Author's response to the reviews of Manuscript No: acp-2014-30 "African dust outbreaks over the western Mediterranean basin: 11-year characterization of atmospheric circulation patterns and dust source areas" by P. Salvador, S. Alonso-Pérez, J. Pey, B. Artíñano, J.J. de Bustos, A. Alastuey and X. Querol.

Dear editor,

First of all we wished to thank Referee #1 and Referee #2 for the very valuable comments and suggestions on the first version of our manuscript. They helped us to write a new version with higher quality.

Next we included a point-by-point response to the reviews and comments and a list of all relevant changes made in the manuscript.

Reply to Anonymous Referee #1:

Reply to general comments:

- 1. We would like to clarify some statements expressed by Referee #1 in the first paragraph of his review.
- a) The analysis was not performed using time series of PM10 data. We used time series of estimations of African dust contribution to daily PM10 levels. The difference between both parameters must be highlighted because the use of dust contributions assures that the intensity of the African dust outbreak is being considered. Weak African dust outbreaks can contribute with a low dust load to the daily PM10 bulk levels at a specific site but it does not assure that the daily PM10 levels would be low.
- b) It must be noted that we considered more accurate to define the western Mediterranean basin as the area of study, than the Iberian Peninsula or simply Spain. In spite of the fact that data from Portuguese air quality monitoring stations were not included for the reason exposed in page 5501, lines 8-10, most of the regions of the Iberian Peninsula have been analyzed. As we also included data from stations within the Balearic Islands we extended the study area further along the Iberian Peninsula. Besides, the Canary Islands were not included in this study for some reasons that will be discussed later. Consequently it would not be strictly correct to talk about the influence of the African dust outbreaks over Spain.
- c) Finally, although perhaps the reviewer had interpreted, we cannot say that "the impact index was higher in spring". This index was not computed on a seasonal basis.

2. Referee #1 declared that our results were not original and suggested building up a story from our previous work.

In spite of the fact that the characterization of synoptic meteorological situations leading to African dust outbreaks were addressed in previous works for some authors of this work, among others, it should be stressed that they were produced over specific areas of the Iberian Peninsula with different methodologies, during different time periods. As it was noted in page 5508 lines 14-16 and page 5509 lines 4-5, two main circulation types were obtained in these works. Namely, the mode 4 (or CT-4) that was identified in our paper, which is the responsible of causing the most frequent and intense African dust outbreaks and other mode which incorporates features of the modes 1 and 2 that we identified in our paper.

Our aim was to complement and improve these studies, by analyzing all the events produced over the whole area of study during a longer time period and then classify the main circulation types which are responsible of them by applying a systematic and objective methodology. Then, the impact of each circulation type over each specific area would be estimated.

In page 110 of the Alonso-Perez et al. (2011a) paper the same inference was used to justify its characterization of meteorological scenarios, favoring high African dust concentrations in the subtropical eastern north Atlantic region.

One of the main results of this work is the demonstration that the circulation types identified produced different degrees of impact, in terms of frequency of occurrence of events and African dust contribution to PM10 levels, over the different regions analyzed. For this reason this study must be considered as necessary and original.

3. In relation with the manuscript of Alonso-Perez et al. (2011a) it was not considered because it was exclusively dedicated to the African dust outbreaks influencing the Canary Islands. The phenomenology of these events is different in this case in comparison with the western Mediterranean basin, with clear differences in their seasonal trends and the associated meteorological patterns (Viana et al., 2002; Alonso-Perez et al., 2011a). You suggested that "It would be valuable to know if the present results differ from Alonso-Perez et al., (2001, Meteorol. Atmos. Phys., 113, 109-124)". As expected, the circulation types obtained in our work strongly differed from those determined by these authors, specially the so-called "return African mass episodes" which are produced in winter-spring months over the subtropical eastern north Atlantic region.

However, we agree with the reviewer that similar methodologies and some common objectives exist between both papers, so that we thank the reviewer to notice this and provide the opportunity to improve our work with additional comments based on the comparison of both works that were included in the revised version of the paper.

4. For our study, the selection of the classification method (K-means) was based in four main criteria:

1) cluster separability ability, 2) ability to reproduce known circulation patterns, 3) computational needs, and 4) physical meaning of the results. When we talk about "physical explainability" we are referring to circulation patterns that were detected during all the years of the period of study, with a common seasonal trend and with influence over the same geographical areas.

Gong and Richman (1995) and Huth (1996) demonstrated that there is not a classification method which could be considered as the best in all aspects among other tested. Methodological deficiencies can be found in any classification method and some subjective decisions must be taken when any of them are applied (Huth et al., 2008).

