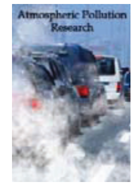
Atmospheric Pollution Research 6 (2015) 70-81



Atmospheric Pollution Research

www.atmospolres.com

Assessing the mineral dust from North Africa over Portugal region using BSC–DREAM8b model

Alexandra Monteiro [1], Ana Patricia Fernandes [1], Carla Gama [1], Carlos Borrego [1], Oxana Tchepel [2]

[1] *CESAM & Department of Environment and Planning, University of Aveiro, 3810–193 Aveiro, Portugal*
[2] *CITTA, Department of Civil Engineering, University of Coimbra, 3030–788 Coimbra, Portugal*

ABSTRACT

Over the last decade, air pollution has become a major problem in Portugal mainly due to the high concentrations of particulate matter in the atmosphere, which surpassed the daily limit values. An abundant type of natural atmospheric aerosol is related with the suspension and long–range transport of mineral dust from North Africa deserts. The main objective of this work was to assess the mineral dust over Portugal, namely in what concerns both long–term period (one year) and episode peaks. The BSC–DREAM8b v1.0 model was applied for the entire year of 2011 and the modeled surface concentrations were explored. The annual mean of the simulated dust has a magnitude of 2–6 μg m^{-3}. The monthly average analysis highlights the largest mineral dust average values in April and May (about 4 μg m^{-3} higher than the other months). The influence of the transport of mineral dust from North Africa to Portugal is limited on time scale, since in 50% of the time this contribution is below 0.2 μg m^{-3}. Only when high percentiles are analyzed the dust surface concentrations over Portugal become relevant (>3 μg m^{-3}; with peak contribution around 10–25 μg m^{-3}). To characterize the strongest episodes of dust, a group of days with modeled surface daily concentrations above 5 μg m^{-3} was selected, and data were extracted for 7 sites, spatially distributed along Portugal. A cluster analysis of the air parcels back trajectories that arrive at each site was performed in order to identify the mean flow patterns associated to each mineral dust episode. The prevalence of the flow regimes coming from North Africa during the episode days was different for the studied sites, with high frequency (above 70%) at south sites. This work contributes to the characterization and assessment of the dust episodes that affect Portugal, on a yearly basis and based on a modeling approach.

Corresponding Author:
Alexandra Monteiro
☎ : *+351-234-370220*
🖷 : *+351-234-370309*
✉ : *alexandra.monteiro@ua.pt*

Article History:
Received: 22 April 2014
Revised: 18 July 2014
Accepted: 18 July 2014

Keywords: *Mineral dust, BSC–DREAM8b model, long–term assessment, episodes, cluster analysis*

doi: 10.5094/APR.2015.009

1. Introduction

Atmospheric aerosols cause detrimental health effects (e.g. Pope and Dockery, 2006; WHO, 2006a; WHO, 2006b) and have direct and indirect effects on the climate system (IPCC, 2001; IPCC, 2007). Additionally to the anthropogenic pollution, natural sources of the atmospheric aerosol may play an important role in the occurrence of pollution episodes (Perez et al., 2006a; Niemi et al., 2009). On a global scale, most of the atmospheric particles are emitted by natural sources, mineral dust being the second most abundant component after sea–spray derived aerosols (IPCC, 2007). A large amount of mineral dust is mobilized over arid regions and injected into the atmosphere under specific weather conditions (Prospero, 1999; Ansmann et al., 2003).

Natural contributions to air pollution in Europe have been debated in the EU Report (IES–JRC, 2007). According to this document, the contribution of the natural sources to the PM levels at European level may range from 5% to 50%. Mineral dust outbreaks are one of the main causes of high PM_{10} particle mass concentrations in Southern Europe (Rodriguez et al., 2001; Borge et al., 2007; Wagner et al., 2009) and several studies stress the importance of mineral dust long–range transport from North African deserts to specific Mediterranean Southern European countries like Portugal (Reis et al., 2002; Fialho et al., 2006; Freitas et al., 2007).

The European Directive for air quality (2008/50/EC) provides the possibility to subtract the contribution of natural sources before comparing the ambient air pollutant concentrations to the limit values, which may therefore reduce the number of exceedances in the reporting system due to the non–anthropogenic origin of the pollution (EC, 2011). The identification of such episodes and quantification of daily and annual contributions of desert dust to PM is currently necessary. In the absence of a single, recognized standard procedure, various studies have been conducted to assess the impact of Saharan dust on PM_{10} concentrations in Europe, especially in Spain (Escudero et al., 2007a; Escudero et al., 2007b).

Air quality models are powerful tools to predict the fate of aerosols after their release into the atmosphere. Several models were designed to describe the atmospheric life cycle of the eroded desert dust, which take into account all major processes of the dust life cycle such as production, horizontal and vertical diffusion and advection and wet and dry deposition. The World Meteorological Organization (WMO) recently established the Sand and Dust Storms Warning Advisory and Assessment System (SDS-WAS, 2014) to enhance the ability of countries to deliver timely and quality sand and dust forecasts, observations, information, and knowledge to users. Within this program, the North Africa, Middle East and Europe Regional Center SDS–WAS, hosted by the Spanish Meteorological Agency (AEMET) and the Barcelona Supercomputing Center – Centro Nacional de Supercomputacion (BSC–CNS), aims to lead the development and implementation of a system for dust observation and forecast. Currently, this Regional Center distributes forecasts over North Africa and Europe from eight models, including the regional BSC–DREAM8b model (Perez et al., 2006a; Perez et al., 2006b; Basart et al., 2012a), which are near–real–time evaluated.

In this study, the influence of African dust outbreaks over Portugal is carried out using mineral dust surface concentrations simulated by the BSC–DREAM8b v1.0 model (Perez et al., 2006a; Perez et al., 2006b; Basart et al., 2012a) over the whole year of 2011. The spatial and temporal patterns of the mineral dust over Portugal are analyzed, in terms of long–term and high peak episode concentrations. The synoptic patterns associated to the high episodes are also analyzed using back–trajectories clustering approach and their relevance are discussed.

2. Methodology

2.1. BSC–DREAM8b model description

The BSC–DREAM8b v1.0 model (Perez et al., 2006a; Perez et al., 2006b; Basart et al., 2012a), developed at the BSC–CNS, has been used in this work to characterize long–range transport of mineral dust from North African deserts to Portugal. The model solves the Euler–type partial differential non–linear equation for dust mass continuity, and it is fully embedded as one of the governing prognostic equations in the Eta atmospheric model (Black, 1994). Thus, the model is able to simulate and predict the 3–dimensional field of dust concentration in the troposphere taking into account all major processes of dust life cycle.

The main features of BSC–DREAM8b, described in detail in Perez et al. (2006a), include a source function based on the aridity categories of the 1 km USGS land use data set, a dust size distribution profile described by 8 size bins within the 0.1–10 µm radius range according to Tegen and Lacis (1996), a source distribution derived from D'Almeida (1987), and dust radiative feedbacks (Perez et al., 2006a). The emission scheme implemented in BSC–DREAM8b, described in detail in Basart et al. (2012a), directly entrains dust–sized particles into the atmosphere and includes the influence of soil structure and particle size distribution, as well as of the atmospheric conditions – the near–surface wind speed must exceed the local threshold velocity to force dust mobilization.

2.2. Model application

In this study, the simulation domain covers a large area, including North Africa, Middle East and Europe, with the region of Portugal highlighted in blue (Figure 1a). The initial state of the dust concentration is defined by the 24–h forecast of the previous–day model run. Meteorological fields are initialized every 24 h, at previous–day 12 UTC, and boundary conditions updated every 6 h with the Global Forecast System (GFS) of NCEP. The resolution is set to 1/3° in the horizontal and to 24 layers extending up to approximately 15 km in the vertical, where the near–surface layer corresponds to approximately 86 meters AGL. Model outputs are processed in a 3–hour basis.

Several case studies have outlined the good skills of BSC–DREAM8b. In the last years, this model has been used for dust forecasting and as dust research tool in North Africa and Southern Europe (Jimenez–Guerrero et al., 2008; Amiridis et al., 2009; Klein et al., 2010; Pay et al., 2010; Alonso–Perez et al., 2011; Basart et al., 2012a; Kokkalis et al., 2012) and in the Cape Verde region (Gama et al., 2014). The model has also been evaluated and tested over longer time periods over Europe (Basart et al., 2012b; Pay et al., 2012; Tchepel et al., 2013) and against measurements at source regions (Todd et al., 2008; Haustein et al., 2009).

