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African dust contribution to ambient aerosol levels across central Spain: Characterization of long-range transport episodes of desert dust

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Abstract — This work presents a summary of the results obtained from the impact of the African dust on levels of atmospheric suspended particulate matter registered among different monitoring sites in the Madrid Air Basin, in the centre of the Iberian Peninsula. African dust outbreaks were identified over the period 2001-2008. Lidar measurements helped to assess the temporal evolution of the dust layers and its impact on surface boundary layer. Monthly trend was analyzed resulting in a late spring-summer maximum occurrence of episodes and the most common synoptic meteorological situations causing the transport of the African dusty air masses were identified over the seasons. Time series of particulate matter daily concentrations recorded across Air Quality Network monitoring stations and sampling campaigns were collected and analysed. The contribution of mineral dust source in airborne particulates was estimated at the sampling campaign sites by means of receptor modelling analyses. Our results have shown that the contribution of mineral dust to PM10 (particulate matter lower than 10 μm, aerodynamic diameter) during African dust outbreaks is highly significant at rural and urban sites of the Madrid Air Basin, giving rise to exceedances of the PM10 daily limit value (50 μg/m^3). Quantification of the emissions of mineral dust would help to establish efficient abatement strategies to reduce the concentration of PM10 with origin in anthropogenic sources, during African dust episodic days in which the PM10 DLV was exceeded in Madrid sites.

Keywords —African Dust, Daily Limit Value, PM10, Long-Range Transport

1 INTRODUCTION

On a global scale, the mineral fraction is the main component of atmospheric aerosol [1]. Mineral aerosols influence the atmospheric radiative balance through scattering and absorption processes and by acting as cloud condensation nuclei. Besides, may also greatly increase the ambient levels of particulate matter (PM) recorded in monitoring sites by means of local and regional dust resuspension processes and long-range transport episodes of desert dust (dust outbreaks). This kind of episodes is generated by massive resuspension processes in basins where a huge quantity of fine-grained mineral particulate matter was accumulated due to the erosion when torrential rains occur [2]. The main arid zones exporting mineral particles are located in North Africa, the Middle East and Central Asia. The effects of dust outbreaks are especially relevant in the Mediterranean area due to proximity of populated sites to the African mainland. European directive 2008/50/EC on air quality, takes specifically into account the potential exceedance of the PM10 (PM lower than 10 μm) daily limit value (DLV = 50 μg/m^3), due to the influence of natural sources, including "the transport of natural particles from arid regions" (article 2.15). In this work we tried to properly assess the role of desert dust on concentration levels and chemical composition of airborne PM, registered among different monitoring sites in the Madrid Air Basin during the period 2001-2008.

Firstly, the occurrence of African dust outbreaks over this area, were documented on the basis of a meteorological analysis and information obtained from satellite imagery and numerical models. Lidar measurements were analysed during specific events to check the entrainment of the dust layers on the boundary layer.

Then, the impact of the African dust transported by the air masses was evaluated on levels of PM10 recorded at rural background monitoring sites, located away from the direct influence of agglomerations and industrial sites. These sites quantify the natural background levels of PM in the study area and consequently the contribution of mineral dust will clearly increase these levels during African dust outbreaks.

Next, time series of PM10 daily concentrations recorded during specific sampling campaigns (performed at representative rural, urban-background and urban sites) and across different Air

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Quality Network (AQN) monitoring stations (urban-background, urban and urban traffic hot spots) were analysed to assess the influence of African dust on PM10 levels and composition and its impact on the exceedances of the PM10 daily limit value.

2 METHODOLOGY

The study area is located in the centre of the Iberian Península (Fig. 1). It is bordered to the north-northwest by the Guadarrama Range located 40 km from the metropolitan area and to the south by the Toledo Mountains. Industrial activity in this area consists essentially of light factories. Consequently the Madrid plume is typically urban, fed by traffic emissions and by domestic heating in the cold season.

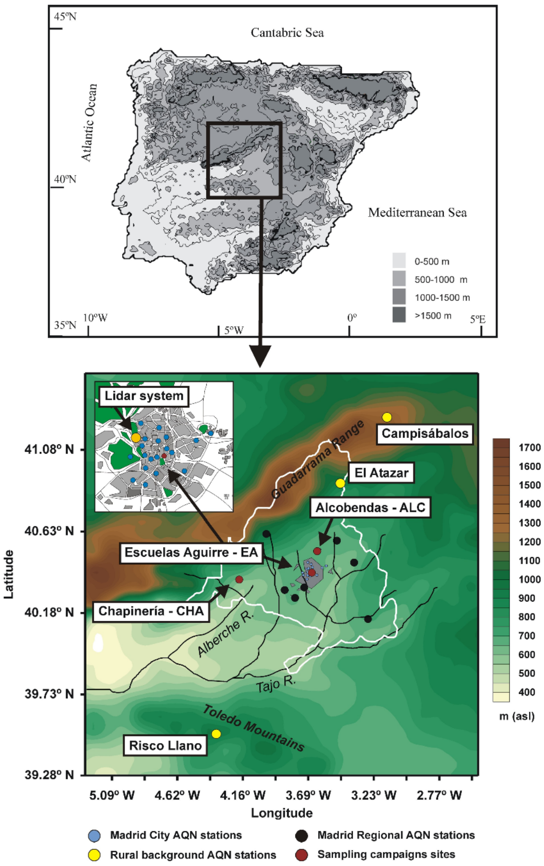

Fig. 1. Bi-dimensional topography of the Madrid Air Basin. The locations of the Lidar system, the AQN monitoring stations and the sampling campaign sites are shown. The metropolitan area, represented by the city of Madrid (up-left) and satellite towns has been shaded. White line represents the boundary of the Madrid province territory.