Principal Components Analysis (PCs) has been widely used for circulation patterns classification although, as all computer-assisted classification method, they have some weaknesses. First of all, as explained in Huth et al. (2008), PCs in S-mode (grid points being in columns and cases in rows) are widely used for atmospheric pattern classification. However, S-mode actually detects modes of variability, which are the elements that through a linear combination, form circulation patterns. So, it is a common error to identify circulation patterns as the first solutions of the S-mode PCs. The correct procedure is to use PCs in T-mode (grid point values in rows and cases in columns), but this method also has a major drawback: the physical meaning of the resulting solutions can be troublesome, and usually a rotation of the principal components is necessary to deal with this issue. Rotations add extra computational time and power needs. In addition, rotated PC solutions have overlapping solutions, i.e., the variables do not necessarily belong to any component, or some variables could belong to more than one component (Gong and Richman, 1995). Moreover, PCs may have two phases, positive and negative (this is more common in the case of using anomalies data; Huth, 1996), and only one of them should be taken to interpret results.

As it can be seen in Table 2 of Huth et al. (2008) and in Huth (1996), K-means provides very good separability (poor in the case of PCs), good temporal and spatial stability (not worse than for PCs), and a moderate ability to reproduce known underlying structure of data, so we chose to use this nonhierarchical algorithm with our data. After comparing several classification methods (including K-means and Principal Components), Huth (1996) stated that "if the preferred property is the separation (among clusters as well as between clusters and the whole data set), the K-means method is best."

For these reasons we preferred to use the K-means methodology which is less complex in the mathematical discrimination of different patterns than other methods, but allowed us obtaining a reasonably number of patterns which could be physically interpreted. Alonso-Perez et al. (2011a) used the same reasoning when they decided not to apply rotations to the PCA solutions because unrotated solutions resulted in patterns with an easy physical meaning.

Finally we would like to emphasize that from the Alonso-Perez et al. (2011a) work it could not be interpreted that the K-means method provides erroneous results in comparison with Principal Components Analysis, as Reviewer#1 alleged. In page 112 of this paper it is clearly declared that "Among all possible methods, K-means is the most suitable partitional clustering technique for dealing with a great number of patterns in a high dimensional space, which is a habitual case in

climatology". Alonso-Perez et al. (2011a) also stated that K-means and PCs resulted to be complementary methods, so the use of both methods was useful Moreover, these authors confirmed in page 113 that "In most cases, the two leading PCs patterns are very similar to the two leading patterns obtained with K-means method". Giving the successful use of both methods, there was no clear reason presented in this study to choose one over the other.

With the aim to validate our results we have carried out a Principal Component Analysis in T-mode with the same 850 hPa geopotential height data base used in our paper. 4 PCs were retained to be compared to the 4 clusters obtained with the K-means procedure. Different rotations (Oblimin as in Huth, (1996) but also Varimax) were checked and the 4 final composite maps were calculated by averaging 850 hPa geopotential height using the fields corresponding to the days assigned to each PC.

5. Regarding your comment about the discussion of the effects of NAO on dust inter-annual variability in Alonso-Perez et al. (2011a), it was concluded that the findings of other authors regarding this relationship were in agreement with the finding of a high-pressure system over Southern Europe and North Africa in their three first clusters in January and July. A study on the correlation of NAO and dust inter-annual variability in the Subtropical Eastern North Atlantic Region was done subsequently by Alonso-Perez et al. (2011b) but, unlike our current study, no links with synoptic patterns were done.

It must be said that we included a discussion in pages 5510-5511 on the relationship between the occurrence of African dust episodic days for the difference circulation types and the values of the NAO index. This relationship was especially outstanding for circulation types 2 and 3.

6. In relation with this discussion we completely disaccord with your statement that, "circulation patterns of mode 2 and 3 in Figure 2 appeared very similar".

A visual inspection of Figure 2 may lead to wrong conclusions. For instance, mode 2 and mode 3 were characterized by the North-African high located over western Lybia. However the trough placed over the western Iberian Peninsula in mode 2 was not detected in mode 3.

In spite of the fact that both modes were more frequently produced in spring, there are many outstanding differences between them that have been showed in the manuscript (Figures 4 and 5)

Figure 4 revealed an opposite behavior of the annual occurrence of both modes in relation with the annual NAO index. Figure 5 illustrated these differences. In spring 2011, a period characterized by the advection of Atlantic air masses at latitudes higher than 45°N (higher than normal NAO index values) the occurrence of African dust episodic days was dominated by mode 2. In these days African air masses moved northward towards the Iberian Peninsula helped by the Atlantic low pressure system and the North-African high. In spring 2005, a period during which the advection of Atlantic air masses was produced at lower latitudes than usual (lower than normal NAO index values), African air masses moved northeastward towards the eastern side of the

Iberian Peninsula and the Balearic Islands mostly during episodic days dominated by mode 3. As a consequence, different regions of the western Mediterranean basin were influenced by the African dust under both modes (Figure 8).