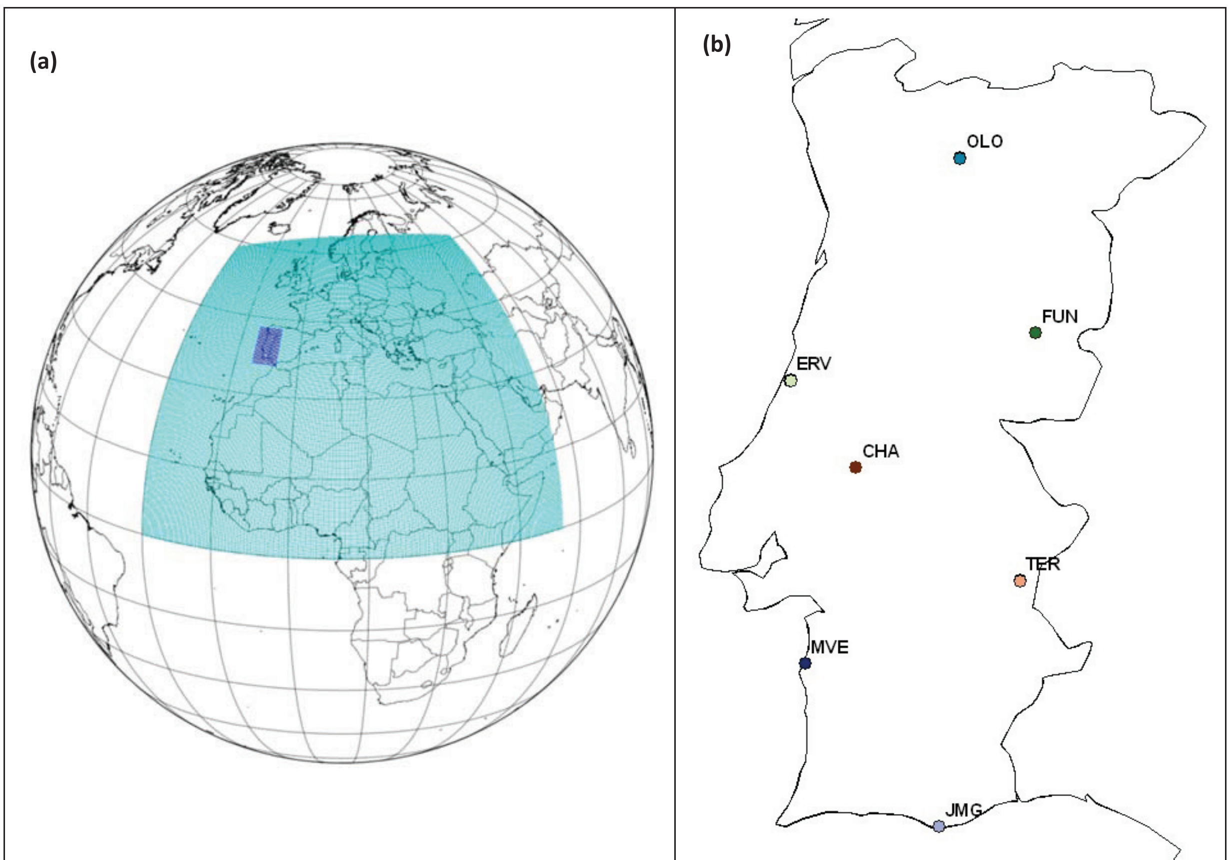


Figure 1. (a) *BSC–DREAM8b v1.0 model simulation domain, with the domain over Portugal highlighted [corresponding to a 396 grid cells (18x22)];* ***(b)*** *PM_{10} monitoring sites over Portugal.*

Furthermore, daily evaluation of BSC–DREAM8b with near–real–time observations is conducted at BSC–CNS (BSC–CNS, 2014). Currently, the daily operational model evaluation includes observations from satellites (MODIS and MSG) and AErosol RObotic NETwork (AERONET) sun photometers. The work presented here uses the BSC–DREAM8b surface concentration data estimated for Portugal to assess and characterize the mineral dust over one year period (2011), after a model validation exercise regarding this particular application over Portugal.

2.3. Model validation

In order to support the use of model results for the assessment of mineral dust over Portugal region, a validation exercise was performed using the comparison between the modeled mineral dust concentration with diameter below 10 µm (MD_{10}) and the observed particulate matter with aerodynamic diameter below 10 µm (PM_{10}). This was processed for 7 different locations, spatially distributed along mainland Portugal, and corresponding to background air quality monitoring sites where PM_{10} is measured continuously (Figure 1b). The daily mean time series comparison is shown in Figure 2.

As expected, Figure 2 shows that the PM_{10} observed concentrations are, in general, higher than the MD_{10} contribution from North Africa. In general, the peaks of dust concentrations coincide with PM_{10} peaks, which suggest a good performance of the BSC–DREAM8b model. Nevertheless, the model overestimates the dust concentrations in some episode cases, namely in April (e.g. in OLO, FUN, CHA). This overestimation in the spring period has been already identified and published. According to Basart et al. (2012a), the model shows an overestimation of the dust activity in Northern Algeria when compared to satellite estimates, affecting thus the dust transported towards the Western and Central Mediterranean mainly in spring.

A quantitative analysis was performed using statistical indicators, namely the Root Mean Square Error (RMSE) and the Mean Systematic Error (BIAS) which are measures of the unsystematic (random) and systematic errors obtained within the observed-predicted pairs of results, respectively (Borrego et al., 2008). The statistical results are shown in Table 1, regarding only the episodes period (simulated ground dust concentration > 5 µg m^{-3}), since in the other periods dust concentration is approximately zero.

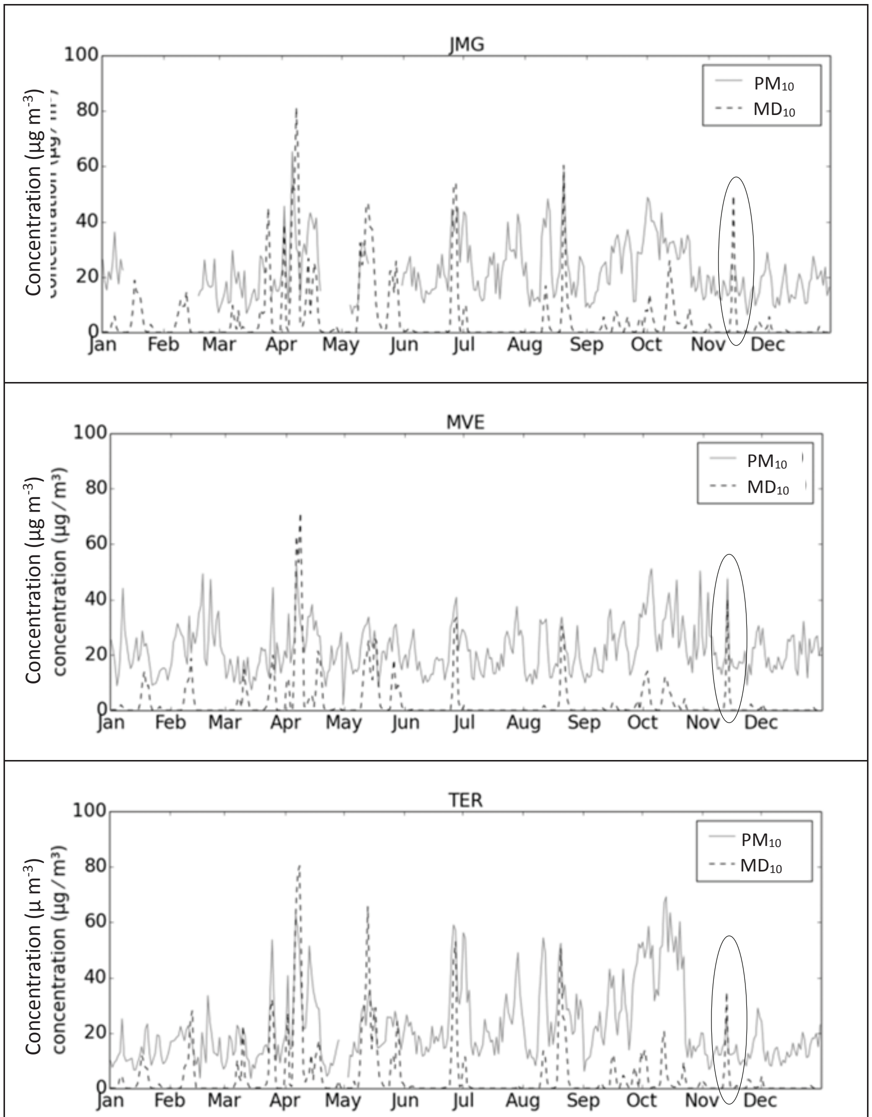


***Figure 2.** Daily mean time series of observed PM_{10} (solid gray line) and simulated MD_{10} (dashed black line) for the different sites.*

