MONAGREGA station in a 3 day period (from August 25 to 27). A similar decrease was reported at the other stations (Figure 4-20).

September-2000. From September 2 to 10 several Atlantic cold fronts crossed Eastern Spain giving rise to low PM and O_3 concentrations. From September 12 to 17 a high PM, O_3 and T episode was reported under conditions favouring the breeze circulation: weak gradient conditions over the Iberian peninsula and Western Mediterranean together with high pressures over the Mediterranean. Even the ITL was developed on some days (Figure 4-22H). During this event high PM and O_3 concentrations were again reported in the range between 25 and 45µg/m^3 for PM and between 100 and 150µg/m^3 for O_3. In the September 18-24 period, Atlantic cold fronts reached the Western Mediterranean (Figure 4-22I), with a fall in the T and the PM and O_3 concentrations. Subsequently, an African event induced a PM peak on September 25-27 (Figure 4-21). Owing to the high mineral dust load, the daily PM10 levels at the MONAGREGA station reached 54µg/m^3, exceeding the forthcoming EU limit value for PM10 (50µg/m^3). Low O_3 concentrations were reported owing to the influence of the relatively "unpolluted" African air masses. Subsequent Atlantic episodes again diminished the PM levels.

October-2000. In the October 1-16 period, several Atlantic episodes caused frequent low PM and O_3 events (Figure 4-22J). From October 17 to 20 weak gradient conditions and a slight anticyclonic situation prevailed (Figure 4-22K), and a high T, PM and O_3 episode was again reported. From October 21 to 24 intensive rains depleted the PM concentrations. An African dust event (Figure 4-21) on October 25 and 26 produced a PM peak. On October 31 a cold front reached the Western Mediterranean (Figure 4-22L) and led to a decrease in the T, PM and O_3 concentrations.

The highest PM events (daily concentrations between 40 and 60µg/m^3 in the July – October 2000 period) were reported during the African dust episodes in late August, late September and late October (Figures 4-20 and 4-21). High PM and low ozone concentrations are recorded during the African episodes as observed in studies carried out in Atlantic regions (Savoie et al., 1992; Rodríguez and Guerra, 2001).

The second highest PM events (daily concentrations between 20 and 45µg/m^3 in the July – October 2000 period) are recorded during periods characterised by a weak pressure horizontal gradient over the Western Mediterranean, which is often associated with the ITL development. As previously reported (section 4.1.2), advection of air masses is not significant under this scenario, and the scarce air mass renovation caused by the breeze circulation and the development of meso-scale meteorological processes accounts for the ageing of air masses and the accumulation of particulate pollutants in the regional atmosphere. More details on the features of these Regional PM episodes associated with increases in the temperature and in the ozone levels (Figure 4-20) are provided in the following section.

The lowest PM episodes are recorded during periods characterised by intense advections of Atlantic air masses toward the Western Mediterranean. These events occur in some meteorological scenarios, such as Atlantic depressions developing off the western or northern coasts of the Iberian peninsula (Figure 4-22I), the North Atlantic anticyclone bringing strong northern winds over Spain (Figure 4-22B), or the North Atlantic anticyclone inducing entries of Atlantic air masses via the Gulf of Lions (Figure 4-22E). Most of these Atlantic events are associated with rainfall and with a cold front crossing the Iberian peninsula. Since the Western Mediterranean is surrounded by high coastal ranges (Figure 4-18), these Atlantic events represent entries of cool air which "clean" the Mediterranean atmosphere, leading to a decrease in the temperature and PM and ozone concentrations (Figure 4-20 and Table 4-5). In the period July – October 2000 the lowest PM events were recorded during intense Atlantic episodes, e.g. daily PM concentrations <20 µg/m^3 in the periods 9-18 July, 3-6 August or 4-5 September (Figure 4-20).

4.3.3 Local meteorological features during the Regional episodes

This section is focused on the study of the special features of the Regional PM episodes given that they account for the second highest PM events at the rural sites in summer.

Wind direction and velocity did not undergo daily cycles during periods dominated by synoptic scale meteorological processes. As expected, southern winds prevailed throughout the day during advection of African dusty air masses. Northern winds prevailed throughout the day during the intense Atlantic episodes caused by the North Atlantic anticyclone. However, where these are caused by Atlantic depressions, the prevalence of northern or southern winds depends on the latitude of the depression.

In periods of weak gradient conditions over the Western Mediterranean, often associated with the ITL, the wind speed shows marked daily cycles with maximum values during daylight owing to the activation of up-slope winds over the mountain slopes (mountain breeze) and the inland sea breeze. In most of these events, the wind direction is characterised by SE winds during daylight and NW winds during the night, but in some periods of intense ITL development light SE winds may blow even during the night. In this typical summer scenario, the back-trajectory analysis shows that the air masses entering the mainland from the Mediterranean sea during daylight have a regional origin. These back-trajectories show a short route (Figure 4-23D, 4-24C, 4-25D) owing to the weak gradient conditions over the Western Mediterranean. An anticyclonic curvature caused by the compensatory subsidence (which typically occurs between Eastern Spain and the Balearic Islands, Figure 4-18) associated with the thermal buoyancy over the warmer terrain is also observed in the trajectories (examples in Figures 4-22A and 4-22C). Examples of these types of back-trajectories are shown in Figures 4-23D, 4-24C and 4-25D, discussed below. Both the breeze circulation and the lack of significant advection of air masses during these events account for the regional origin of the airborne particulates.

Figure 4-23 shows the hourly values of wind velocity and direction recorded at the MORELLA station for the Regional PM episode in mid September 2000 (Figure 4-20 and Table 4-5). From September 7 to 10 the wind direction and speed did not show significant variations during the day (except on September 8), and the trajectory analysis showed transport patterns from the North Atlantic and Western Europe (Figure 4-23C). From September 13 to 17 weak gradient conditions prevailed over the Iberian peninsula, and even the ITL was developed for some days (Figure 4-22H), and an increase in the PM concentrations was reported (Figures 4-20 and 4-23). During this period the wind speed and direction showed marked daily cycles due to the up-slope winds and the sea breeze circulation. Moreover, the trajectory analysis showed that the air masses entering the mainland during daylight had a regional origin. The scarce renovation of the air masses due to the development of the breeze circulation and the lack of significant advection of air masses accounts for the accumulation of airborne particulates in the regional atmosphere. From September 18 to 20 a significant fall in the PM concentrations at the