7. It is also suggested by the reviewer to remark similarities and/or differences between the results of this study and those obtained by Pey et al. (2013). In page 5499, lines 22-25 we clearly expressed that they used part of our data set, among others, to characterize the African dust outbreaks over the western, central and eastern Mediterranean basin, with different purposes than ours. Consequently it was not easy to find similarities and/or differences between both studies. An additional effort will be done to find links between our results and the comments on the atmospheric patterns which governed the 2001-2011 summer periods showed in the Pey et al. (2013) work.

Reply to more detailed comments:

Abstract:

We agree with Referee #1 that the description of the methodology in the abstract could be shortened. The Abstract will be rewritten emphasizing in the key findings, in the new version of the manuscript.

1. In relation with the statement that we were "unable to identify sources of dust beyond the entire northern Africa", it should be noted that Prospero et al. (2002) demonstrated that most regions of the African continent at latitudes higher than 10°N were active sources of dust. Ginoux et al. (2001; 2012) also identified dust source regions all over Northern Africa. This fact is well illustrated in the Figure 7 in page 14 of the Ginoux et al. (2012) paper.

Figure 7 in our paper showed that most of the potential sources areas of dust identified in the RCF agreed with maxima in the dust source function S (Ginoux et al., 2001). However, some other outstanding maxima in the geographic distribution of the function S in northern Africa, such as those representing the Bodelé depression, the eastern Lybian Desert or the Qattarah depressions in northern Egypt (Ginoux et al., 2012) were not identified as potential sources of dust.

It must be taken into account that the RCF showed in Figure 6, were calculated including all the episodic days identified for each circulation type and all the African dust values estimated at the nine monitoring sites over 11 years. As we showed that the contribution of the sources dust varied with the circulation type (Figure 6) and also with the season (Figure S03) new RCF computed for specific time periods and with values of dust from specific regions of the study area, aiming to provide a more detailed identification of source areas of dust and to differentiate between source

- areas. This information was included in the Revised version of the manuscript to strengthen the Identification of potential source areas of dust section. Thus, the new findings were highlighted in the abstract and the conclusions section.
- 2. In relation with the reviewer's comment "Is there not a physical way to characterize the circulation patterns?" it has been noted before that we included a discussion in pages 5510-5511 on the relationship between the occurrence of African dust episodic days for the difference circulation types and the values of the NAO index.
- 3. "Last line is confusing???". It will be corrected to be sure that the abstract will finish with a key finding.

Introduction:

- 1. Lines 14-19 (we supposed that Referee #1 referred to page 5498, Line 2-7 in the web version of the manuscript published for discussion) will be removed in the Revised version of the manuscript.
- 2. In this section the interest of dust over the western Mediterranean basin and in particular over Spain will be underlined. Under the light of recent researches acute effects on human health in this study area could be attributed to the African dust (Pérez et al., 2008; Tobías et al., 2011a-b). More recently, Reyes et al. (2014) found a significant increase in respiratory-cause hospital admissions associated with PM10 and PM10-2.5 fractions during African dust outbreaks in Madrid (Spain).
- 3. "Page 5, Line 11 to 14: reformulate more clearly". The methodology used to estimate the impact of the African dust contributions on the PM10 daily records (Page 5, Line 11-14 in the preliminary version of the manuscript; Page 5501, Line 13-16 in the web version of the manuscript published for discussion) will be more clearly reformulated in the revised paper. It was basically the same description showed in the Pey et al. (2013) paper (page 1398). More detailed comments of this methodology have been included in the "Response to Anonymous Referee 2".

Circulation classification methodology:

1. The description of the K-means circulation classifications methodology was basically the same showed by Alonso-Perez et al. (2011a) in page 112. They explained that: "K-means is based on the minimization of the sum of quadratic distances between the data points and the corresponding centroid of each group. In our case, this algorithm is therefore used to globally diminish the intragroup distance, classifying our geopotential height patterns into K groups". Formula 1 in our paper represents "the sum of quadratic distances between the data points and the corresponding

centroid of each group" and Formula 2 represents the "intra-group distance". These Formula were obtained from Philipp A. et al. (2007). This section will be also rewritten in the revised version with the aim to clarify all the steps of the selection procedure and to avoid possible mistakes.

2. We use raw data instead of anomalies for one main reason. In Huth (1996) no significant differences between using K-means with raw or anomalies data were found. However, clear differences are found for rotated T-mode PCs using raw data (excellent reproduction of predefined patterns) or anomalies data (creation of artificial types of patterns). As it seems to be irrelevant to use raw data or anomalies data, and our final goal was to establish a classification of circulation types (not circulation anomalies), raw data were used.

Across the text we always referred to daily fields because the results of the circulation classifications were showed as composite maps by averaging geopotential height at 850 hPa using the data (fields) corresponding to all days assigned to a particular cluster.