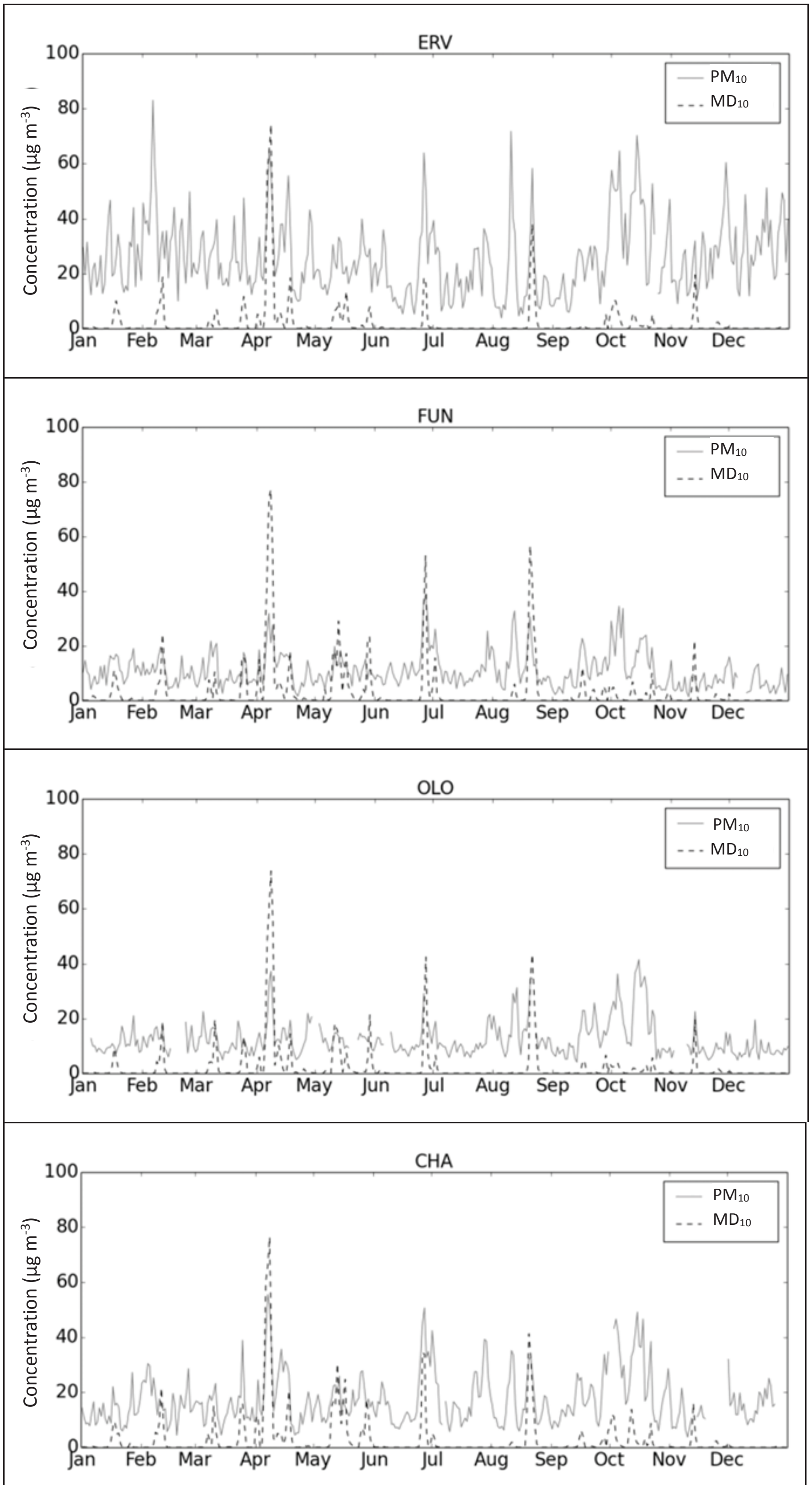


Figure 2. *Continued.*

Table 1. *Statistical indicators used for model validation (RMSE and BIAS)*

Monitoring Stations	Observed Mean (PM_{10})	Simulated Mean (MD_{10})	RMSE [a] ($\mu g\ m^{-3}$)	BIAS [b] ($\mu g\ m^{-3}$)
OLO	16.9	11.9	14.9	–4.8
CHA	25.7	14.8	16.5	–10.9
ERV	34.0	13.3	24.4	–23.7
FUN	17.1	16.0	14.4	–1.0
JMG	34.5	22.2	23.2	–12.4
MVE	28.3	14.8	18.4	–13.5
TER	35.3	22.8	22.1	–13.5
Average	27.45	16.02	19.1	–11.3

[a] $RMSE = \frac{1}{N}\sqrt{\sum_{i=1}^{N}(\mathrm{Model_i} - \mathrm{Obs_i})^2}$

[b] $BIAS = \frac{1}{N}\sum_{i=1}^{N}(\mathrm{Model_i} - \mathrm{Obs_i})$

On average, the deviation of the BSC–DREAM8b model results is below 20 μg m^{-3}, and the negative values of the BIAS indicate that, in general, model results (MD_{10}) are below PM_{10} observations. Since we are comparing just one fraction of the total PM_{10}, the magnitude of this error is low. During the dust episodes the values of PM_{10} can contain large fractions of other particulate matter content besides the mineral dust portion (MD_{10}).

3. Results

3.1. Assessment of mineral dust over Portugal

The mineral dust surface concentrations simulated with the BSC–DREAM8b v1.0 model over mainland Portugal during an entire year period (2011) were analyzed in terms of monthly averages and percentiles, in order to assess its spatial and temporal distribution. In this Section, not only the mineral dust fraction with diameter below 10 μm (MD_{10}) is analyzed, but the total mineral dust simulated by the model (MD), which includes particles with diameter up to 20 μm, as presented in Section 2.1. Figure 3 shows the BSC–DREAM8b v1.0 model results for the mineral dust monthly averages over the year 2011.

Figure 3 highlights that the mineral dust average values are significantly higher in April and May than the rest of the year. During these spring months mineral dust over Portugal has average contributions over 10 μg m^{-3} (10–18 μg m^{-3}), while during summer and winter seasons the mean values are below 6 μg m^{-3} (0–6 μg m^{-3}). April is the month when expected mean values are highest and December exhibits the lowest contribution. The spring events were already noticed in previous works, over the Iberian Peninsula and Western–Southern Europe (Laurent et al., 2008; Basart et al., 2012a; Gkikas et al., 2012; Pey et al., 2013). However, in the annual BSC–DREAM8b model evaluation shown in Basart et al. (2012a) it is highlighted that the model overestimates the Aerosol Optical Depth (AOD) in Morocco and North Algeria during spring, which may indicate that the strength of this source is being overestimated, thus influencing the modeled surface concentrations in the nearby areas such as the Iberian Peninsula.

The annual mean pattern, shown in Figure 4, reflects the several months where the influence of the Saharan mineral dust is not frequent. Only the southwest region of Spain exhibits a high annual average of mineral dust (>10 μg m^{-3}). Over the Portuguese territory, the magnitude of the annual average values varies between 2–6 μg m^{-3}.