2.1 Detection of African dust outbreaks reaching the Madrid Air Basin

Synoptic charts and atmospheric back-trajectories, were analysed on a daily basis to estimate the origin of the air masses arriving over Madrid area.

5-day 3D backward air trajectories were computed using the FLEXTRA model and meteorological data provided from ECMWF (European Centre for Medium-Range Weather Forecasts) by the Norwegian Institute for Air Research (NILU). The model generated trajectories daily at 00:00, 06:00, 12:00 and 18:00 h UTC for the study period, with origin over Madrid metropolitan area (40.45°N, 3.72°W) at 1500 m asl.

A determination of the dominant synoptic wind flow patterns over the study area was carried out using a Cluster Analysis "[3], [4]". A non hierarchical method known as the k-means procedure was applied [5] using trajectory endpoints as the clustering variables. Synoptic meteorological situations associated to the transport of African air masses towards the Madrid Air Basin were determined and represented with meteorological variables from the NOAA/OAR/ESRL PSD-USA NCEP/NCAR Reanalysis datasets files [6].

The occurrence of African dust outbreaks were studied on the basis of this meteorological analysis and information obtained from satellite imagery and numerical models (TOMS-NASA, NRL, SKIRON, and ICoD-DREAM aerosol and dust maps (TOMS, http://www.jwocky.gsfc.nasa.gov; NRL, http://www.nrlmry.navy.mil/aerosol; SKIRON: http://www.forecast.uoa.gr; DREAM: http://www.bsc.es/projects/earthscience/DREAM), and satellite images provided by the NASA SeaWIFS project (http://seawifs.gsfc.nasa.gov/SEAWIFS.html)).

2.2 Lidar measurements

The lidar technique, which stands for LIght Detection And Ranging, operates in a similar way as radar, but using light instead of microwaves. Lidars have been used for several years to determine the planetary boundary layer height because of the large gradient in aerosol concentration that occurs between the top of the boundary layer and the free troposphere. The basic lidar products are vertical profiles of optical properties of aerosols, namely the extinction and backscattering coefficients. These properties are related with mass concentration through the so-called specific extinction coefficient, which depends on size distribution, shape and chemical composition of the mixture of aerosols.

The CIEMAT lidar system (Fig. 1) (40.42°N, 3.70°W, 669 m asl) is a laboratory equipment based on a Nd:YAG laser source (Spectra Physics LAB170-30, 115 mJ/pulse) operating at the 2nd harmonic (532 nm), a 30 cm diameter Newtonian telescope and photon-counting acquisition system.

Other instrument characteristics as well as the mathematical treatment of the raw signals have been described elsewhere [7].

This lidar station forms part of EARLINET (European Aerosol Research Lidar NETwork) since 2006. This network of lidar stations comprises 25 stations distributed over Europe and study tropospheric aerosols on continental scale. The network activity is based on scheduled measurements twice a week, a rigorous quality assurance program both at algorithm and instrument level, and a standardized data exchange format. CIEMAT lidar measurements were used to study the temporal evolution of aloft dust-rich layers during African dust outbreaks.

2.3 Time series analysis

Time series of PM10 daily concentrations recorded at rural-background stations were analysed to assess the impact of the African dust on surface PM10 levels.

During the period 2001-2008 there were two different AQN that covered the Madrid province territory taken as work domain in this study (Fig. 1). Madrid City AQN stations were located in almost of the districts of the city. Otherwise Madrid Regional AQN stations were distributed throughout the province. However, rural stations were not incorporated at these AQN for PM sampling until the year 2006. For this reason two rural background monitoring sites included in the rural background network of UNECE/LRTAP/EMEP (European Monitoring and Evaluation Programme) were utilized in this study for the 2001-2006 period. These two sites were located on the far Northeastern (Campisabalos) and Southwestern (Risco Llano) limits of the Madrid Air Basin, respectively (Fig. 1) and provided regularly PM10 daily concentration levels in this term. Both stations were not working for long lasting periods after the year 2006. Therefore, PM10 concentration data obtained at a rural station from the Madrid Regional AQN, El Atazar (Fig. 1) were used for the 2007-2008 period.

The PM10 measuring method of the Madrid City and Madrid Regional AQN stations was continuous, based on the Tapered Element Oscillating Microbalance (TEOM) and beta attenuation method, respectively. The standardised gravimetric manual method was only used at the EMEP stations.

Furtherly, several 1-year sampling campaigns were carried out to characterize mean PM10 and PM2.5 concentration levels and their chemical composition at three representative sites of this area (Fig. 1). The first study was conducted throughout the year 2001 at an urban park located in Alcobendas (ALC; 40.55° N, 3.63° W, 667 m asl), a smaller town 13 Km away from Madrid city. The second study was carried out from May 2004 to April 2005 at the outskirts of Chapinería (CHA; 40.38° N, 4.20° W, 675 m asl), a small village 25 Km southwest from Madrid City located in a rural area (Fig. 1). ALC and CHA sites can be considered as urban-background and rural-background sites, respectively. The third one was conducted from January 2007 to April 2008 at an urban traffic site (Escuelas Aguirre-EA; 40.42° N, 3.68° W, 672 m asl) in the Madrid city downtown.