3. Page 7 line8 (page 5503, line 20 in the web version of the manuscript published for discussion): "Cycle 31r2". It is the version of the ECMWF's Integrated Forecast System used by the ERA-Interim atmospheric model and reanalysis system to generate fields of atmospheric variables. More information can be consulted in:

http://www.ecmwf.int/publications/library/ecpublications/_pdf/era/era_report_series/RS_1_v2.pdf

With the aim to clarify the paragraph, it will be rewritten as:

"The ERA-Interim atmospheric model and reanalysis system uses the cycle 31r2 version of the ECMWF's Integrated Forecast System, which was configured for the following spatial resolution:...".

Identification of potential source areas of dust:

1. We are very sorry that the Referee #1 considered this section so excruciating to read and without sense. More work will be done with the aim to clarify the methodology and the results obtained.

We agree that the methodology for the computation of Redistributed Concentration Fields has been published previously and consequently this section could be shortened. However Stohl (1998) showed that there are many trajectory statistical methods such as residence time analysis (Ashbaugh et al. 1985), Concentration Fields method (Seibert et al., 1994) or Redistributed Concentration Field Method (Stohl, 1996) among others, which have been used and compared by many authors. Moreover some of them have modified the original methods by applying different smoothing procedures or filters (Lupu and Maenhaut, 2002; Kaiser et al., 2007; Han et al., 2004) or using them with data sets from more than one receptor site (Zeng and Hopke, 1989; Lupu and Maenhaut, 2002).

Our aim was to describe explicitly the methodology used in this specific case. Do not hesitate that we will rewrite this section for the sake of clarity.

Estimation of the Impact Index:

1. "Formula 4 is unclear". ADL was defined in page 5498, as the net African Dust Load transported during African Dust Outbreaks. This parameter is an estimation of the African dust contribution to the daily PM10 concentration levels registered at regional background air quality monitoring sites, during these events.

For each sampling site, ADL_i is the mean African dust contribution estimated during the N_i episodic days produced by the circulation type i. ADL is the mean African dust contribution estimated during all the episodic days (N_t).

Thus, the Impact Index i (II_i) at one sampling site is an estimation of the impact of the circulation type i which give rise to African Dust events, on the mean African dust contribution estimated at this site.

Results and discussion:

1. Page 10, line1 (page 5507, lines 1-2 in the web version of the manuscript published for discussion): "...increased gradient from the N (21% at O Saviñao and Niembro) to the S (65% at Viznar)"????.

With the aim of clarifying the paragraph, it will be rewritten as:

"The episodic days occurred less frequently at northern locations (21% at O Saviñao and Niembro) than at central (30-50% at Els Torms, Monagrega, Risco llano, Bellver and Zarra) and southern locations (>50% at Barcarrota and Viznar) of the western Mediterranean basin. At most southern locations it is evident a higher frequency of episodic days due to the higher proximity to the African mainland".

Conclusions:

This section will also be rewritten, avoiding the abusive use of acronyms and emphasizing in the key findings.

Reply to Anonymous Referee #2:

Referee #2 detected a number of small mistakes in the paper (mainly those referred as **Minor comments 2-10**) which will be corrected in the revised manuscript.

1. In relation with **Minor comment 1**, we agree that the term "dust load" has not been appropriated used as it may be interpreted as an integrated columnar dust concentration on the site such as those provided, for example, by Cimel sunphotometers.

What we called "dust load" must be interpreted as an estimation of the African dust content deposited during African dust outbreaks, which is contributing to the ambient PM10 daily mean values, recorded at regional background air quality monitoring sites. Hence, only those events exerting any influence on the PM concentration levels at the surface level were taken into account in our study.

The procedure used to obtain these estimations of African dust contribution (Escudero et al., 2007a) is based on the identification of African episodes affecting PM levels in different areas of the Western Mediterranean Basin and the analysis of time series of PM10 levels from regional background air quality monitoring stations. Firstly, atmospheric back-trajectories were visually analysed together with meteorological charts to determine, on a daily basis, the origin of the air masses at a synoptic scale. Different numerical models (SKIRON, DREAM/BSC-DREAM8b, NAAPs) generate maps every 6 hours showing estimations of the dust concentration over geographical locations. These maps help to identify desert dust plumes moving towards and reaching different areas of the Western Mediterranean Basin. The evolution of the dust plumes can frequently be tracked with satellite imagery. All this information was obtained and analysed on a daily basis to identify the appearance of African dust plumes over the study area. This is a qualitatively way to detect the occurrence of African dust outbreaks.

It should be taken into account that different sources of error during the calculation of back-trajectories may also generate occasionally a highly different wind flow regime than the real one. Besides, models results can over or underestimate the dust load transported by the air masses and the geographical position of the dust plumes. For this reason, daily PM concentration values registered at the regional background stations were simultaneously analysed to detect increases during African dust episodic days. No increases in the PM concentration values at these stations prevent the occurrence of the African dust outbreak at this area from happening, from the point of view of the air quality at the surface level. A high number of exceedances of the PM10 Daily Limit Value (50 μ g/m³) have been recorded in regional background sites, especially in southern Europe owing to the influence of African dust outbreaks (Viana et al., 2002; Querol et al., 2004; Salvador et al., 2008). Escudero et al. (2007b) have showed that the majority of the exceedances of the PM10 DLV (>70% in most stations, reaching 100% in stations located in the central and southeastern areas of the Iberian Peninsula) in 13 regional background stations of the Iberian Peninsula were caused by African dust outbreaks for the 2001-2003 period.