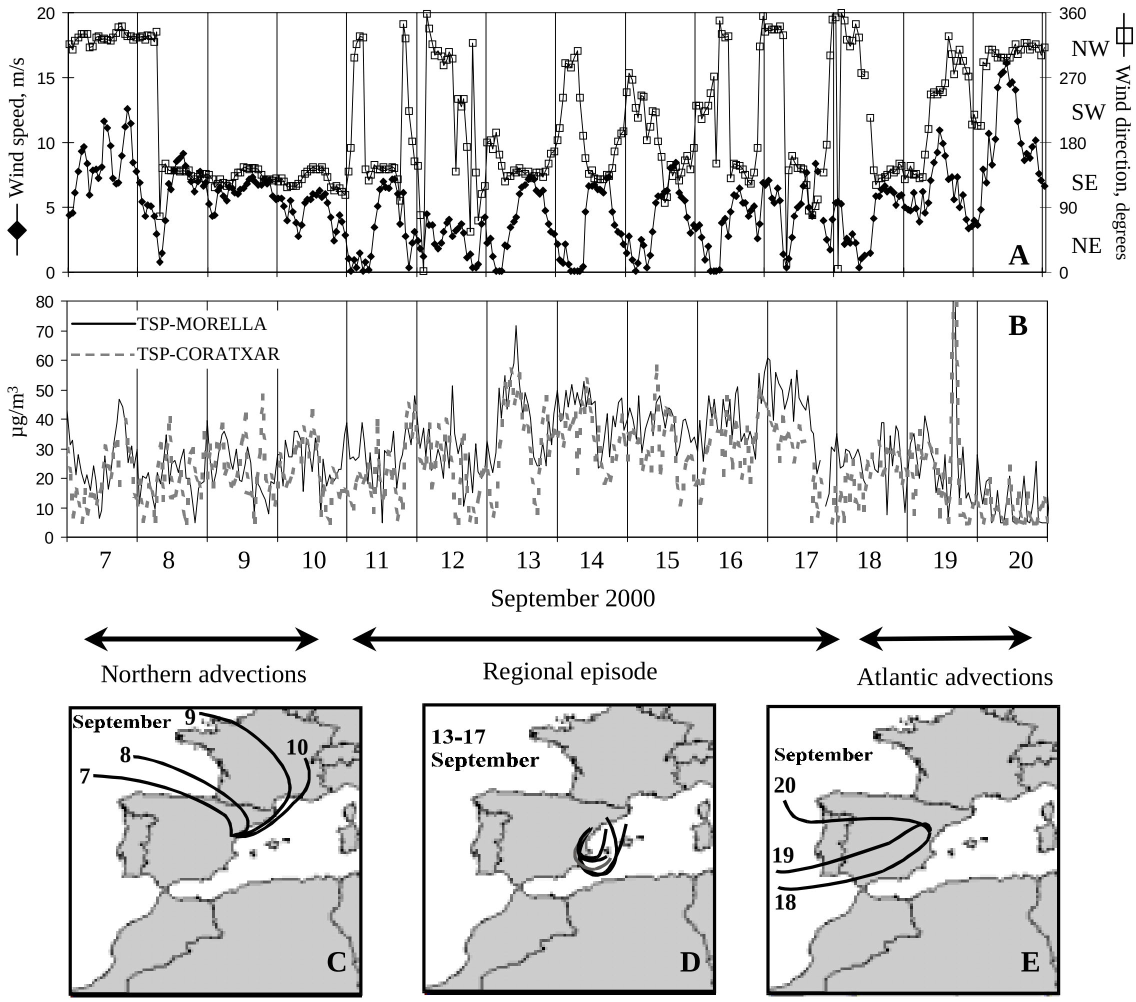


Figure 4-23. Hourly values of: (A) wind velocity and direction at MORELLA and (B) TSP at MORELLA and CORATXAR. Isentropic air back-trajectories (calculated at 12:00 UTC and 3 days backward for 1000m.s.a.l.) in the period 7-20 September 2000 (C, D and E).

rural sites was brought about by the entry of cool Atlantic air masses into the Western Mediterranean basin (Figures 4-22I, 4-20 and 4-23).

These Regional PM episodes were also reported in July (18-23; Figures 4-20, 4-22C and 4-24) and August (8-23; Figures 4-20, 4-22F and 4-25). Figure 4-25 shows hourly data of wind speed and direction and TSP concentrations at the MORELLA mountain site from August 4 to 28. From August 4 to 7 advections of Atlantic air masses produced strong northern winds resulting in low PM concentrations (Figures 4-22E and 4-25C). From August 8 to 23 weak gradient conditions over the Western Mediterranean and the ITL prevailed (Figure 4-22F), resulting in a PM event of regional origin (Figures 4-20 and 4-25D). Southern winds associated with breeze circulation prevailed in this period and back-trajectories showed the anticyclonic curvature over the Western Mediterranean and Eastern Spain. From early August 24 to early 26 southern winds prevailed during the day, and hourly PM concentrations >100µg/m^3 were reported owing to an African dust outbreak (Figure 4-21 and 4-25E). From mid August 26, an