Besides the monthly and annually characterization, the frequency of low, median and high values of mineral dust was also analyzed. Figure 5 presents the results obtained with the BSC–DREAM8b v1.0 annual simulation for various percentiles, in order to show about the spatial distribution of the different magnitude of mineral dust values.

Starting with the Percentile 25 (P25), and as it would be expectable, the low values of mineral dust over mainland Portugal are close to 0 μg m^{-3}. The analysis of P50 indicates that the influence of the transport of mineral dust over Portugal is limited on time scale, since in 50% of the time this contribution is below 0.2 μg m^{-3}. Only when high percentiles are analyzed (e.g. P75), the concentration of mineral dust over Portugal domain becomes relevant (≥2 μg m^{-3}). At Percentile 95 (P95), it is possible to observe the magnitude and location of the episodes of mineral dust which occurred in Portugal over 2011. These episodes are significantly stronger in the south–east region of the country exhibiting P95 values higher than 30 μg m^{-3}. In the rest of the Portuguese territory the peak contribution of mineral dust is around 10–25 μg m^{-3}.

3.2. Analysis of mineral dust episodes

Identification and selection of mineral dust episodes. In addition to the long–term assessment of the mineral dust over Portugal, this work aims to assign and analyze the days when a significant contribution of mineral dust is transported over Portugal domain. For that, a threshold above 5 μg m^{-3} (above the magnitude of the simulated annual average; see Figure 4) was considered to select this group of days. Simulated mineral dust data were extracted for the 7 locations of Figure 1b (monitoring sites used to analyze the model behavior) taking into account the above mentioned threshold.

Seventeen episodes were selected from the time series with mineral dust daily mean concentrations above 5 μg m^{-3} over all the analyzed sites. The duration of all episodes is more than one day (from 2 to 9 days period). Table 2 summarizes the selected episodes, the corresponding days and the maximum concentration of mineral dust (and where it was registered).

The selected episodes (concentrations of mineral dust higher than 5 μg m^{-3}) occurred at the same days in all analyzed sites, but with different magnitude. The episodes are distributed over the entire year, with predominance in April, May and June. The longest episode was registered in May (episode no. 8) and lasted for 9 days (10–18 May 2011). Besides this one, the duration of the majority of the episodes was about 2–6 days, which is consistent with the synoptic patterns lifetime (Cahynova and Huth, 2009).

The highest episode in terms of magnitude was simulated in April (5th–9th), when mineral dust surface concentrations reached around 60–80 μg m^{-3} over all the different locations. The southern sites (JMG, TER and MVE) show higher intensity on the peak episodes. The last episode identified – November 12th–14th – is a good example of the higher magnitude over the southern sites (pointed out with a black circle in Figure 2).

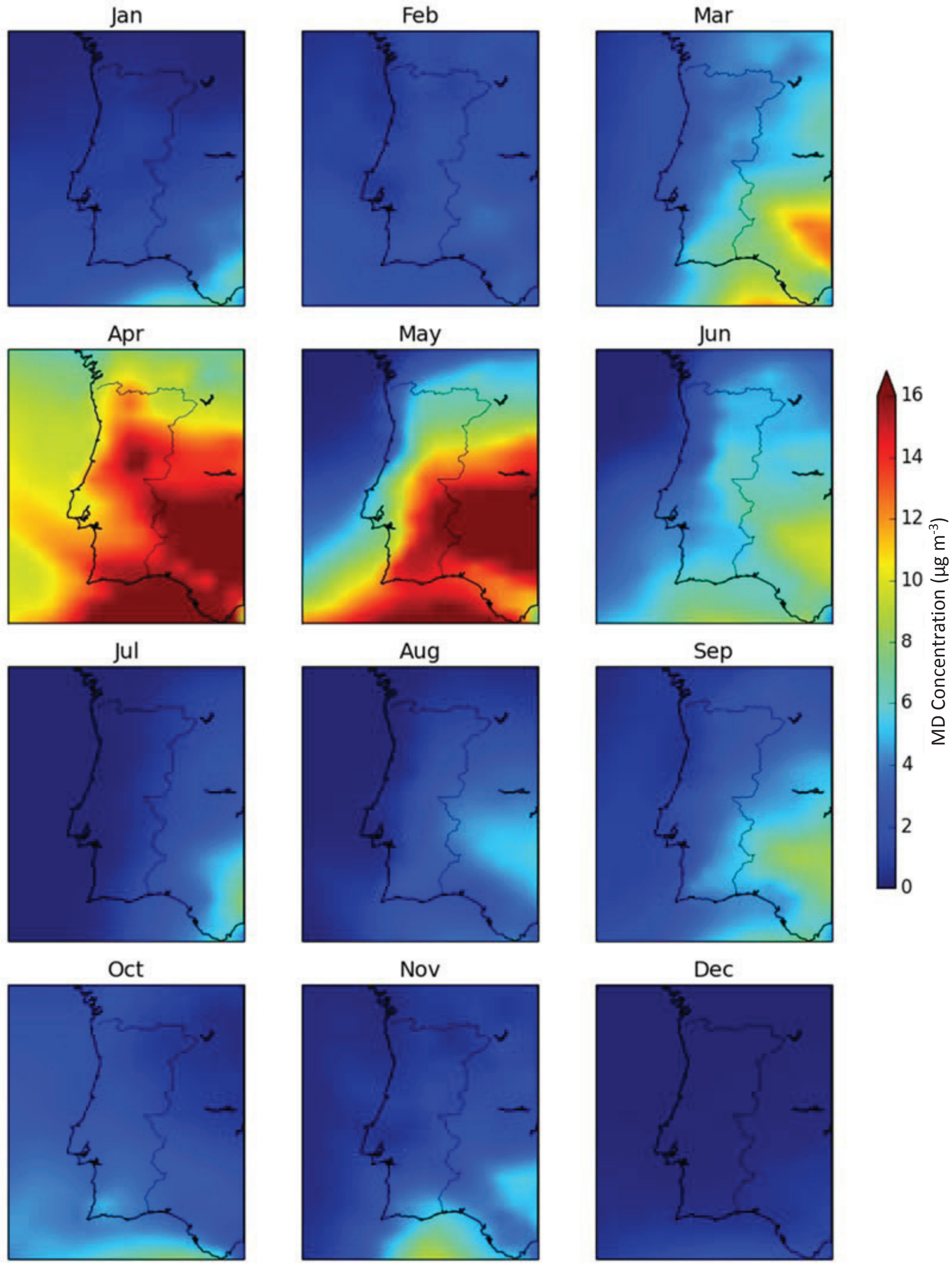


Figure 3. *Monthly mean of modeled mineral dust surface concentrations (μg m^{-3}) for 2011.*

Clustering analysis of the mineral dust episodes. In this section, the origin of the air parcels trajectories that arrive at each monitoring site during the selected episode days was studied. A cluster analysis of the obtained back trajectories was performed to reduce the number of ''origins of air masses'', appointing every calculated individual back trajectory to the most appropriate cluster, in order to identify and to better interpret mean synoptic situations related to the mineral dust episodes.

The back trajectories technique is a useful tool in tracing source regions of air pollution and determining transport patterns at receptor sites (Jorba et al., 2004). In this study, five–days (120 h) kinematic back trajectories arriving at 12 UTC at 1 000 m AGL at each of the study sites were estimated for the whole 2011. The air parcel trajectories were obtained with the version 4.8 of the HYbrid Single–Particle Lagrangian Integrated Trajectory model (HYSPLIT) developed by the National Oceanic and Atmospheric Administration (NOAA)'s Air Resources Laboratory (ARL) (Draxler and Hess, 1997; Draxler and Hess, 1998). Meteorological data from the NCEP Global Data Assimilation System – GDAS (Kanamitsu, 1989), with a horizontal resolution of 1.0° and a 3 hourly time resolution, was used to compute the trajectories. Even though the meteorological inputs for HYSPLIT and BSC–DREAM8b model is different, the predicted synoptic patterns are quite similar, at least for the periods studied in this paper.