Escudero et al. (2007a) demonstrated that the 30 days moving percentile 40 determined for each day, excluding the African dust episodic days, reproduces rather suitably the regional background levels at EMEP and other regional background stations in the Western Mediterranean Basin during periods with prevailing atmospheric advective conditions (Atlantic, Mediterranean and European). Thus, at regional background monitoring sites, the origin of the PM10 levels recorded during these days must be local or regional. The methodology used to estimate the net dust load during African dust episodic days, subtracts this regional background levels from the PM10 recorded at the regional background sites. This is the way we have discriminated the impact from local and regional sources from the African dust contribution in African dust episodic days at the regional background sites.

Occasionally, as suggested by the Referee #2, dust layers reach very high altitudes or the dust load transported is very low. Consequently the dust deposition processes over the area of study are inexistent or very weak and the time series of PM concentration values registered at the rural background stations do not show significant increases. These specific events are out of the scope of this study because they do not contribute to ambient aerosol concentration levels in the study area. Otherwise, Escudero et al. (2007a) specifically stated in the page 5519 of their paper that in some cases the African dust transported at relative high altitudes "would reach the ground surface in approximately 3 days. Thus, levels of PM could remain high at a given regional background (RB) site 2 days after the episode has ended and, therefore, the African period could also include these dates".

This is a quantitative and simple methodology which was validated by correlating the net African dust load determined by chemical speciation at three regional background sites in Spain (Escudero et al., 2007a), obtaining a high degree of correlation in any case. Published results showed that the method is applicable across the whole Southern Europe (Querol et al., 2009) and when compared to other measurement-based methods to quantify the African dust contribution to PM10 levels, this one was demonstrated to be the most adequate available at the present moment (Viana et al., 2010). Moreover, it has been accepted as one of the methodologies to be included in the EC Guidance for Member States regarding natural events, for evaluating the occurrence of African dust outbreaks and quantifying its contributions (http://www.magrama.gob.es/es/calidad-yevaluacion-ambiental/temas/atmosfera-y-calidad-del-aire/Directrices_Comisi%C3%B3n-SEC_208_final-en_tcm7-152574.pdf).

We have decided to replace in p. 5501, l.11, "A procedure for the quantification of the net African dust load transported during each ADO was applied to estimate the impact of the African dust contributions on the PM10 daily records" with "A procedure for the quantification of the African dust contribution (ADC) deposited during each ADO was applied to estimate the impact of the African dust contributions on the PM10 daily records". The terms "African Dust Load" (ADL) and "dust load" has been replaced with "African Dust Contribution" (ADC) all across the manuscript.

- 2. **Minor comment 2:** page 5500, line 1. The term "topography" was replaced with "pressure level" elsewhere.
- 3. **Minor comment 3:** page 5500, line 15: "The" was replaced with "This".
- **4. Minor comment 4:** page 5508, line 8: "A shift of the North African high to the west" was replaced with "A shift of the North African high to the east".
- 5. **Minor comment ·5:** page 5508, line 21: "Argelia" was replaced with "Algeria" elsewhere.
- 6. In relation with **Minor comment 6**, we agree that this paragraph (pp. 550, l. 13-16) was somewhat obscure (ambiguous). It will be replaced with:

"When this pressure gradient between the Icelandic low and the subtropical high is more intense than normal (positive NAO) the westerly winds are stronger across northern Europe, bringing Atlantic air masses over the continent associated with mild temperatures and higher precipitation. On the opposite, dryer conditions than usual are produced at lower latitudes across southern Europe".

- 7. In relation with **Minor comment 7**, "dust load" refers to the African Dust contribution in PM10. The aim of this figure was to compare the areas with higher values (>22 μ g/m³) in the RCF of dust load in PM10 displayed in Figure 6, with a global distribution of dust sources, represented by the dust source function S (Ginoux et al., 2001). We demonstrated by this way that the main potential source areas of African dust obtained in the RCF, correspond to real sources of dust. This Figure has disappeared in the new version of the manuscript.
- 8. **Minor comment ·8:** page 5514, line 3: The four images in Figure 8 were numbered as 8a, 8b, 8c and 8d.
- **9. Minor comment ·9:** page 5515, line 24: "Displacement to the west" was replaced with "Displacement to the east".

- 10. **Minor comment ·10:** page 5516, line 21: "their prediction and analysis of aerosol properties" was replaced with "forecast and analysis of aerosol properties".
- 11. In relation with **Minor comment 11**, we agree with the referee #2 that it would be very interesting finding a link between the source areas of the African dust and its chemical composition. Two main problems arise when we tried to tackle this subject.