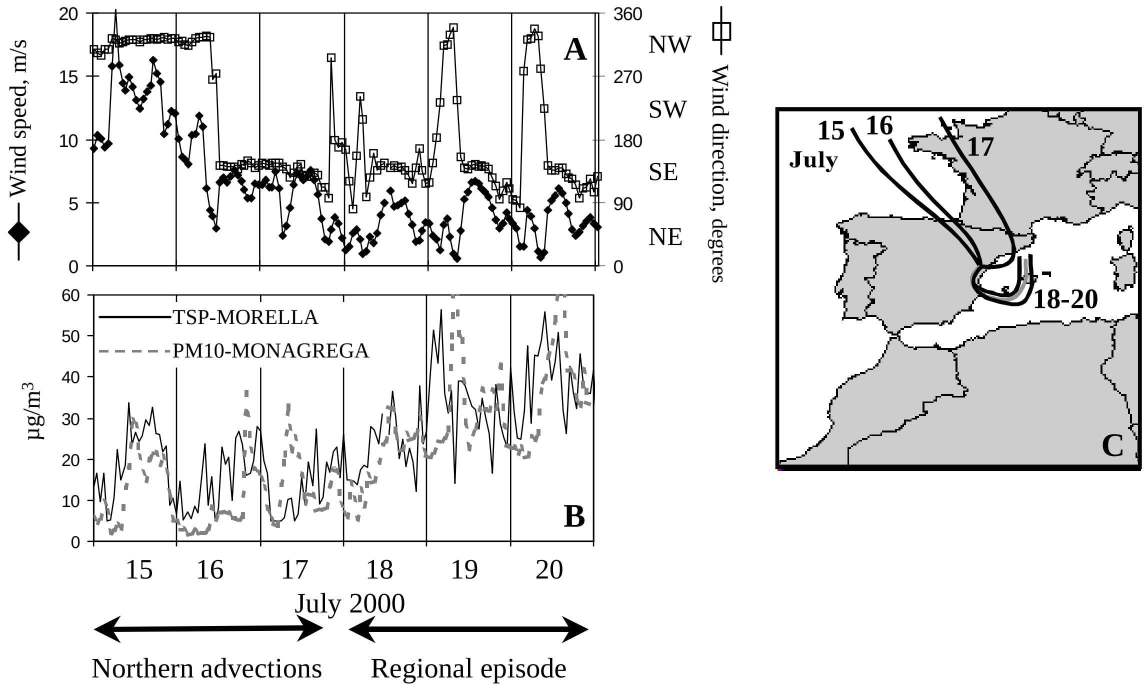


Figure 4-24. (A) Hourly values of wind velocity and direction at MORELLA. (B) Hourly values of TSP and PM10 at MORELLA and MONAGREGA. (C) Isentropic air back-trajectories (calculated at 12:00 UTC and 3 days backward for 1000m.s.a.l.) in the period 15-20 July 2000.

abrupt entry of cool northern Atlantic air masses in the Mediterranean basin led to reductions in the PM levels (Figures 4-22G and 4-25E).

In an attempt to elucidate the origin of the high ozone episodes in this region, Millán et al. (1997, 2000) proposed a conceptual model in which the polluted air masses undergo vertical re-circulations over the Eastern coast of Spain owing to the inland up-slope winds, the westerly winds over the top of the mountains, the subsidence over the Mediterranean sea and the subsequent inland entry of the sea breeze. In this scenario, Millán et al. (1997) propose that polluted air mass trajectories tend to describe an helicoidal curve along the eastern coast of Spain in a southward direction. The subsidence (relative high pressures) over the Western Mediterranean sea is attributed to the compensatory sinking associated with the thermal buoyancy over the warmer land together with the large scale anticyclonic subsidence. This subsidence accounts for the back-trajectories showing the anticyclonic curvature described above for the Regional PM episodes (e.g. Figures 4-23D, 4-24C and 4-25D). Salvador et al. (1999) have argued that given the abrupt orography of the region and given that the movement of air masses is influenced by the heating of mountain and valley slopes, meteorological models able to simulate the atmospheric dynamics need a very high resolution, at least 2 km x 2 km to account for 95% of terrain variance. Using high resolution trajectory analysis, Gangoiti et al. (2001) have demonstrated the re-circulations of polluted air masses over the Western Mediterranean and the Eastern coast of Spain. Moreover, back-scattering Lidar measurements have documented the transport of aerosols towards a high altitude by up-

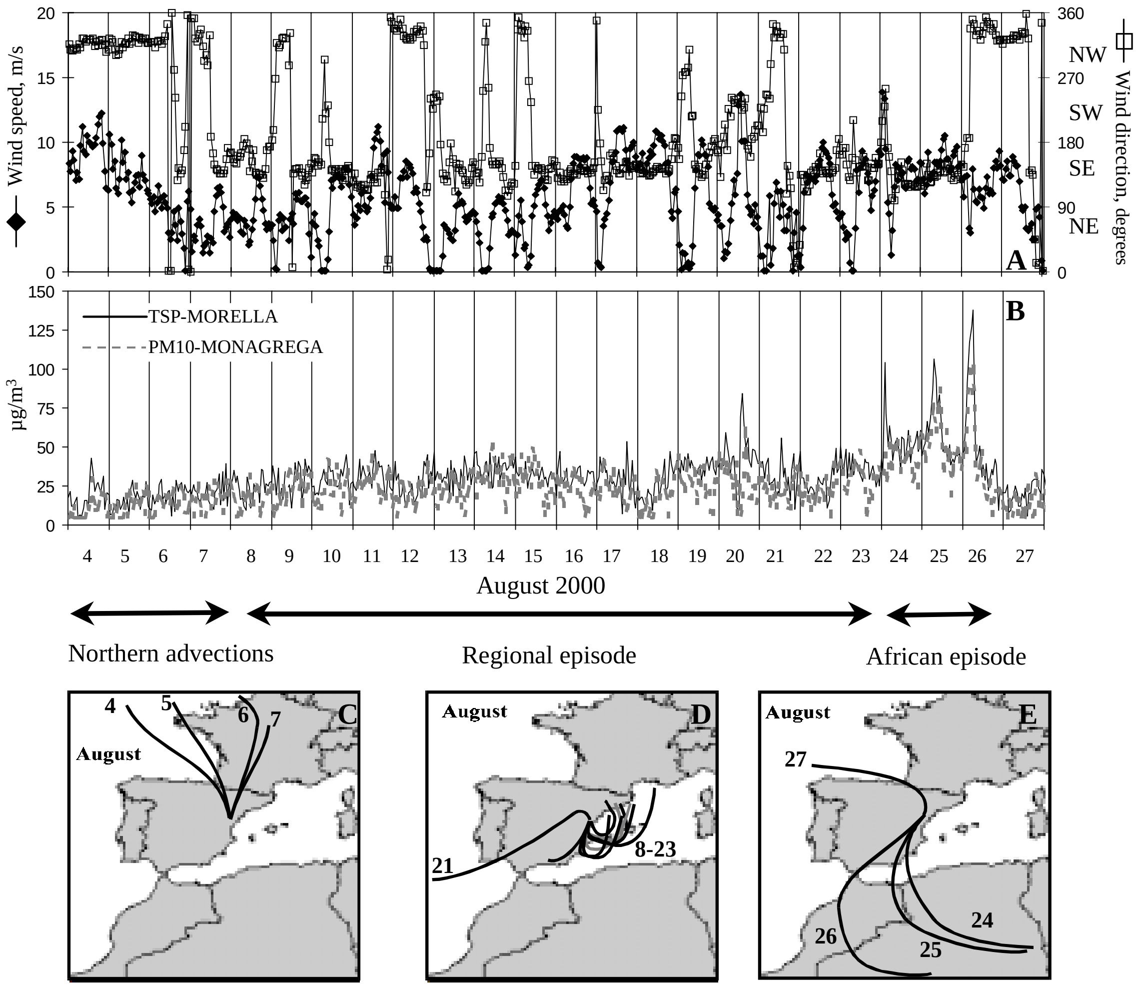


Figure 4-25. (A) Hourly values of wind velocity and direction at MORELLA; (B) hourly levels of TSP at MORELLA and of PM10 at MONAGREGA. (C, D and E) Isentropic air back-trajectories (calculated at 12:00 UTC and 3 days backward for 1000m.s.a.l.) in the period 8-27 August 2000.

slope winds on the coastal ranges in Eastern Spain (Soriano et al., 2001). Thus, although the back-trajectories associated with the Regional PM episodes (Figures 4-23D, 4-24C and 4-25D) probably do not represent the actual and detailed movement of the air masses, they may be used as tracers of the regional air mass and particulate sources.