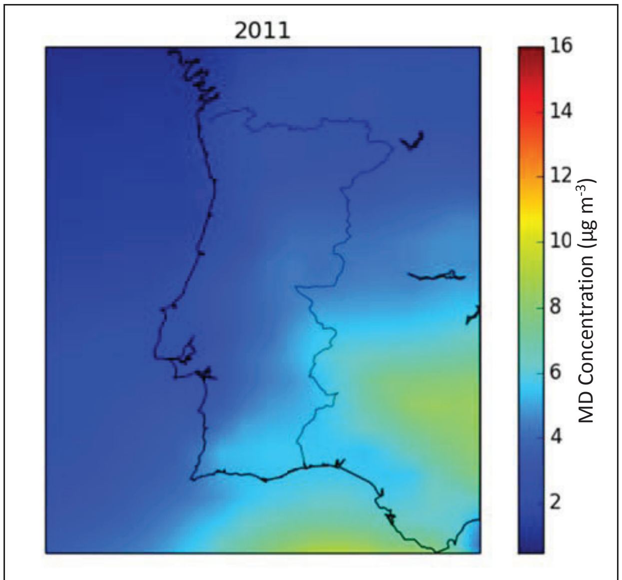


Figure 4. *Annual mean of modeled mineral dust surface concentration (µg m^{-3}) for 2011.*

A hierarchical clustering method was chosen to extract a natural number of clusters for the different individual back–trajectories, corresponding to the study day's episodes and to the whole year as well. The clustering iterations involved the calculation of a statistical measure, the cluster spatial variance (SPVAR, defined as the sum of the squared distances between the endpoints of the cluster's component trajectories and the mean of the trajectories in that cluster), for every possible combination of trajectory/cluster pairs. In each iteration, a pair of clusters was returned, chosen to be the one associated with the lowest increase in the total spatial variance (TSV, defined as the sum of all the SPVAR). Before the first clustering iteration, each trajectory was defined to be a cluster, e.g. there were N trajectories and N clusters, and TSV was equal to zero. After the first iteration, the number of clusters was N–1. The iterations continued until the last two clusters were combined, resulting in all the individual trajectories combined in the same cluster (Dorling et al., 1992).

In the first several clustering iterations the TSV increases greatly, then for much of the clustering it typically increases at a small, generally constant rate, but at some point it again increases rapidly, indicating that the clusters being combined are not very similar. This latter increase suggests where to stop the clustering method and it is clearly seen in a plot of the percentage of change in TSV vs. the number of clusters (not shown). Using the minimum optimum number of clusters as a selection criterion, different groups of clusters were identified for each specific site and are plotted in Figure 6, together with the respective mineral dust modeled statistics associated to each of the designed trajectories.

Between three and five major types of flow patterns are identified for the study period. These mean flow pathways (clusters) obtained for the different sites can easily be distinguished in terms of spatial, dynamic and temporal patterns.

The site of JMG (in the south coast of Portugal) is the one where the air masses trajectories coming from the North African coast are more pronounced (cluster 1 with a frequency of occurrence of 52% and cluster 2 with 35%, Figure 6) and are responsible for the highest modeled mineral dust concentrations (box plot, Figure 6). The site of TER, closer to JMG in the North direction, also exhibits a prevalence of the flow regimes coming from the African coast (cluster 1 and cluster 2 with 32% and 38% of frequency, respectively) but with less expressive contribution for modeled concentrations.

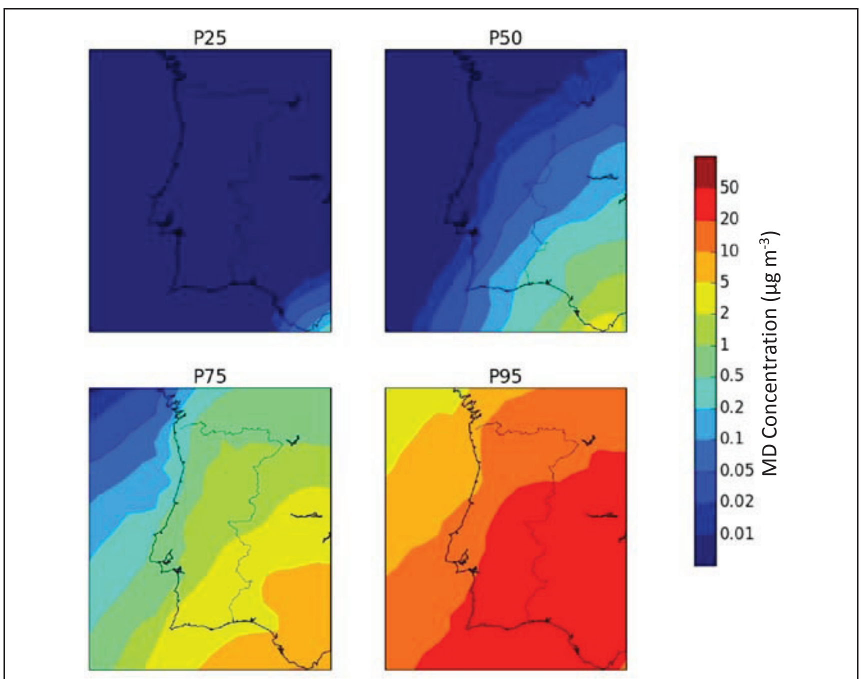


Figure 5. *Spatial representation of percentile 25 (P25), 50 (P50), 75 (P75) and 95 (P95) of the modeled mineral dust concentrations for 2011.*

MVE site, located in the south–west coast of Portugal, and FUN, sited in the interior central domain, also present a high predominance of the flow regimes coming from North Africa during the selected episode days (MVE – cluster 1: 49% and cluster 3: 25%; FUN – cluster 1: 72% and cluster 3: 17%), associated with higher modeled concentrations.