On the one hand, we have not available time series of detailed PM10 chemical composition data at regional background sites in the Western Mediterranean Basin. Across the period of study (2001-2011) we obtained a number of short-time data bases (50-100 daily samples obtained during a 12-15 months sampling period) of PM10 and PM2.5 chemical composition at some urban and urban-background sites. At these sites local and regional contributions of typical crustal tracers (Ca, Fe, Mn, Al, Ti, Sr) are relatively high, hampering the discrimination of their contributions due to long-range transport of dust during African dust outbreaks. Aside from natural phenomena such as atmospheric resuspension processes and strong winds, which contribute with crustal components from the local and regional mineral dust sources to the PM10 bulk levels determined at the sampling sites, the resuspension of road dust may be an important carrier of vehicle and construction-demolition related pollutants in urban areas. In fact, it has been demonstrated that road transport has become the main source of mineral dust and metals in large cities without influence of industrial emissions, due to the non-exhaust emissions: road dust resuspension, brake abrasion and tyre wear (Amato et al., 2013 and references therein).

On the other hand, some authors stated that long range transport of dust will inevitably tend to alter the bulk aerosol chemistry by mixing and size fractionation, thus obscuring geological signatures from specific source areas (Schutz and Sebert, 1987; Moreno et al., 2006; Gullu et al., 1996; Claquin et al., 1999).

It is well known (Chiapello et al., 1997; Moreno et al., 2006; Avila et al., 2007) that the Tunisia and most of the western Sahara lie upon carbonated lithology. In the occidental Sahara, the Coastal Basin is composed of Mesozoic and Cenozoic carbonatic sediments, dolomites and marls. By contrast, Precambrian and Paleozoic Massifs with low carbonate content cover more southern parts comprising Chad, Sudan, Mali and Mauritania. Consequently higher contents of calcitedolomite derived elements (Ca, Mg, Sr) should be expected for the African dust contribution produced during many CT-2 and CT-3 episodic days. These circulation types are able to generate the transport of African dust from western Saharan regions and from the Atlantic coastal strip, especially during the spring period. Otherwise, higher contents of clay-silicates derived elements (Al, Fe, K, V, P) should be obtained during episodic days produced during most CT-1 and CT-4 episodic days which may produce the transport of African dust from the Hoggar massif located in central and southern Algeria.

Unfortunately until the present moment our results did not found any statistically significant between PM10 samples obtained at the urban and urban background sites during the different

circulation types for their content of crustal tracers. It should be noted that individual particle characteristics of North African dust were determined in 6 Particulate Matter samples obtained in Madrid (Coz et al., 2009) by means of a Computer-Controlled Scanning Electron Microscope coupled to energy-dispersive X-ray spectroscopy (CCSEM/EDX). This is a very expensive and complex procedure that could not be carried out systematically to the samples that composed our data bases.

References:

Alonso-Pérez, S., Cuevas, E., Querol, X.: Objective identification of synoptic meteorological patterns favouring African dust intrusions into the marine boundary layer of the subtropical Eastern north Atlantic región, Meteorol. Atmos. Phys., 113, 109-124, 2011a.

Alonso-Pérez, S., Cuevas, E., Pérez, C., Querol, X., Baldasano, J.M., Drazler, R., de Bustos, J.J.: Trend changes of African Airmass Intrusions in the Marine Boundary Layer over the Subtropical Eastern North Atlantic Region in Winter; Tellus B, 63:255-265, 2011b.

Amato, F., Schaap, M., Denier van der Gon, Hugo, A.C., Pandolfi, M., Alastuey, A., Keuken, M. and Querol X.: Short-term variability of mineral dust, metals and carbon emission from road dust resuspension. Atmospheric Environment 74, 134–140, 2013.

Ashbaugh L., Malm W. and Sadeh W., 1985. A residence time probability analysis of sulfur concentrations at Grand Canyon National Park. Atmospheric Environment, 19 (8), 1263-1270.

Avila, A., Alarcón, M., Castillo, S., Escudero, M., García Orellana, J., Masqué, P. and Querol, X.: Variation of soluble and insoluble calcium in red rains related to dust sources and transport patterns from North Africa to northeastern Spain, J. Geophys. Res. 112, D05210, doi: 10.1029/2006JD007153, 2007.

Chiapello, I., Bergametti, G., Chatenet, B., Bousquet, P., Dulac, F. and Santos Soares, E.: Origins of African dust transported over the North-Eastern Tropical Atlantic. J. Geophys. Res. 102, 13701–13709, 1997.

Claquin, T., Schulz, M. and Balkanski, Y.J.: Modelling the mineralogy of atmospheric dust sources. J. Geophys. Res. 104 (D18), 22243–22256, 1999.