Although these Regional PM episodes are more frequently observed and have a longer duration in summer, they are also observed in the semi-warm periods (late spring and early autumn). A typical example, which is also associated with an ozone episode, occurred in the period October 17-20 2000 (Figure 4-20 and Table 4-5). This event was brought about by an anticyclonic situation together with a weak pressure horizontal gradient over the Iberian peninsula and the Western Mediterranean (Figure 4-22K). The temperature increase (Figure 4-20) was not as high as during the summer event, and consequently the ITL was not present.

The PM events attributed to regional sources are associated with ozone episodes. When interpreting the occurrence of the simultaneous high PM and O_3 events both the role of meteorology and that of photo-chemistry should be taken into account. From the point of view of meteorology, the increase in the PM and O_3 concentrations during the Regional episodes and the decrease during the Atlantic events account for the correlated daily O_3 and PM time series. In the photo-chemical context, the formation of secondary particles by photochemical processes is expected to be significant in these aged polluted air masses transported to the rural environment. Under strong summer insolation, photochemical reactions involving NOx, organic vapours and SO_2 give rise to acid compounds, secondary particles, O_3 and other oxidants Moreover, O_3 is involved directly (oxidation of gases by O_3) and/or indirectly (e.g. oxidation of gases by the OH radicals formed by O_3 photolysis) in the formation of secondary particles (e.g. sulphate, nitrate, organic components).

The crustal particulate matter is also a key component of PM10 in these regional episodes. This is due to the fact that the temperature increases favour the turbulent dynamics in arid areas. Querol et al. (1998b) found a higher load of crustal PM in summer than in winter in the Ebro basin.

4.4 Seasonal patterns of PM

Table 4-6 shows the monthly mean number of days (from 1996 to 2000) with trajectories from each sector considered in this study (Figure 4-3). The number of trajectories per month from 1996 to 2000 from each sector is shown in Figure 4-26. The annual frequency of each transport pattern was: 55% Atlantic, 20% Europe, 13% North Africa and 12% Regional. The Regional transport pattern is more frequently recorded in summer (Figure 4-26A). In the summer months, the Regional transport pattern may account for 10-15 of the days of the month (Figure 4-26A). Atlantic events are frequent in the cold season, mainly from November to February, and also in April (Figure 4-26B).

Figure 4-27 shows the monthly mean concentrations of PM measured at rural (Figure 4-27A), urban and industrial sites (Figure 4-27B), the difference between the monthly PM levels measured at the urban/industrial and rural sites (Figure 4-27C), and the monthly CO and NO_x concentrations averaged from a set of urban/industrial stations (Figure 4-27D). At rural sites (COR, MON, MOR and VIL) the highest monthly PM levels are recorded in summer, but a second order maximum is evident in March in some years. Conversely, the lowest PM levels are measured in winter. Most of the sub-urban and urban sites (FOR, CUB, PEN and L´HO) and the OND industrial site also recorded this seasonal PM trend, but the winter minimum is less pronounced. At the highly polluted sites, the highest PM levels are measured in autumn-winter (IGU, MAR and G.VIA) or constantly along the year (e.g. GAN, SAO and ERM).

As Figure 4-27C shows, the difference between the monthly PM levels measured at the urban and rural sites and the CO and NO_x levels recorded at urban sites attain an autumn-winter maximum due to the higher frequency of Local urban pollution episodes. As previously

Table 4-6. monthly mean number of days (from 1996 to 2000) with trajectories from each sector considered in this study (Figure 4-3).

	Source region					
	ANW	**AW**	**ASW**	**NAF**	**RE**	**EU**
JANUARY	5	6	5	7	2	6
FEBRUARY	12	3	2	3	2	7
MARCH	10	3	3	5	3	7
APRIL	10	7	3	1	4	6
MAY	3	6	8	6	2	6
JUNE	6	3	7	4	4	6
JULY	9	4	2	4	9	4
AUGUST	5	2	5	6	9	4
SEPTEMBER	6	5	3	3	4	8
OCTOBER	10	5	4	4	3	5
NOVEMBER	10	9	2	1	1	6
DECEMBER	8	7	6	2	3	5
ANNUAL	**91**	**59**	**49**	**46**	**45**	**73**