At each site, 24-h sampling was carried out by means of a PM10 Graseby-Andersen high volume sampler (68 m^3/h), which is an EN12341 reference instrument, and a MCV high volume sampler (30 m^3/h) equipped with a PM2.5 inlet and quartz micro-fibre filters (QF20 Schleicher and Schuell). PM10 and PM2.5 manual sampling was carried out at a rate of 2 moving days per week. During the campaign performed in ALC only one PM2.5 sample was obtained every week.

A number of PM10 and PM2.5 filters were collected at the ALC urban background (84 and 34), CHA rural (98 and 96) and EA urban (95 and 104) sites. Following sampling and standard gravimetric determination of the PM mass concentration levels, major and trace elements and compounds were analysed in PM10 and PM2.5 filters, with a total of 57 determinations per sample (see methodology in [8]).

Finally, receptor modeling analyses were performed using Positive Matrix Factorization-PMF [9] to characterise the main PM10 sources and quantify the contribution of mineral dust in airborne particulates. This receptor model assesses correlations between the input variables and provides as output a number of factors which are interpreted as emission sources. Uncertainties used in the receptor modelling analysis were calculated based in [10]. Source contributions to daily PM10 concentrations were estimated by means of multilinear regression analysis.

2.4 Assessment of the impact of the African dust outbreaks on the exceedances of the PM10 DLV

As it was stated in the Introduction section, European directive on air quality (2008/50/EC) takes specifically into account the potential exceedance of the PM10 DLV due to the transport of natural particles from arid regions. In these cases the exceedances caused by natural episodes may be discounted by the Member States after careful justification of their natural origin.

With the aim to qualitatively estimate the impact of the African dust contributions on the daily concentrations of PM10, the PM10 DLV exceedances registered at the different Madrid AQN monitoring stations during days with African dust transport, were recorded during the 2001 – 2008 period. For this study we have analysed only the time series of pollutants from those stations with the best data coverage (daily data coverage > 70% for every year of the 2001-2008 period) which were not re-located in this period. Finally, the time series of PM10 daily levels used were obtained from 20 urban stations, from which 7 were located at hot spots strongly affected by road traffic (mean annual levels of NO_x, NO_2 and CO higher than 170 µg/m^3, 75 µg/m^3 and 0.9 mg/m^3, respectively) and 2 urban background stations (Fig. 1).

Results obtained with the receptor modeling analyses allowed to estimate the mean contribution of mineral dust to the PM10 mass during days with African dust transport, hereafter African dust episodic days and days exceeding the PM10 DLV at the urban, urban-background and rural sampling sites.

Afterwards a procedure for the quantification of the net African dust load transported was applied to estimate the impact of the African dust contributions, on the PM10 DLV exceedances registered at Madrid monitoring stations during African dust episodic days. This methodology built on the identification of days with African dust transport and statistical analyses based on the calculation of the 30 days moving 40th percentile for regional background PM10 time series "[2], [11]". This percentile is an indicator of the non-African regional background to be subtracted from the daily PM10 levels during African dust outbreaks, and thus allows calculating the daily net African dust contribution. This methodology became the Spanish and Portuguese reference method to identify and quantify African dust contributions to PM10 levels since 2004. It has been applied to data on levels of PM10 measured at regional background sites across the Mediterranean Basin [12] and compared to other measurement-based methods to quantify the African dust contribution to PM10 levels [13]. The results showed that the method is applicable across the whole Southern Europe and is the most adequate available at the moment, from an EU policy perspective. It has been recently accepted as one of the methodologies to be included in the EC Guidance for Member States regarding natural events, currently under preparation according to directive 2008/CE/50.

3 RESULTS AND DISCUSSION

3.1 Occurrence of African dust outbreaks

During the period 2001-2008, 145 African dust episodes were identified by means of the meteorological analysis and the satellite imagery and numerical model results inspection, accounting for a total of 519 episodic days. On average 18 episodes per year (65 days/year) were identified increasing the daily concentration levels of PM10 recorded in the rural-background stations, with a mean duration of 3.6 days per episode. The highest number of episodes was recorded in 2004 and 2007 (21 and 22 episodes respectively) and the lowest in 2006 (15 episodes).

The monthly mean distribution of African episodes is shown in Fig. 2.

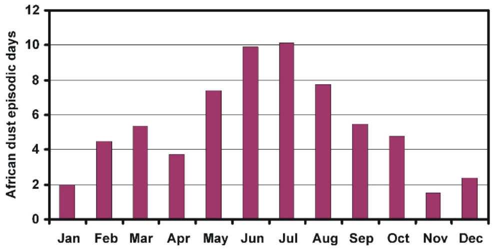

Fig. 2. Monthly number of African dust episodic days, averaged over the 2001-2008 period.

There is a clear monthly trend towards a higher frequency of African dust episodes during the late spring-summer months (from May to August) and in lesser extent during the February-March and September-October terms. In particular the highest number of African dust episodic days per month was registered in the period June-July (10 days per month on average). The lowest number of African dust episodic days per month was recorded from November to January (2 days per month on average).

The effect of the dust outbreaks was clearly detectable in the PM10 values determined at the Risco Llano, Campisábalos and El Atazar rural-background stations. Fig. 3 shows how the main peaks of the time series PM10 coincide with dust events.