Coz, E., Gómez-Moreno, F.J., Pujadas, M., Casuccio, G.S., Lersch, T.L., Artíñano, B.: Individual particle characteristics of North African dust under different long-range transport scenarios. Atmos. Environ. 43, 1850-1863, 2009.

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

Escudero, M., Querol, X., Ávila A. and Cuevas, E.: Origin of the exceedances of the European daily PM limit value in regional background áreas of Spain, Atmos. Environ. 41, 730-744, 2007b.

Ginoux, P., Chin, M., Tegen, I., Prospero, J., Holben, B., Dubovik, O., and Lin, S. J.: Sources and global distributions of dust aerosols simulated with the GOCART model, J. Geophys. Res., 106, 255–273, 2001.

Ginoux, P., Prospero, J. M., Gill, T. E., Hsu, H. C., and Zhao, M.: Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products, Rev. Geophys., 50, RG3005, doi:10.1029/2012RG000388, 2012.

Gong, X. and Richman, M. B.: On the application of cluster analysis to growing season precipitation data in North America east of the Rockies, J. Climate, 8, 897–931, 1995.

Gullu, G.H., Olmez, I. and Tuncel, G.: Chemical concentrations and elements size distributions of aerosols in the eastern Mediterranean during strong dust-storms. In: Guerzoni, S., Chester, R. (Eds.), The Impact of Desert Dust Across the Mediterranean. Kluwer Academic Publishers, Dordrecht, pp. 339–347. 1996.

Han, Y., Holsen, T. M., Hopke, P. K., Cheong, J., Kim, H., and Yi, S.: Identification of source locations for atmospheric dry deposition of heavy metals during yellow-sand events in Seoul, Korea in 1998 using hybrid receptor models, Atmos. Environ., 38, 5353–5361, 2004.

Huth, R.: An intercomparison of computer-assisted circulation classification methods. Int J Climatol 16:893–922. 1996.

Huth, R., Beck, C., Philipp, A., Demuzere, M., Ustrnul, Z., Cahynová, M., Kyselý, J. and Tveito, O.E.: Classifications of atmospheric circulation patterns: recent advances and applications, Ann. N.Y. Acad. Sci. 1146, 105-152, 2008.

Kaiser, A., Scheifinger, H., Spangl, W., Weiss, A., Gilge, S., Fricke, W., Ries, L., Cemas, D., Jesenovec, B., 2007. Transport of nitrogen oxides, carbon monoxide and ozone to the alpine global atmosphere Watch stations Jungfraujoch (Switzerland), Zugspitze and Hohenpeissenberg (Germany), Sonnblick (Austria) and Mt. Krvavec (Slovenia). Atmospheric Environment 41, 9273-9287.

Lupu, A. and Maenhaut, W.: Application and comparison of two statistical trajectory techniques for identification of source regions of atmospheric aerosol species, Atmos. Environ., 36, 5607–5618, 2002.

Moreno, T., Querol, X., Castillo, S., Alastuey, A., Cuevas, E., Herrmann, L., Mounkaila, M., Elvira, J. and Gibbons, W.: Geochemical variations in aeolian mineral particles from the Sahara-Sahel Dust Corridor. Chemosphere, 65, 261-270; 2006.

Pérez, L., Tobías, A., Querol, X., Kunzli, N., Pey, J., Alastuey, A., Viana, M., Valero, N., González-Cabré, M., and Sunyer, J.: Coarse Particles from Saharan Dust and Daily Mortality, Epidemiology, 19, 800–807, 2008.

Pey, J., Querol, X., Alastuey, A., Forastiere, F., and Stafoggia, M.: African dust outbreaks over the Mediterranean Basin during 2001–2011: PM10 concentrations, phenomenology and trends, and its relation with synoptic and mesoscale meteorology, Atmos. Chem. Phys., 13, 1395–1410, doi:10.5194/acp-13-1395-2013, 2013.

Philipp, A., Della-Marta, P. M., Jacobeit, J., Fereday, D. R., Jones, P. D., Moberg, A., and Wanner, H.: Long-term variability of daily North-Atlantic-European pressure patterns since 1850 classified by simulated annealing clustering, J. Climate, 20, 4065–4095, 2007.

Prospero, J. M., Ginoux, P., Torres, O., Nicholson, S. E., and Gill, T. E.: Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product, Rev. Geophys., 40, 1002, doi:10.1029/2000RG000095, 2002.

Querol, X., Pey, J., Pandolfi, M., Alastuey, A., Cusack, M., Pérez, N., Moreno, N., Viana, M., Mihalopoulos, N., Kallos, G. and Kleanthous, S.: African dust contributions to mean ambient PM10 mass-levels across the Mediterranean basin, Atmos. Environ., 43, 4266-4277, 2009.

Querol, X., Alastuey, A., Rodriguez, S., Viana, M., Artíñano, B., Salvador, P., Mantilla, E., Garcia, S., Fernandez, R., de la Rosa, J., Sanchez, A., Menendez, M. and Gil, J.: Levels of particulate matter in rural, urban and industrial sites in Spain. Sci. Total Environ. 334-335, 359-376, 2004.