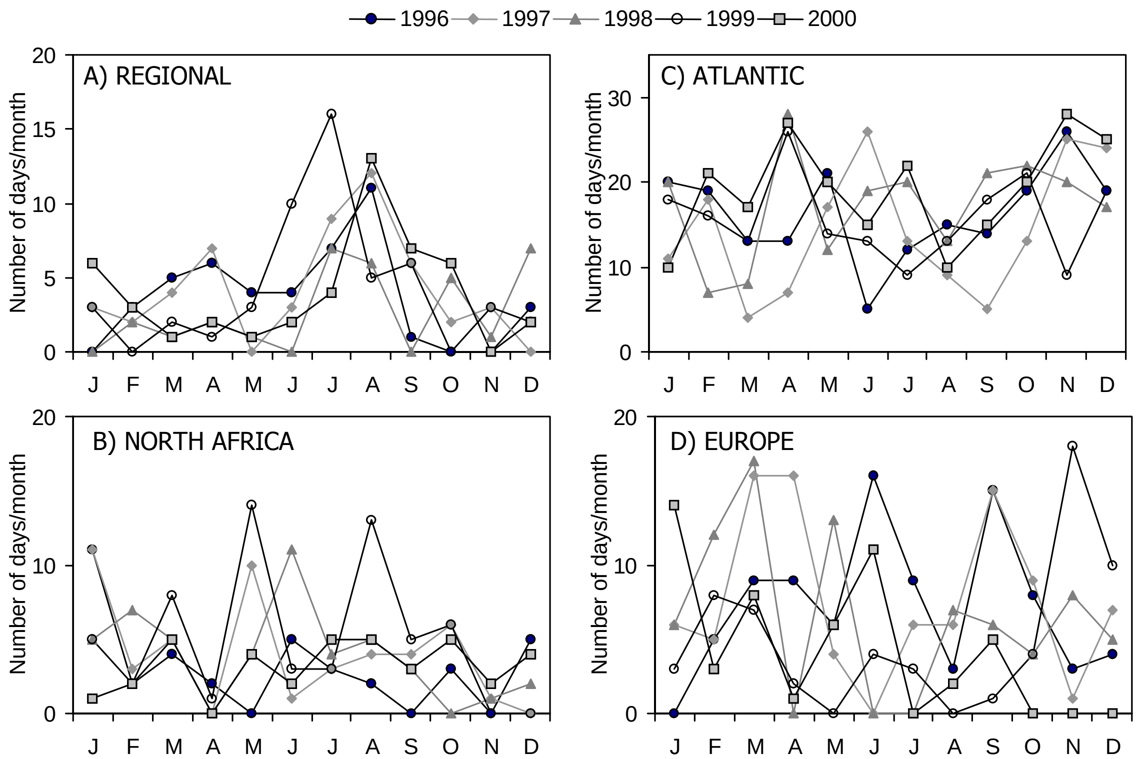


Figure 4-26. Monthly number of days of air back-trajectories from the different sources areas of air masses considered in this study (Figure 4-3).

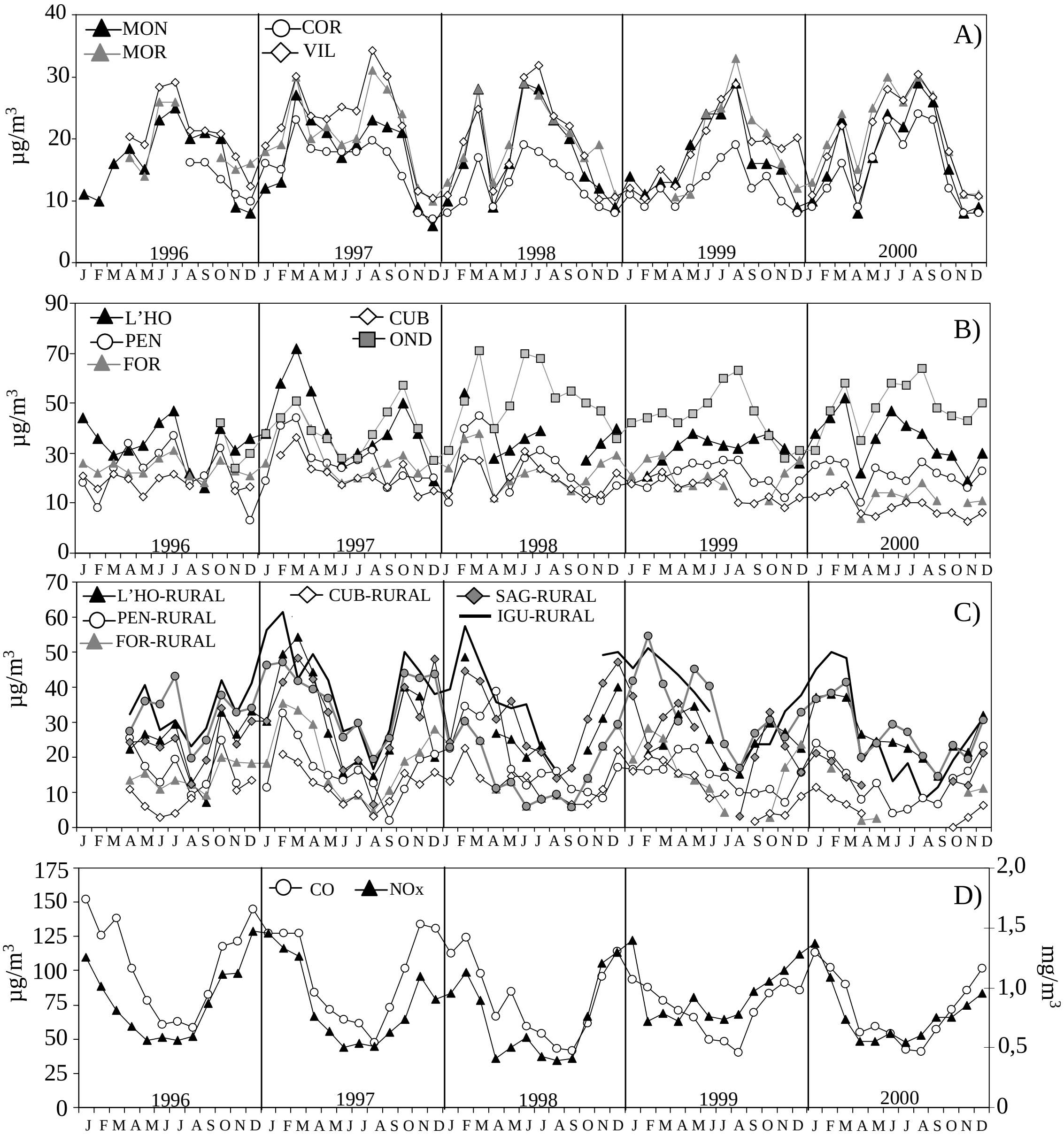


Figure 4-27. Monthly levels of PM10 (MON for 1996-2000 and SAG, L´HO and S.AND for 2000) and TSP at rural (A) and urban (B) sites. Difference between the monthly PM levels at urban/industrial and rural sites (C). Monthly levels of NOx and CO averaged for selected urban/industrial sites (SAG, L´HO, IGU, FOR, MAR, MOT, OND; D).

Table 4-7. Number (N) of exceedances of the EU daily limit values for PM10 at rural and urban stations in Eastern Spain, showing the number of exceedances caused by African dust and non-African dust events.

N>50µgPM10/m^3		AFRICAN	NON AFRICAN
CORATXAR	Rural	2	0
MONAGREGA	Rural	4	0
MORELLA	Rural	5	0
FORNELLS	Sub-urban	2	4
PENYETA	Sub-urban	8	6
L´HOSPITALET	Urban	13	38
IGUALADA	Urban / Industrial	13	38
ONDA	Industrial	23	50

stated in the period of March to October, the occurrence of Regional episodes predominates over the Local urban/industrial pollution episodes. The sporadic occurrence of African dust outbreaks (mainly from January to October) cause high PM episodes and could also influence the PM seasonal evolution. Thus, the higher summer frequency of Regional episodes and the occurrence of intense African dust events account for the maximum PM levels recorded at rural sites in the warm season (the monthly number of trajectories from each sector is included in appendix 3).