In fact there is a positive relationship between the number of African dust outbreaks and the number of PM10 DLV exceedances registered at rural-background stations in this area. In the period 2001-2006, 30 out of 31 and 20 out of 21 PM10 DLV exceedances were registered during African dust outbreaks at the Risco Llano and Campisabalos stations, respectively. Moreover the whole number of PM10 DLV exceedances registered at El Atazar in 2007 and 2008 (n=16) occurred during African dust outbreaks.

These results shown that African dust usually entrain into the boundary layer during outbreaks, subsequently descending to ground-level, where it may be detected by AQN monitoring stations. The mixing of the lower troposphere with dust-rich layers is enhanced during summer by the greater thickness of the mixing layer [14]. In winter this mixing can not be assured unless vertically-resolved information is available because of the smaller development of the mixing layer. The lidar technique can close this gap by providing vertically-resolved aerosol optical properties.

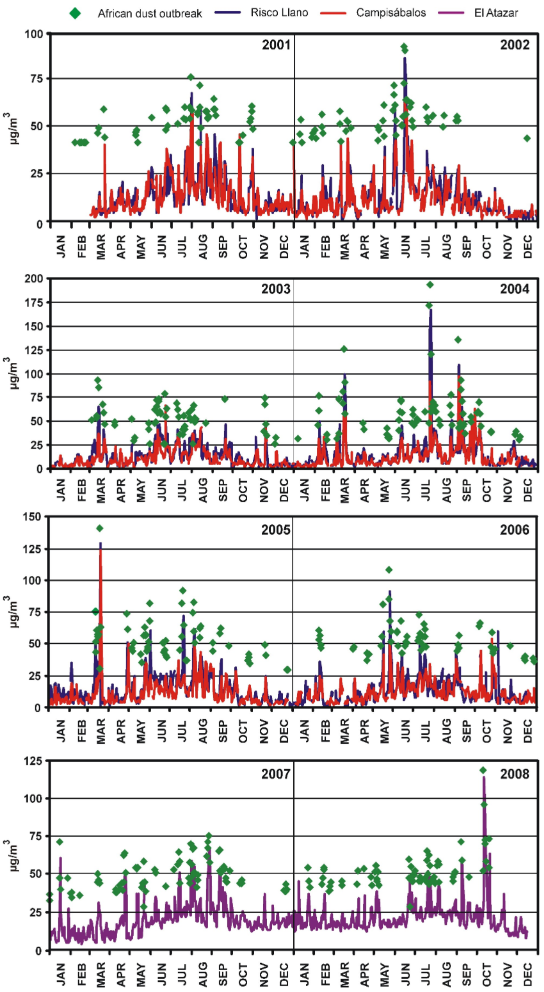

Fig. 3. Daily PM10 concentration levels from the rural stations of Risco Llano, Campisábalos and El Atazar over the 2001-2008 period. African dust outbreaks episodic days are highlighted with green rhombus.

An example of the detection of aloft dust-rich layers during an African dust outbreak and its temporal evolution by interpolation of Lidar profiles can be seen in Fig. 4.

It shows the extinction coefficient expressed as m^{-1}, derived from Lidar measurements, for June 28th 2008 at the first 12 hours. Vertical profiles were interpolated using Kriging algorithm in order to obtain a visual representation of the boundary layer evolution, using aerosols as tracers. A relevant vertical structure was found on this period, with a high aerosol load layer located between 1.5 and 3 km and probably decoupled from surface (Fig. 5a-b). Such structure could be due to the arrival of dust-rich air mass from the Sahara desert. The meteorological analysis confirmed the occurrence of an African dust outbreak over the Madrid Air Basin for this day. During the morning, the aerosol load in the boundary layer was mixed with the layer aloft, as it can be seen in the right-most part of Fig. 4 and specifically in the vertical profiles of the extinction coefficients obtained at 11:00 and 12:00 h UTC (Fig. 5c-d). Thus it is demonstrated that African dust particles can be mixed with PM emissions from local and regional sources inside the boundary layer of the Madrid Air Basin.

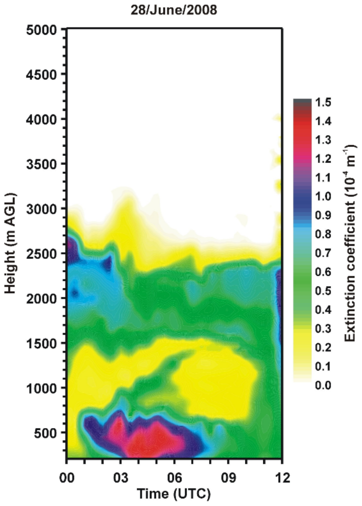

Fig. 4. Extinction coefficient profiles interpolated over the 00:00-12:00 h (UTC) 28/June/2008 period.

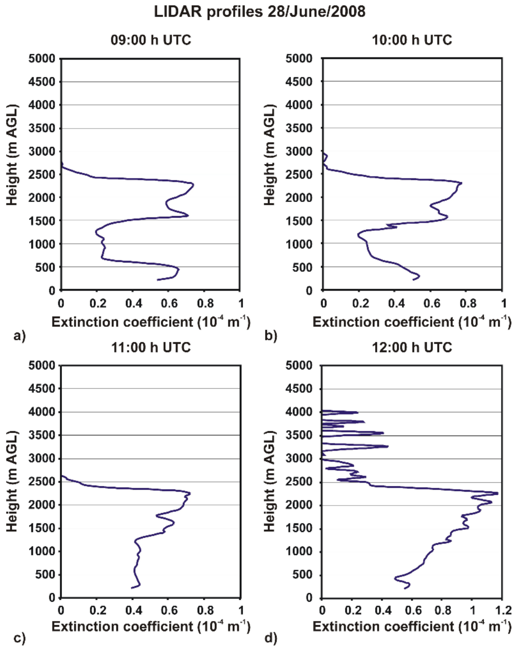

Fig. 5. Vertical profiles of the extinction coefficient derived from Lidar measurements at 09:00 (a), 10:00 (b), 11:00 (c) and 12:00 (d) h UTC for June 28.