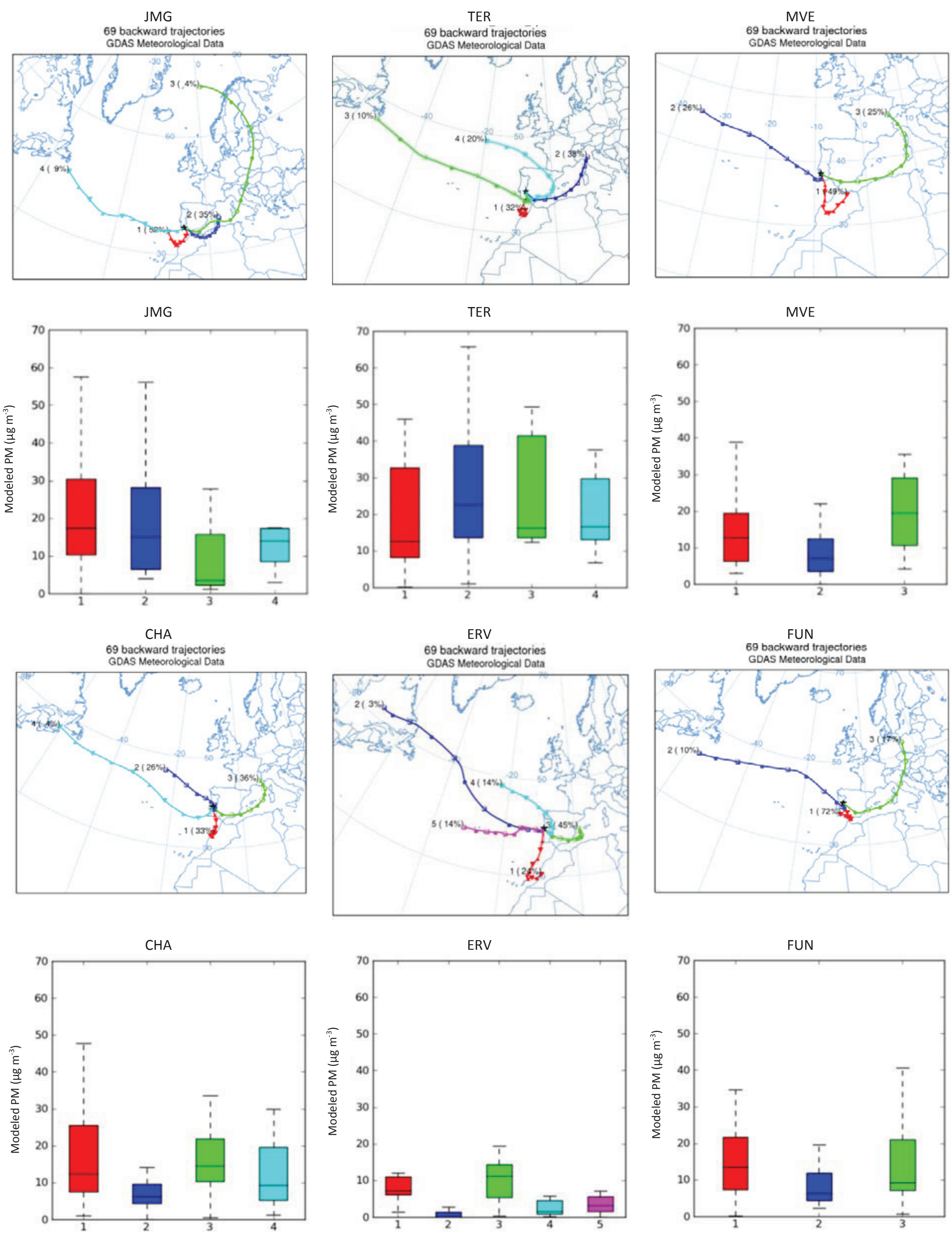


Figure 6. *Cluster mean trajectories identified for each study site (JMG, MVE, TER, CHA, ERV, FUN and OLO) during the selected episodes days and modeled mineral dust concentration statistics associated to each of the identified trajectories (same color representation).*

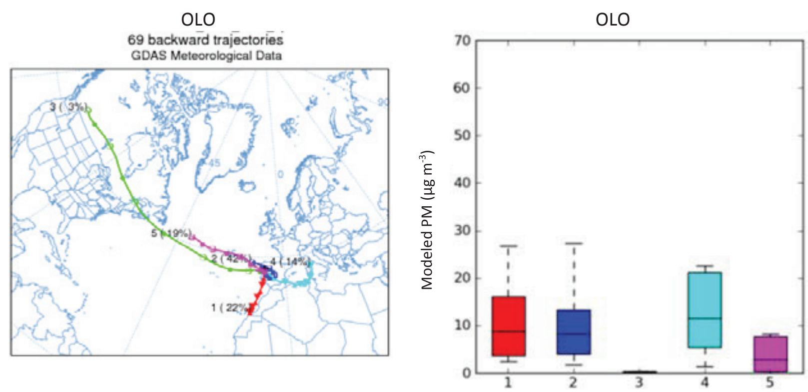


Figure 6. *Continued.*

Table 2. *Identification and characterization of the selected episode days*

Episode No.	Months	Days	Duration (No. of Days)	Maximum Concentration of MD_{10} ($\mu g\ m^{-3}$) and Location	
1	January	17–19	3	18.9	JMG
2	February	08–12	5	28.1	TER
3	March	10–11	2	22.8	TER
4	March	24–26	3	44.9	JMG
5	April	01–02	2	39.1	JMG
6	April	05–09	5	81.1	JMG
7	April	12–19	8	26.8	JMG
8	May	10–18	9	65.5	TER
9	May	28–29	2	25.8	JMG
10	June	25–28	4	53.7	JMG
11	July	01–02	2	15.6	FUN
12	August	19–23	5	60.2	JMG
13	September	15–17	3	12.2	TER
14	September/October	30/01–03	4	14.1	MVE
15	October	11–13	3	25.9	JMG
16	October	21–22	2	9.9	FUN
17	November	12–14	3	49.4	JMG

The three last sites – CHA, ERV and OLO – located in the central and northern regions of Portugal, present a lower influence of the air flow trajectories coming from North Africa (cluster 1 with 33%, 24% and 22% of frequency; respectively) associated with lower mineral dust concentration values (Figure 6). The lowest influence of the North Africa flow regimes and thus lower associated PM concentrations were found at OLO site, which can be justified mainly due its more far/remote location in the North of Portugal. Nevertheless, even in this distant place, during the selected episode days, a contribution of 22% of the air masses regime (cluster 1) was estimated and concentrations of 5–25 $\mu g\ m^{-3}$ of mineral dust (associated to these air masses trajectories) were predicted by the model.

In order to analyze the representativeness and relevance of the selected episode days regarding the entire year of 2011, Figure 7 presents the cluster annual mean trajectories for the whole 2011, for the different study sites.

There are two distinct groups of sites regarding the relevance of the air flow patterns coming with influence from North Africa. The first group includes the sites of JMG, TER and MVE, located in the south and south–west coasts of Portugal, where the frequency of occurrence of these southern patterns is about 8–16% of the total air masses trajectories occurred during the entire year. This percentage is, however, not significantly high, when compared to the dominant flow patterns from the Atlantic Northwest corridor.

All the other study sites – FUN, CHA, ERV and OLO – located in the central and north part of Portugal show a cluster pattern with lower southern influence, which is more evident for OLO site. This indicates that in these particular sites the prevailing air flow patterns have other origins, predominantly from the Atlantic northwest direction, but also from the northeast inner part of the Peninsula. This Atlantic Northwest dominant pattern is even more evident for OLO site, where the entire mean clusters have this direction.

4. Conclusions

This work focused on the study of the mineral dust transported from the North African deserts to Portugal, in terms of long–term assessment and also high episode peaks. The application of BSC–DREAM8b v1.0 model (already validated in previous studies) to a domain which include North Africa and Middle East source regions, for the entire year of 2011, allows the characterization of the magnitude and spatial distribution of the mineral dust over Portugal. The annual mean pattern of the simulated dust has a magnitude of 2–6 $\mu g\ m^{-3}$, where the monthly average highlights the largest mineral dust values in April and May (in average 4 $\mu g\ m^{-3}$ higher than in the rest of the months). The influence of the transport of mineral dust to Portugal is limited on time scale, since in 50% of the time (percentile 50) this contribution is below 0.2 µg m^{-3}. Only when high percentiles are analyzed the concentration of mineral dust over Portugal's domain becomes relevant (>3 µg m^{-3}; with peak contribution around 10–25 µg m^{-3}).