PM levels at rural sites in June and July 1997 were lower than those recorded for these months in other years (Figure 4-27). These anomalous low summer PM levels in 1997 were induced by the occurrence of intensive Atlantic episodes, associated with Atlantic depressions, cold fronts and rain in June and July 1997. The summer 1997 (mainly June and July) was classified by the Spanish National Institute of Meteorology as cool and wet (Calendario Meteorológico, 1998). The frequent occurrence of these Atlantic episodes inhibited the development of Regional PM events, and low PM levels were recorded.

In addition to the summer PM maximum recorded at the rural sites, in 1997, 1998 and 2000, a second order maximum was also evident in March. This secondary PM maximum was caused by heavy outbreaks of African dust over the Iberian peninsula in the periods March 3-7 1997, March 3-7 1998 and March 6-8, 14-15 and 18-19 2000 (previously discussed in section 4.1.4 and shown in Figure 4-10). These African events accounted for 16% of the days in March 1997 and 1998, and for 24% of the days in March 2000. Thus, the high PM levels recorded at the rural sites during these African episodes (up to 70 µgPM10/m^3 as a daily mean) account for the increase in the monthly PM concentrations.

4.5 PM levels and EU standards

Table 4-7 reports the 1996-2000 mean annual exceedances of the daily EU PM10 limit value caused by African-dust and non-African episodes at nine rural, urban and industrial monitoring stations (presented for each year in appendix 4). These estimations are based on direct PM10 measurements at MON and on TSP measurements at MOR and COR. In the two

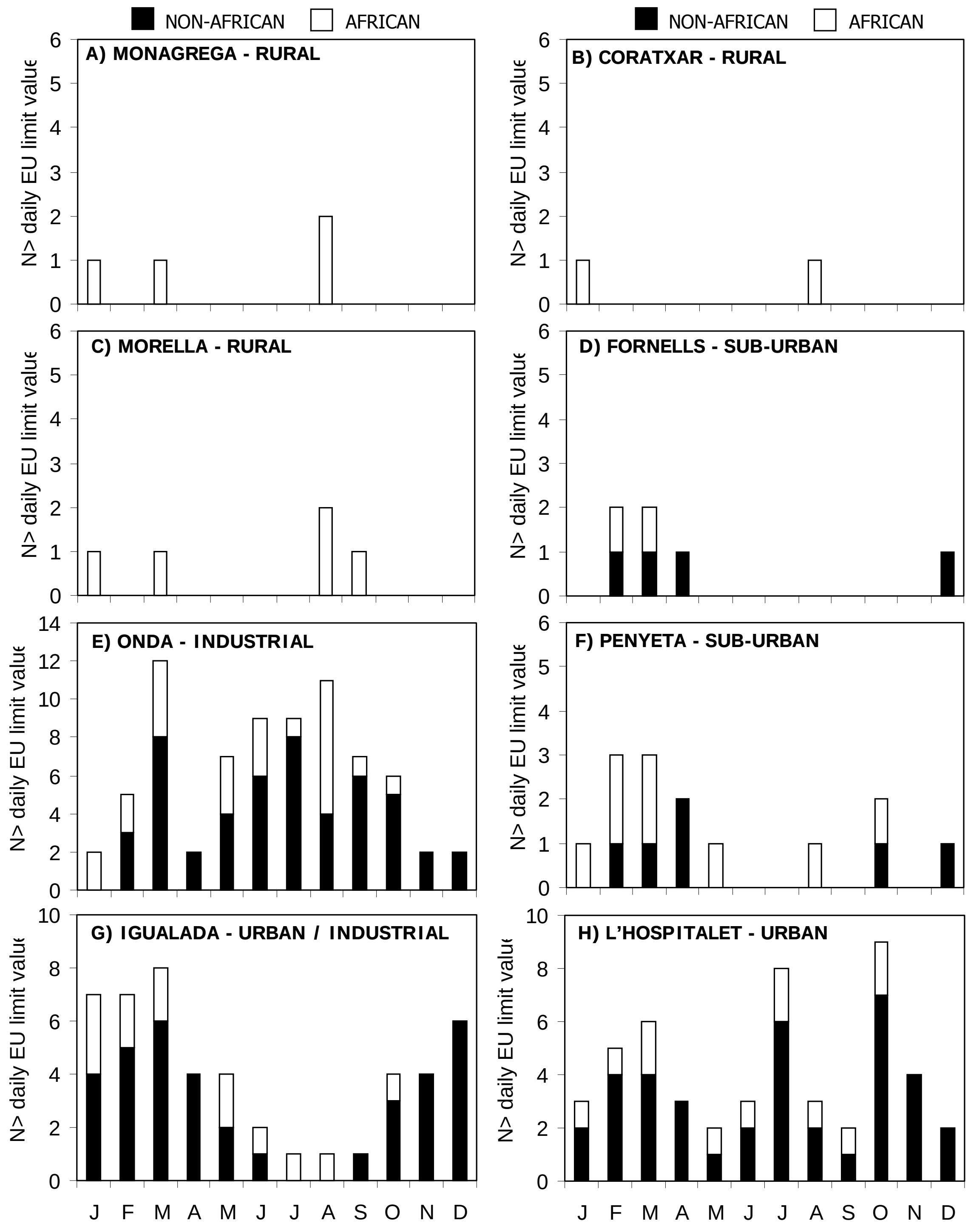


Figure 4-28. Averaged monthly exceedances (from 1996 to 2000) of the daily EU PM10 limit value caused by African-dust and non-African episodes at eight selected air quality monitoring stations from Eastern Spain.

latter rural sites it was assumed that PM10/TSP≈1 since a significant contribution to TSP levels of particles >10µm is not expected since these sites are located in ranges covered by forest. At the sub-urban, urban and industrial sites, these estimations (Table 4-7) are based on TSP measurements and on the experimentally determined ratio PM10/TSP=0.7 in Eastern Spain (discussed in chapter 6), with the exception of L'HOSPITALET in 2000 since PM10 was directly measured.

At the rural sites (MON, COR and MOR) there is a strong predominance of African over non-African (anthropogenic) exceedances. The African and non-African exceedances are balanced at the sub-urban sites (FOR and PEN), whereas anthropogenic exceedances prevail at the urban and industrial sites (SAG, L'HO, IGU and OND).

The number of exceedances of the daily PM10 limit value (50µg/m^3) induced by African events at the rural (n=2-5, mainly recorded in January, March and August, Figure 4-28) and at the sub-urban (n=2-8, mainly recorded from January to October, Figure 4-28) sites is very close to that permitted for 2010 (n=7). The number of African induced exceedances of the daily PM10 limit value at the urban (n=13) and industrial (n=23) sites (mainly recorded from January to October) is higher than that permitted for 2010.