3.2 Atmospheric transport scenarios

Cluster Analysis was used to group trajectories into homogeneous groups, depending on direction and speed of transport. Each cluster represents a dominant flow pattern. 8 different clusters were obtained as the best number for describing significantly different air-flow patterns, depending on their direction and speed (Fig. 6). Most clusters show marked seasonality.

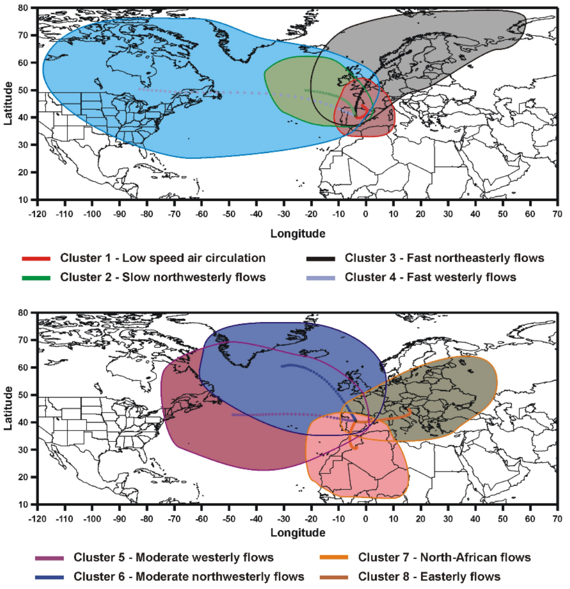

Fig. 6. Cluster centres (average of the members of each cluster) resulting from the analysis. Horizontal standard deviations about each cluster centre are represented by the shaded contour.

Cluster 1 – 17% of the total number of trajectories, contained short trajectories of air mass recirculating within the Iberian Peninsula (Fig. 6, Top panel). A slow moving north-westerly flow was represented by cluster 2 – 17% (Fig. 6, Top panel). These air flows were produced in the summer as a consequence of the influence of the quasi-persistent Azores high over the Iberian Peninsula.

Cluster 3 – 5% represented north-easterly flows moving fast along the North European coastal corridor (Fig. 6, Top panel). The fastest-moving trajectories from the Atlantic Ocean and North America were grouped in cluster 4 – 11% (Fig. 6, Top panel). Both kind of fast air flows were more frequent during the winter, being generated by strong baric gradients.

The advection of moderate flows from Atlantic Western (cluster 5 – 22%) and Northern regions (cluster 6 – 12%) took place more frequently during the spring and autumn months and less during the summer (Fig. 6, Bottom panel).

Cluster 7 – 9% was characterized by trajectories from the North-African regions of Morocco, Algeria, Tunisia, Libya, Western Sahara, Mauritania, Mali and Niger and from Atlantic areas near the north western African coast (Fig. 6, Bottom panel). These flows were registered throughout the year, although they could be more likely in the June-October period.

No clear seasonal trends were found for the occurrence of cluster 8 – 8%. It contained trajectories of air mass coming from the European continent and the western and central Mediterranean area (Fig. 6, Bottom panel).

On the basis of these results, different synoptic meteorological situations, depending on the season, originated the transport of African dusty air masses towards the centre of the Iberian Peninsula, represented by the Cluster 7. The most common synoptic situations causing this transport have been represented by composite 850 mb geopotential height maps in Fig. 7.

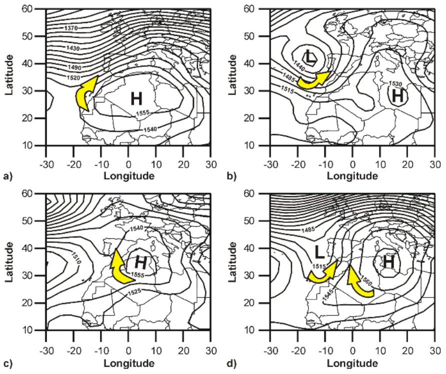

Fig. 7. Composite 850 mb geopotential height (m) for the African flows arriving over the Madrid Air Basin during the winter (a), spring (b), summer (c) and autumn (d) at 12:00 h UTC..

The winter meteorological scenario was identified by a high pressure system centred over Argelia and extended from Western Sahara to Libya (Fig 7a).

The spring transport scenario was characterised by a deep low, centred west of the Portuguese coast (Fig 7b).

In the summer, the intense heating of the Northern-Africa surface generated the development of a thermal low. A compensatory high pressure system was formed in the upper atmospheric levels (850 hPa), frequently over Northern Argelia (Fig 7c).

During the autumn, the African high was shifted to the North and a trough is observed southwest of the Portuguese coast (Fig 7d).

The summer and autumn typical synoptic meteorological situations generated events which are characterised by a wider extension of the African air mass invading the Iberian Península. On the contrary the winter and spring synoptic situations frequently generate the development of a mass of mineral dust with a well defined convex morphology, which turns over the Atlantic Ocean reaching the Iberian Península from the west side “[2], [15]”.