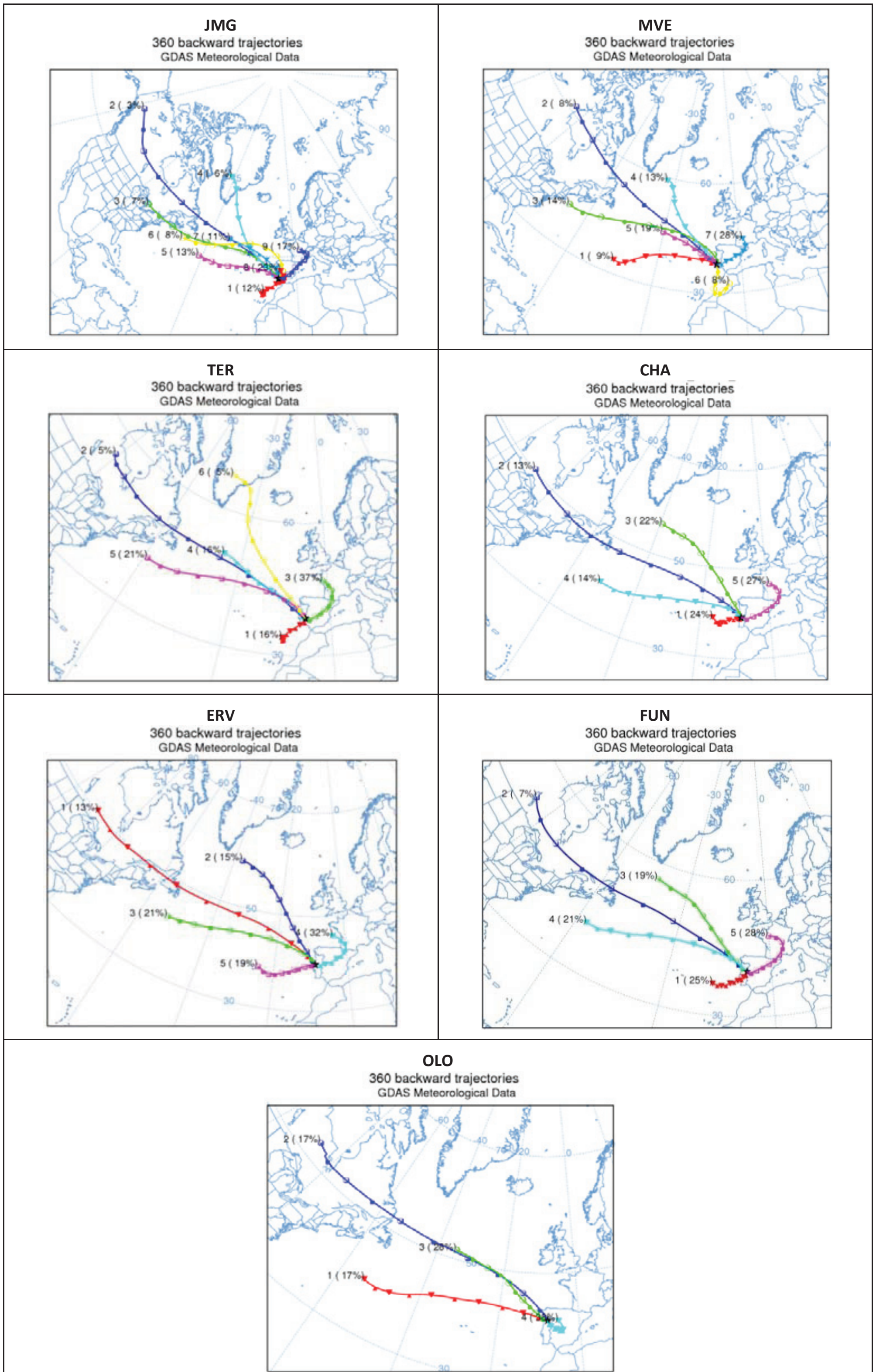


Figure 7. *Cluster mean trajectories obtained for the entire year of 2011, for the different sites (JMG, MVE, TER, CHA, ERV, FUN and OLO).*

In order to characterize the highest episodes of dust occurred in Portugal, a group of episode days with dust daily concentration above 5 $\mu g\ m^{-3}$ was selected, and data were extracted for 7 different locations, spatially distributed along the country (and coincident with monitoring sites). A cluster analysis of the air parcels back trajectories that arrive at each site was performed in order to identify the mean flow patterns associated to each mineral dust episode. At the south study sites, during the episode days, the air masses trajectories coming from the North African coast are predominant (with frequency above 70%). The prevalence of the flow regimes coming from North Africa during the episode days decrease for the upper latitude sites: the two central region sites still exhibit a frequency above 50%, but this frequency decrease to below 33% for the two sites located in the north region.

Acknowledgments

This work was supported by the Portuguese 'Ministerio da Ciencia, da Tecnologia e do Ensino Superior' through the Post–Doctoral fellowship of Alexandra Monteiro (SFRH/BPD/63796/2009) and the PhD fellowships of Ana Patricia Fernandes (SFRH/BD/86307/2012) and Carla Gama (SFRH/BD/87468/2012). The authors gratefully acknowledge the Barcelona Supercomputing Center – Centro Nacional de Supercomputacion (BSC–CNS) for the provision and support with the BSC–DREAM8b dust model, and NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and the READY website (http://www.arl.noaa.gov/ready.html) from which the meteorological data used to force HYSPLIT has been downloaded.

References

Alonso–Perez, S., Cuevas, E., Perez, C., Querol, X., Baldasano, J.M., Draxler, R., De Bustos, J.J., 2011. Trend changes of African airmass intrusions in the marine boundary layer over the subtropical Eastern North Atlantic region in winter. *Tellus Series B–Chemical and Physical Meteorology* 63, 255–265.

Amiridis, V., Kafatos, M., Perez, C., Kazadzis, S., Gerasopoulos, E., Mamouri, R.E., Papayannis, A., Kokkalis, P., Giannakaki, E., Basart, S., Daglis, I., Zerefos, C., 2009. The potential of the synergistic use of passive and active remote sensing measurements for the validation of a regional dust model. *Annales Geophysicae* 27, 3155–3164.

Ansmann, A., Bosenberg, J., Chaikovsky, A., Comeron, A., Eckhardt, S., Eixmann, R., Freudenthaler, V., Ginoux, P., Komguem, L., Linne, H., Marquez, M.A.L., Matthias, V., Mattis, I., Mitev, V., Muller, D., Music, S., Nickovic, S., Pelon, J., Sauvage, L., Sobolewsky, P., Srivastava, M.K., Stohl, A., Torres, O., Vaughan, G., Wandinger, U., Wiegner, M., 2003. Long–range transport of Saharan dust to Northern Europe: The 11–16 October 2001 outbreak observed with EARLINET. *Journal of Geophysical Research–Atmospheres* 108, art. no. 4783.

Basart, S., Perez, C., Nickovic, S., Cuevas, E., Baldasano, J.M., 2012a. Development and evaluation of the BSC–DREAM8b dust regional model over Northern Africa, the Mediterranean and the Middle East. *Tellus Series B–Chemical and Physical Meteorology* 64, art. no. 18539.

Basart, S., Pay, M.T., Jorba, O., Perez, C., Jimenez–Guerrero, P., Schulz, M., Baldasano, J.M., 2012b. Aerosols in the CALIOPE air quality modelling system: Evaluation and analysis of PM levels, optical depths and chemical composition over Europe. *Atmospheric Chemistry and Physics* 12, 3363–3392.

Black, T.L., 1994. The new NMC mesoscale Eta–model – Description and forecast examples. *Weather and Forecasting* 9, 265–278.

Borge, R., Lumbreras, J., Vardoulakis, S., Kassomenos, P., Rodriguez, E., 2007. Analysis of long–range transport influences on urban PM_{10} using two–stage atmospheric trajectory clusters. *Atmospheric Environment* 41, 4434–4450.

Borrego, C., Monteiro, A., Ferreira, J., Miranda, A.I., Costa, A.M., Carvalho, A.C., Lopes, M., 2008. Procedures for estimation of modelling uncertainty in air quality assessment. *Environment International* 34, 613–620.

BSC–CNS (Barcelona Supercomputing Center–Centro Nacional Supercomputacion), 2014. http://www.bsc.es/earth–sciences/mineral–dust–forecast–system/bsc–dream8b–forecast, accessed in 2014.

Cahynova, M., Huth, R., 2009. Enhanced lifetime of atmospheric circulation types over Europe: Fact or fiction? *Tellus Series A–Dynamic Meteorology and Oceanography* 61, 407–416.

D'Almeida, G.A., 1987. On the variability of desert aerosol radiative characteristics. *Journal of Geophysical Research–Atmospheres* 92, 3017–3026.

Dorling, S.R., Davies, T.D., Pierce, C.E., 1992. Cluster–analysis – A technique for estimating the synoptic meteorological controls on air and precipitation chemistry – Method and applications. *Atmospheric Environment Part A–General Topics* 26, 2575–2581.

Draxler, R.R., Hess, G.D., 1998. An overview of the HYSPLIT_4 modelling system for trajectories, dispersion and deposition. *Australian Meteorological Magazine* 47, 295–308.

Draxler, R.R., Hess, G.D., 1997. Description of the HYSPLIT 4 Modeling System, NOAA Technical Memorandum ERL ARL–224, 27 pages.

EC (European Commission), 2011. Commission Staff Working Paper, Establishing for Demonstration and Subtraction of Exceedances Attributable to Natural Sources under the Directive 2008/50/EC on Ambient Air Quality and Cleaner Air for Europe, http://ec.europa.eu/environment/air/quality/legislation/pdf/sec_2011_0208.pdf, accessed in August 2014.

Escudero, M., Querol, X., Avila, A., Cuevas, E., 2007a. Origin of the exceedances of the European daily PM limit value in regional background areas of Spain. *Atmospheric Environment* 41, 730–744.

Escudero, M., Querol, X., Pey, J., Alastuey, A., Perez, N., Ferreira, F., Alonso, S., Rodriguez, S., Cuevas, E., 2007b. A methodology for the quantification of the net African dust load in air quality monitoring networks. *Atmospheric Environment* 41, 5516–5524.

Fialho, P., Freitas, M.C., Barata, F., Vieira, B., Hansen, A.D.A., Honrath, R.E., 2006. The Aethalometer calibration and determination of iron concentration in dust aerosols. *Journal of Aerosol Science* 37, 1497–1506.