At the sub-urban sites, the number of non-African exceedances (n=4-6) of the daily PM10 limit value is much lower than that permitted for 2005 (n=35), and slightly lower than that permitted for 2010 (n=7). At the urban sites, the number of exceedances of the daily PM10 limit value (n=38) is slightly higher than that permitted for 2005 (n=35) and much higher than that permitted for 2001 (n=7). The selected industrial site would not meet the requirements for 2005 and 2010 with respect to the exceedances of the daily limit value.

In 2000, when direct PM10 measurements were performed, 110 non-African exceedances were recorded at L'HOSPITALET (appendix 4), which is a number significantly higher than that recorded in the prior years (around 40 per year). In the period June 1999 to June 2000, when 115 samples of PM10 were taken (data discussed in chapter 5) with the EN 12341 reference method demanded by the EU directive 1999/30/CE, 86 exceedances of the EU daily limit value were recorded. In 2001, 74 non-African exceedances were recorded at SAGRERA (Viana, personal communication, 2002). This underestimation in the number of exceedances of the daily EU limit value is attributed to the underestimation in TSP levels in autumn-winter due to the use of beta monitors (discussed in chapter 3). Thus, around 80 non-African exceedances per year are estimated for the urban kerbsides stations in Eastern Spain.

The non-African exceedances were mostly recorded in autumn and winter at the urban sites because of the higher frequency of Local pollution episodes (Figure 4-28). The maximum number of exceedances was recorded at the OND industrial site during summer (Figure 4-28) owing to the scarce renovation of polluted air masses induced by the sea breeze circulation (the local atmospheric dynamic in this area is discussed in detail in chapter 6).

Based on PM10 measurements performed with the EN 12341 reference method (chapter 5), the annual PM10 concentrations are estimated in the ranges 17-20µg/m^3 at the rural, 30-45µg/m^3 at the urban and 45-60µg/m^3 at the industrial sites. The EU annual PM10 limit value for 2005 (40µg/m^3) would be met at the rural sites, but not at most urban traffic stations or industrial sites. At the rural sites, annual PM10 levels are slightly lower than the annual EU limit value for 2010. The urban traffic and industrial sites would not meet the annual EU limit value for 2010.

The annual TSP concentrations reached maximum levels in 1997 and 1998 at 12 of the 19 studied monitoring stations (appendix 1). These inter-annual differences are higher at the urban and industrial sites than at the rural ones.

4.6 Comparison between the PM10 measurements performed with the TEOM and the EN12341 reference method: reconstruction of winter PM10 levels

As discussed in chapter 3, the PM10 and TSP measurements interpreted in this chapter were performed with PM monitors based on tapered element oscillating micro-balance (TEOM) and on the attenuation of beta radiation. Earlier studies demonstrated that these automatic PM monitors underestimate PM10 levels when compared with the EN 12341 reference method demanded by the EU Directive 1999/30/CE (Allen et al., 1997; APEG, 1999; European Commission, 2001) owing to the volatilisation of semi-volatile species during the collection. This is due to the heating of the sampled air to remove particle-bound water. In this study simultaneous PM10 measurements using TEOM and gravimetric (which meet the EN12341 requirements) methods were performed at the MONAGREGA rural site (at the rate of two samples per week) from March 1999 to July 2000. A comparison of the results obtained with these systems was performed in order to quantify the potential degree of underestimation.

As Figure 4-29A shows, PM10 measurements obtained with TEOM fit very well those performed with the gravimetric method in warm and semi-warm periods (March to October). However, in the cold period of the year (November to February) TEOM underestimates PM10 levels in 32% with respect to the gravimetric method (Figure 4-29 B). This underestimation of PM10 levels is caused by the occurrence of ammonium-nitrate in winter (demonstrated in chapter 5) which is volatilised during the heating of the sample.

This underestimation of PM10 levels in winter should be taken into account in the interpretation of PM10 data. Figure 4-30A shows the daily PM10 levels recorded with the TEOM and the gravimetric methods at MONAGREGA. In summer, PM10 presents the aforementioned high background concentrations, and PM10 levels measured with both systems (TEOM and beta) in the range 20-50µg/m^3. However, low background PM10 concentrations were recorded in winter. In the period November-February, PM10 levels were mostly in the interval 5-20µg/m^3 according to TEOM measurements and in the range 5-35µg/m^3 according to the gravimetric measurements. The monthly mean PM10 concentrations obtained with TEOM are very close to those obtained with the gravimetric system from March to October, but a mean underestimation of 32% is observed in the period from November to January (Figure 4-30B).

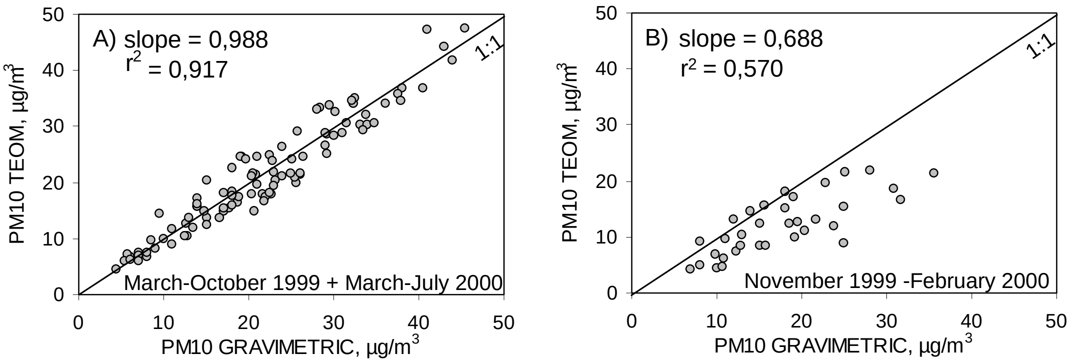


Figure 4-29. Levels of PM10 measured with TEOM versus those obtained with the gravimetric method for the periods March-October 1999 + March July 2000 (A) and November 1999 – February 2000 (B). Measurements performed at the MONAGREGA rural site.