3.3 Chemical characterization

With the aim to quantify the impact of the African dust input on the ambient PM levels, the chemical composition of the PM10 and PM2.5 samples obtained during the sampling campaigns at EA, ALC and CHA was studied.

During the experimental studies, mean PM concentrations reached 41 µgPM10/m^3 and 21 µgPM2.5/m^3 at EA urban site, 32 µgPM10/m^3 and

25 µgPM2.5/m^3 at ALC urban background site and 32 µgPM10/m^3 and 17 µgPM2.5/m^3 at CHA rural site. Results obtained from a previous study at EA shows that PM2.5 concentrations reduced by 38% from 1999 to 2008. This behaviour in the trend of fine PM concentrations in Madrid can be attributed to a reduction in the emissions from residential coal burning and in a lesser extent from traffic. The same fact could be the reason why mean PM2.5 concentrations registered at the urban-background site in 2001 were higher than those registered at the urban site in 2007-2008. On the contrary mean PM10 levels at ALC and CHA were equal and 22% lower than at EA. It suggests the existence of relevant contributions of coarse PM from different origins at rural and urban sites in the Madrid Air Basin.

Crustal material contribution (sum of elements typically found in rock-forming minerals, including Al_2O_3, SiO_2, CO_3^{2-}, Ca, Fe, K, Mg, Mn, Ti and P) to PM10 and PM2.5 bulk mass was elevated, especially for the coarse fraction. In relative terms this contribution was higher at the rural site (35% and 28%, respectively) than at the urban (31% and 12%, respectively) and urban-background (27% and 11%, respectively) sites.

3.4 Source apportionment study

Receptor modelling analysis agrees with the identification of four main sources or origins for PM10 at EA, ALC and CHA sampling sites: Emissions from **vehicular traffic** (with C, Fe, Sb, Cu as main tracers), **mineral dust** (Al, Ca, K, Ti, Mn,...), **secondary inorganic aerosol - SIA** (SO_4^{2-}, NO_3^-, V and NH_4^+) and **sea salt** (Cl^-, Na and Mg). **residential coal combustion** (As, Se,...), **Industrial** (Cr, Ni, Pb,..) and **biomass burning** (P, C,...) were sources identified exclusively at the urban, urban-background and rural-background sites, respectively. An **African dust** (Al, Ti, K, Mn, Sr...) source category was discriminated as an individual source in the urban site [13]. However with the aim to compare mean source contributions among different sites and periods, we grouped the contributions of the **African dust** and local-regional **mineral dust** sources into the single category **mineral dust** at the site. Having identified the main PM10 sources, their contribution to the daily samples mass were estimated. Mean contributions to bulk PM10 levels are shown in Table 1.

The mean contribution of mineral dust to the PM10 mass was obtained for African dust episodic days and days exceeding the PM10 DLV. Results are shown in Fig. 8.

In any case a relevant mineral contribution was obtained, higher in the rural than in the urban background and urban-traffic sites. Mineral contribution ranged from 48 to 66% (22.3-35.3 µgPM10/m^3) of the total bulk mass of PM10 during African dust episodic days (Fig. 8a).

Table 1. Source contribution to gravimetric PM10 (in µg/m^3) resulting from the PMF analysis, at EA, ALC and CHA.

	EA	ALC	CHA
Mineral	12.7	8.0	12.8
Traffic	14.6	8.4	4.2
SIA	7.3	8.8	8.6
Sea salt	2.8	2.4	1.8
Industrial	-	4.2	-
Coal combustion	3.9	-	-
Biomass burning	-	-	3.6
Unknown	0.1	0.2	0.0

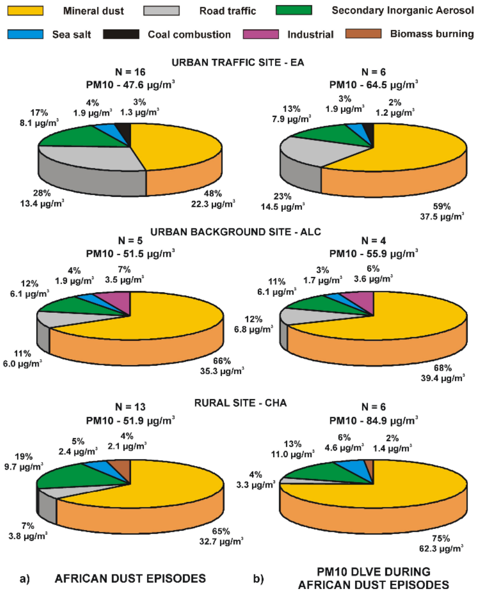

Fig. 8. Mean source contributions to PM10 registered at EA, ALC and CHA sampling sites during African dust episodic days (a) and African dust episodic days exceeding the PM10 DLV (b). N = number of daily cases.

During days exceeding the PM10 DLV this contribution represented 27-54% (19.1-39.5 µgPM10/m^3) of the total mass.

However when the exceedance of the PM10 DLV occurred simultaneously with an African dust episodic day, mineral contribution increased, representing 59% of the total mass of PM10 at the urban-traffic site, 68% at the urban-background site and 75% at the rural site (Fig. 8b).

These results show the high contribution of mineral dust to PM10 during African dust outbreaks recorded at rural and urban sites of the Madrid Air Basin. Mineral dust contribution can give rise to exceedances of the PM10 DLV. However it is still unclear the contribution of the net dust load transported by the African air masses.