Freitas, M.D.C., Dionisloa, I., Fialho, P., Barata, F., 2007. Aerosol chemical elemental mass concentration at lower free troposphere. *Nuclear Instruments & Methods in Physics Research Section A–Accelerators Spectrometers Detectors and Associated Equipment* 579, 507–509.

Gama, C., Tchepel, O., Baldasano, J.M., Basart, S., Ferreira, J., Pio, C., Cardoso, J., Borrego, C., 2014. Seasonal patterns of Saharan dust over Cape Verde – a combined approach using observations and modelling. *Submitted to Tellus B*.

Gkikas, A., Houssos, E.E., Hatzianastassiou, N., Papadimas, C.D., Bartzokas, A., 2012. Synoptic conditions favouring the occurrence of aerosol episodes over the broader Mediterranean basin. *Quarterly Journal of the Royal Meteorological Society* 138, 932–949.

Haustein, K., Perez, C., Baldasano, J.M., Muller, D., Tesche, M., Schladitz, A., Esselborn, M., Weinzierl, B., Kandler, K., von Hoyningen–Huene, W., 2009. Regional dust model performance during SAMUM 2006. *Geophysical Research Letters* 36, art. no. L03812.

IES–JRC (Institute for Environment and Sustainability – Joint Research Centre), 2007. Contribution of Natural Sources to Air Pollution Levels in the EU – A Technical Basis for the Development of Guidance for the Member States, 102 pages.

IPCC (Intergovernmental Panel on Climate Change), 2007. Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K.B., Tignor M., Miller, H.L., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 996 pages.

IPCC (Intergovernmental Panel on Climate Change), 2001. Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, edited by Houghton, J.T., Ding, Y., Griggs, D.J., Noguer, M., van der

Linden, P.J., Dai, X., Maskell, K., Johnson, C.A., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 881 pages.

Jimenez–Guerrero, P., Perez, C., Jorba, O., Baldasano, J.M., 2008. Contribution of Saharan dust in an integrated air quality system and its on–line assessment. *Geophysical Research Letters* 35, art. no. L03814.

Jorba, O., Perez, C., Rocadenbosch, F., Baldasano, J.M., 2004. Cluster analysis of 4–day back trajectories arriving in the Barcelona area, Spain, from 1997 to 2002. *Journal of Applied Meteorology* 43, 887–901.

Kanamitsu, M., 1989. Description of the NMC global data assimilation and forecast system. *Weather and Forecasting* 4, 335–342.

Klein, H., Nickovic, S., Haunold, W., Bundke, U., Nillius, B., Ebert, M., Weinbruch, S., Schuetz, L., Levin, Z., Barrie, L.A., Bingemer, H., 2010. Saharan dust and ice nuclei over Central Europe. *Atmospheric Chemistry and Physics* 10, 10211–10221.

Kokkalis, P., Mamouri, R.E., Todua, M., Didebulidze, G.G., Papayannis, A., Amiridis, V., Basart, S., Perez, C., Baldasano, J.M., 2012. Ground–, satellite– and simulation–based analysis of a strong dust event over Abastumani, Georgia, during May 2009. *International Journal of Remote Sensing* 33, 4886–4901.

Laurent, B., Marticorena, B., Bergametti, G., Leon, J.F., Mahowald, N.M., 2008. Modeling mineral dust emissions from the Sahara Desert using new surface properties and soil database. *Journal of Geophysical Research–Atmospheres* 113, art. no. D14218.

Niemi, J.V., Saarikoski, S., Aurela, M., Tervahattu, H., Hillamo, R., Westphal, D.L., Aarnio, P., Koskentalo, T., Makkonen, U., Vehkamaki, H., Kulmala, M., 2009. Long–range transport episodes of fine particles in southern Finland during 1999–2007. *Atmospheric Environment* 43, 1255–1264.

Pay, M.T., Jimenez–Guerrero, P., Jorba, O., Basart, S., Querol, X., Pandolfi, M., Baldasano, J.M., 2012. Spatio–temporal variability of concentrations and speciation of particulate matter across Spain in the CALIOPE modeling system. *Atmospheric Environment* 46, 376–396.

Pay, M.T., Piot, M., Jorba, O., Gasso, S., Goncalves, M., Basart, S., Dabdub, D., Jimenez–Guerrero, P., Baldasano, J.M., 2010. A full year evaluation of the CALIOPE–EU air quality modeling system over Europe for 2004. *Atmospheric Environment* 44, 3322–3342.

Perez, C., Nickovic, S., Pejanovic, G., Baldasano, J.M., Ozsoy, E., 2006a. Interactive dust–radiation modeling: A step to improve weather forecasts. *Journal of Geophysical Research–Atmospheres* 111, art. no. D16206.

Perez, C., Nickovic, S., Baldasano, J.M., Sicard, M., Rocadenbosch, F., Cachorro, V.E., 2006b. A long Saharan dust event over the western Mediterranean: Lidar, sun photometer observations, and regional dust modeling. *Journal of Geophysical Research–Atmospheres* 111, art. no. D15214.

Pey, J., Querol, X., Alastuey, A., Forastiere, F., Stafoggia, M., 2013. African dust outbreaks over the Mediterranean Basin during 2001–2011: PM_{10} concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology. *Atmospheric Chemistry and Physics* 13, 1395–1410.

Pope, C.A., Dockery, D.W., 2006. Health effects of fine particulate air pollution: Lines that connect. *Journal of the Air & Waste Management Association* 56, 709–742.

Prospero, J.M., 1999. Long–range transport of mineral dust in the global atmosphere: Impact of African dust on the environment of the Southeastern United States. *Proceedings of the National Academy of Sciences of the United States of America* 96, 3396–3403.

Reis, M.A., Oliveira, O.R., Alves, L.C., Rita, E.M.C., Rodrigues, F., Fialho, P., Pio, C.A., Freitas, M.C., 2002. Comparison of continental Portugal and Azores Islands aerosol during a Sahara dust storm. *Nuclear Instruments & Methods in Physics Research Section B–Beam Interactions with Materials and Atoms* 189, 272–278.

Rodriguez, S., Querol, X., Alastuey, A., Kallos, G., Kakaliagou, O., 2001. Saharan dust contributions to PM_{10} and TSP levels in Southern and Eastern Spain. *Atmospheric Environment* 35, 2433–2447.

SDS–WAS (Sand and Dust Storms – Warning Advisory and Assessment System), 2014. http://sds–was.aemet.es/, accessed in August 2014.

Tchepel, O., Ferreira, J., Fernandes, A.P., Basart, S., Baldasano, J.M., Borrego, C., 2013. Analysis of long–range transport of aerosols for Portugal using 3D chemical transport model and satellite measurements. *Atmospheric Environment* 64, 229–241.

Tegen, I., Lacis, A.A., 1996. Modeling of particle size distribution and its influence on the radiative properties of mineral dust aerosol. *Journal of Geophysical Research–Atmospheres* 101, 19237–19244.

Todd, M.C., Karam, D.B., Cavazos, C., Bouet, C., Heinold, B., Baldasano, J.M., Cautenet, G., Koren, I., Perez, C., Solmon, F., Tegen, I., Tulet, P., Washington, R., Zakey, A., 2008. Quantifying uncertainty in estimates of mineral dust flux: An intercomparison of model performance over the Bodele Depression, northern Chad. *Journal of Geophysical Research–Atmospheres* 113, art. no. D24107.

Wagner, F., Bortoli, D., Pereira, S., Costa, M.J., Silva, A.M., Weinzierl, B., Esselborn, M., Petzold, A., Rasp, K., Heinold, B., Tegen, I., 2009. Properties of dust aerosol particles transported to Portugal from the Sahara Desert. *Tellus Series B–Chemical and Physical Meteorology* 61, 297–306.

WHO (World Health Organization), 2006a. Air Quality Guidelines – Global update 2005, WHO Air Quality Guidelines for Particulate Matter, Ozone, Nitrogen Dioxide and Sulfur Dioxide, http://www.who.int/phe/health_topics/outdoorair/outdoorair_aqg/en/, accessed in August 2014.

WHO (World Health Organization), 2006b. Health Risks of Particulate Matter from Long–Range Transboundary Air Pollution, http://www.euro.who.int/_data/assets/pdf_file/0006/78657/E88189.pdf, accessed in August 2014.