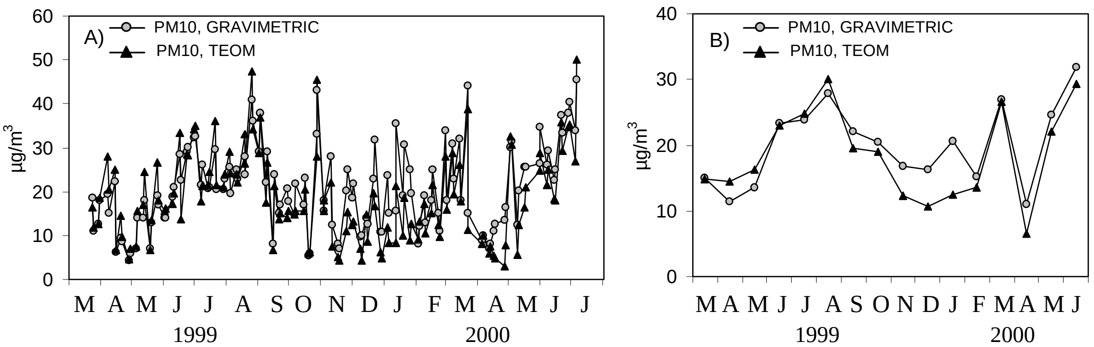


Figure 4-30. Daily (A) and monthly (B) PM10 levels measured at the MONAGREGA rural site from March 1999 to July 2000. Measurements performed at the rate of 2 samples per week.

This underestimation should be taken into account in the quantitative estimations of the seasonal evolution of PM for the rural sites discussed above (Figure 4-27A). At these rural sites, PM levels recorded from 1996-2000 corrected by the TEOM underestimation should present the summer maximum, but winter levels should be higher (around 33% according to the estimations for the period March 1999 – June 2000 period, Figure 4-30B).

Bearing in mind the underestimation of the automatic PM monitors, the number of exceedances of the daily EU PM10 limit value recorded with the gravimetric method may be slightly higher than that recorded with the TEOM or beta during the winter period at urban sites. As stated above, this should also be taken into account in the above estimations of the compliance of the EU limit values (section 4.5), mainly at urban sites.

This winter underestimation of PM levels does not modify the aforementioned seasonal evolution of the difference between the urban and rural PM concentrations (characterised by a winter maximum) given that PM levels have been measured with beta or TEOM equipment at urban and rural sites.

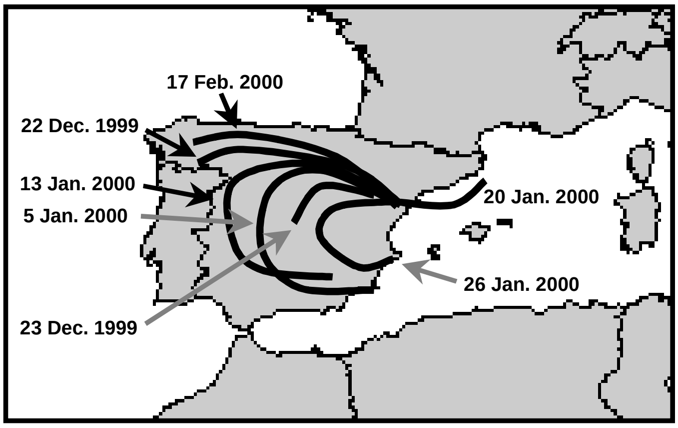


Figure 4-31. Back-trajectories associated with daily PM10 levels > 20µg/m^3 at MONAGREGA rural sites recorded during the period November 1999 – February 2000.

In the period November 1999 – February 2000, 7 daily PM10 samples with PM10 levels >20µg/m^3 were recorded with the gravimetric method. The meteorological analysis indicate that these sporadic PM10 peak events were caused by a regional transport of PM (Figure 4-31). Almost all these events were recorded under the prevalence of an anticyclonic situation over the Iberian peninsula. A NW air flow (the main wind field in the Ebro basin in winter) associated with an anticyclonic circulation over the Peninsula appears to be the main transport pattern (Figure 4-31).

4.7 Summary and conclusions

The results of this study demonstrate that the airborne particulate matter (PM) concentrations in the Western Mediterranean are strongly influenced by the regional meteorology. The Western Mediterranean is characterised by an abrupt coastal orography and frequently undergoes weak gradient baric conditions, which contrasts with the prevailing westerly air mass advection conditions (frequently associated with cold fronts and rain) and the flat terrain in Central and Northern Europe which favour air mass renovation and scavenging. On a day-to-day basis the main episodes affecting PM concentrations are: 1) Atlantic episodes with low PM levels induced by advective conditions, 2) Regional episodes with high PM and O_3 levels at rural and urban/industrial sites, 3) Local urban/industrial pollution episodes with high and low PM levels at urban/industrial and rural sites, respectively, and 4) African dust outbreak episodes with very high PM levels at all sites. The seasonal distribution of these episodes accounts for the PM seasonal evolution. The difference between PM levels at the urban and rural sites reaches a maximum in winter together with urban NOx and CO levels. The high winter urban PM episodes occur under scenarios that inhibit the pollutant dispersion (temperature inversion at low altitude and low wind speed under anticyclonic conditions). PM levels at the rural sites maximise in summer because of the frequent occurrence of meso-scale meteorological processes which favour the transport of pollutants from the urban/industrial to the rural sites, and because of the ageing of polluted air in the Western Mediterranean basin by the re-circulation of pollutants. African dust outbreaks are frequent between spring and early autumn, giving rise to the highest PM events. A secondary PM maximum in March in some years is caused by strong African dust outbreaks.

Exceedances of the daily EU limit value for PM10 (50µg/m^3) are recorded during different types of episodes. Several exceedances of the daily EU limit values are recorded every year because of African dust episodes at the rural, urban and industrial sites. At rural sites, the African dust events constitute the only cause of exceedances of the daily EU limit value. At urban sites, a significant number of exceedances of the daily EU limit value occur during Local urban episodes in autumn and winter. At sites where the breeze circulation induces a scarce renovation of air masses on the local scale, exceedances of the daily EU limit value are recorded mainly in summer.

The frequent occurrence of African and Regional PM episodes has implications for air pollution regulation strategies, and accounts for the marked difference in the features of the airborne particulates between Southern and Northern Europe. The African dust events are more frequently observed in Mediterranean countries than in Central-Northern Europe. Around 10 African dust events occur every year. Regional PM episodes are caused by the specific orography and the prevailing meteorology in the Western-Central Mediterranean which inhibit air mass renovation and scavenging and favour the airborne particulate accumulation in the regional atmosphere. When comparing two regions with the same degree of urban/industrial development (similar emission rates of air pollutants) in the Western Mediterranean and in Central-Northern Europe, higher background levels of airborne particulate pollutants are expected in the Mediterranean region because of the smaller "self-cleansing" capacity of the Mediterranean atmosphere.