3.5 Assessment of the impact of the African dust outbreaks on the exceedances of the PM10 DLV

On average, 35%, 43% and 58% of the annual PM10 DLV exceedances registered at Madrid hot-spots, urban and urban-background stations respectively, were detected during African dust outbreaks in the period 2001-2008 (Fig 9).

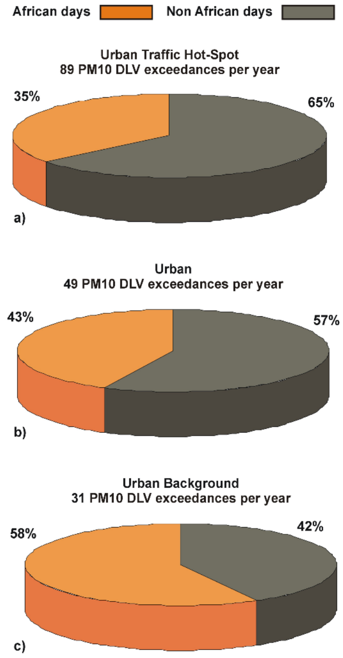

Fig. 9. Mean annual number of daily exceedances of the PM10 DLV produced during African dust episodic days.

However it can not be assured that all these exceedances of the PM10 DLV were exclusively attributed to the net load of African dust.

For this reason the daily net dust load in PM10 was obtained at the rural-background stations for any African dust episodic day, following the procedure previously exposed in section 2.3 [2].

On average the net load of African dust obtained during African dust episodic days was 16 µgPM10/m^3 at Campisábalos and 20 µgPM10/m^3 at Risco Lano during the 2001-2006 term. A mean value of 15 µgPM10/m^3, was obtained at El Atazar for the 2007-2008 period. The highest mean contribution of African dust net load was obtained in the summer months at the three sites, when synoptic meteorological situations as the one described in Fig. 7c, were frequently produced.

For those days in which the PM10 DLV was exceeded at any station (urban-background, urban or urban traffic hot-spots) during an African dust outbreak, the net dust load obtained in the closest rural-background station was subtracted from the PM10 daily mean value registered at this station. If the result of the subtraction was lower than 50 µgPM10/m^3, the PM10 DLV exceedance registered in the considered station was attributed to the African dust contribution. Results are shown in Fig. 10.

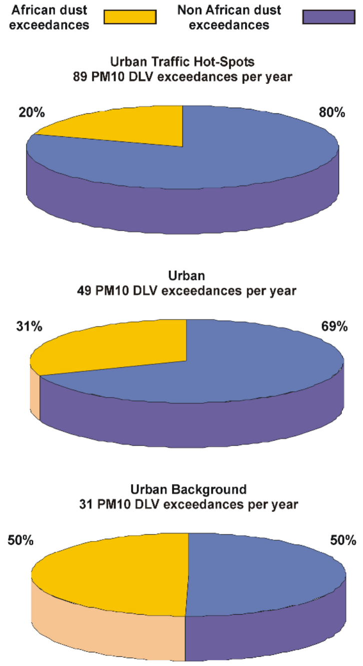

Fig. 10. Mean annual number of daily exceedances of the PM10 DLV due to African dust and due to other causes.

Thus, on average during the 2001 – 2008 period, 18%, 28% and 45% of the total PM10 DLV exceedances registered at Madrid hot-spots, urban and urban-background stations respectively, were exclusively attributed to African dust.

The number of PM10 DLV exceedances attributed to African dust is higher, in relative terms, at urban-background stations than at urban and urban traffic hot-spots stations. As the urban stations are less isolated from the influence of direct anthropogenic emissions, especially in the case of urban traffic hot-spots stations, there is a high proportion of PM10 DLV exceedances attributed to other causes different from African dust. In absolute terms, the number of PM10 DLV exceedances attributed to African dust is very similar at any monitoring site (14 at urban-background and urban stations and 16 at urban traffic-hot spots stations).

Afterwards, the PM10 mean annual concentration was calculated for the AQN stations PM10 daily concentration time series and for the time series obtained after subtracting the net dust load for the African dust episodic days. The difference between both mean annual values is an estimation of the annual contribution of African mineral dust to the mean concentration levels of PM10 [2].

In this study the values obtained of the mean annual net African dust contribution to the annual mean PM10 means recorded at the AQN monitoring stations ranged from 2 to 4 µgPM10/m^3. The annual limit value for PM10 is 40 µgPM10/m^3. Hence, the African dust contribution to the annual limit value is much lower than the daily limit value of PM10 in the Madrid monitoring sites.

4 CONCLUSIONS

The results of this work demonstrate that during African dust episodic days, mineral dust represents a significant contribution to daily PM10 levels registered at rural and urban monitoring sites in the Madrid Air Basin.

On average 18 African dust outbreaks per year (65 days/year) were identified influencing the concentration levels of PM10 recorded in rural-background monitoring stations, with a mean duration of 3.6 days per episode.

Different synoptic meteorological situations, originated the transport of African dusty air masses towards the centre of the Iberian Peninsula in any season. However there is a clear monthly trend towards a higher frequency of African dust episodes during the May-August period and in lesser extent during the February-March and September-October terms.

The effect of the dust outbreaks was clearly detectable in the PM10 values determined at rural and urban AQN monitoring stations and specific sampling sites. Estimations of the mineral contribution, ranged from 48 to 66% of the total bulk mass of PM10 during African dust episodic days, at different representative sampling sites. However when the exceedance of the PM10 DLV occurred simultaneously with an African dust episodic day, mineral contribution increased, representing from 59% to 75% of the total bulk mass of PM10 higher in the rural than in the urban background and urban-traffic sites.

In fact, 35%, 43%, 58% and 96% of the mean annual PM10 DLV exceedances registered at Madrid hot-spots, urban, urban-background and rural-background stations respectively, were detected during African dust outbreaks. Moreover 18%, 28% and 45% of the total PM10 DLV exceedances registered at Madrid hot-spots, urban and urban-background stations respectively, could be exclusively attributed to the African dust net load.

It can be concluded that the net contribution of mineral dust to PM10 during African dust outbreaks, give rise to an elevated number of exceedances of the PM10 DLV, at rural and urban sites of the Madrid Air Basin. On the contrary, the net dust load transported by the African air masses, does not significantly alter the mean annual PM10 concentration values.

Our results suggest that the application of methodologies with the aim to assess the contributions of mineral dust, such as the detection and subsequent quantification of mineral dust contribution to PM levels during African dust outbreaks, would be very helpful in designing effective abatement strategies to reduce the concentration of PM10 with origin in anthropogenic sources, in African dust episodic days in which the PM10 DLV was exceeded in Madrid sites.

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REFERENCES

[1] IPCC. Climate Change 2001: The Scientific Basis, Cambridge University Press, 2001.

[2] Spanish Ministry of the Environment, Rural and Marine Affairs. Methodology for the Identification of Natural Episodes in PM10 and PM2.5, and Justification with Regards to the Exceedances of the PM10 Daily Limit Value. Spanish Ministry of the Environment, Rural and Marine Affairs: Madrid, Spain, January 2010; p 40.

[3] P. Salvador, B. Artiñano, X. Querol and A. Alastuey, "A combined analysis of backward trajectories and aerosol chemistry to characterise long-range transport episodes of particulate matter: The Madrid Air Basin, a case study" Science of the Total Environment 390 pp. 495-506, 2008

[4] P. Salvador, B. Artiñano, C. Pio, J. Afonso, M. Legrand, H. Puxbaum and S. Hammer, "Evaluation of aerosol sources at European high altitude background sites with trajectory statistical methods" Atmospheric Environment 44 pp. 2316-2329, 2010

[5] R.C. Owen, "A Climatological Study of Transport to the PICO-NARE Site Using Atmospheric Backward Trajectories". Civil and Environmental Engineering, Michigan Technological University, Master of Science, Houghton, MI, 361 pp., 2003

[6] E. Kalnay, M. Kanamitsu, R. Kistler, W. Collins, D. Deaven, L. Gandin, M. Iredell, S. Saha, G. White, J.

Woollen, Y. Zhu, M. Chelliah, W. Ebisuzaki, W. Higgins, J. Janowiak, K.C. Mo, C. Ropelewski, J. Wang, A. Leetmaa, R. Reynolds, R. Jenne and D. Joseph, "The NCEP/NCAR 40-year reanalysis project" Bulletin of the American Meteorological Society 77 pp. 437-470, 1996

[7] F. Molero, and F. Jaque, "The laser as a tool in environmental problems" Optical Materials 13 pp. 167-173 1999

[8] X. Querol, A. Alastuey, M.M. Viana, S. Rodriguez, B. Artíñano, P. Salvador, S. Garcia, R. Fernandez, C. Ruiz, J. de la Rosa, A. Sanchez, M. Menendez and J.Gil, "Speciation and origin of PM10 and PM2.5 in Spain" Journal of Aerosol Science 35 pp. 1151-1172, 2004

[9] P. Paatero and U. Tapper, "Positive matrix factorization: a non-negative factor model with optimal utilization of error estimates of data values" Environmetrics 5 pp. 111-126, 1994

[10] A. Polissar, P. Hopke and P. Paatero, "Atmospheric aerosol over Alaska: 2. Elemental composition and sources" Journal of Geophysical Research pp. 19045-19057, 1998

[11] M. Escudero, X. Querol, J. Pey, A. Alastuey, N. Pérez, F. Ferreira, S. Alonso, S. Rodríguez and E. Cuevas, "A methodology for the quantification of the net African dust load in air quality monitoring networks". Atmospheric Environment 41 pp. 5516-5524, 2007

[12] X. Querol, J. Pey, M. Pandolfi, A. Alastuey, M. Cusack, N. Pérez, T. Moreno, M. Viana, N. Mihalopoulos, G. Kallos, S. Kleanthous, "African dust contributions to mean ambient PM10 mass-levels across the Mediterranean basin" Atmospheric Environment 43 pp. 4266-4277, 2009

[13] M.M. Viana, P. Salvador, B. Artiñano, X. Querol, A. Alastuey, J. Pey, A.J. Latz, M. Cabañas, T. Moreno, S. García, M. Herce, P. Diez, D. Romero and R. Fernández, "Assessing the performance of methods to detect and quantify African dust in airborne particulates" Environmental Science & Technology 44 pp. 8814-8820, 2010

[14] S. Crespí, B. Artíñano and H. Cabal, "Synoptic clasification of the Mixed-Layer height evolution" Journal of Applied Meteorology 34 pp. 1666-1676, 1995

[15] P. Salvador, B. Artiñano, D. Alonso, X. Querol and A. Alastuey, "Identification and characterisation of sources of PM10 in Madrid (Spain) by statistical methods" Atmospheric Environment 38 pp. 435-447, 2004