Reyes, M, Diaz, J, Tobías, A; Montero, JC; Linares, C, 2014. Impact of Saharan dust particles on hospital admissions in Madrid (Spain). International Journal of Environmental Health Research, 24(1), 63-72, doi: 10.1080/09603123.2013.782604.

Salvador, P., Artíñano, B., Querol, X. and Alastuey, A.: 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, Sci. Total Environ., 390, 495-506, 2008.

Schütz, L. and Sebert, M.: Mineral aerosols and source identification. J. Aerosol Sci. 18, 1-10. 1987.

Seibert, P., Kromp-Kolb, H., Baltenspenger, U., Jost, D. T., Schwikowski, M., Kasper, A., and Puxbaum, H.: Trajectory Analysis of Aerosol Measurements at High Alpine Sites. A contribution to subproject ALPTRAC, The Proceedings of EUROTRAC Symposium '94, edited by: Borrell, P. M., Borrell, P., Cvitas, T., and Seiler, W., Academic Publishing, The Hague, 689–693, 1994.

Stohl, A.: Trajectory statistics-a new method to establish source—receptor relationships of air pollutants and its application to the transport of particulate sulfate in Europe, Atmos. Environ., 30, 579–587, 1996.

Stohl, A.: Computation, accuracy and applications of trajectories – a review and bibliography, Atmos. Environ., 32, 947–966, 1998.

Tobías, A., Caylá, J. A., Pey, J., Alastuey, A., and Querol, X.: Are Saharan dust intrusions increasing the risk of meningococcal meningitis?, International Journal of Infectious Diseases, 15, e503, doi:10.1016/j.ijid.2011.03.008, 2011a.

Tobías, A., Pérez, L., Díaz, J., Linares, C., Pey, J., Alastuey, A., and Querol, X.: Short-term effects of particulate matter on daily mortality during Saharan dust outbreaks: A case-crossover analysis in Madrid (Spain), Sci. Total Environ., 412–413, 386–389, 2011b.

Viana, M., Querol, X., Alastuey, A., Cuevas, E. and Rodríguez, S.: Influence of African dust on the levels of atmospheric particulates in the Canary Islands air quality network. Atmos. Environ., 36, 5861-5875, 2002.

Viana, M., Salvador, P., Artíñano, B., Querol, X., Alastuey, A., Pey, J., Latz, A.J., Cabañas, M., Moreno, T., García, S., Herce, M., Diez, P., Romero, D. and Fernández, R.: Assessing the performance of methods to detect and quantify African dust in airborne particulates. Environ. Sci. Technol., 44, 8814-8820, 2010.

Zeng, Y. and Hopke, P.: A study of the sources of acid precipitation in Ontario, Canada, Atmos. Environ., 23, 1499–1509, 1989.

Relevant changes made in the manuscript:

Abstract.

- The description of the methodology in the abstract was shortened. The Abstract was rewritten emphasizing in the key findings.
- New findings concerning the identification of specific source areas of dust associated to each circulation type during specific periods of the year, were included.
- Comments on the association found between the occurrence of the African dust episodic days produced by the different circulation patterns and the NAO index were included.

1. Introduction.

- As suggested by Referee#1, lines 2-7 in page 5498, were removed.
- The interest of dust over the western Mediterranean basin and in particular over Spain was underlined.

2. Methodology.

- As suggested by Referee#2 we have replaced in page 5501, line 11, "A procedure for the quantification of the net African dust load transported during each ADO was applied to estimate the impact of the African dust contributions on the PM10 daily records" with "A procedure for the quantification of the African dust contribution deposited during each ADO was applied to estimate the impact of the African dust contributions on the PM10 daily records".
- The terms "African Dust Load" (ADL) and "dust load" has been replaced with "African Dust Contribution" all across the manuscript.
- The methodology used to estimate the net African dust contribution on the PM10 daily records (Page 5501, Line 13-16) was more clearly reformulated.

2.1 Circulation classifications methodology.

- The description of the methodology was simplified as much as possible.
- A detailed discussion on the selection of the non-hierarchical K-means cluster analysis as the classification method was included.

- Additional comments based on the work of Alonso-Pérez et al. (2011) were included.
 Following the methodology used in this paper for circulation classifications and with the
 aim to validate the results showed in the first version of our Manuscript, we proposed to
 carry out a Principal Component Analysis in T-mode with the same data base. 4 principal
 components would be retained to be compared to the 4 clusters obtained with the Kmeans procedure.
- As requested by Referee#1 lines 10-21 in page 5503, were rewritten: "The ERA-Interim atmospheric model and reanalysis system uses the cycle 31r2 version of the ECMWF's Integrated Forecast System, which was configured for the following spatial resolution:...".

2.2 Identification of potential source areas of dust

- The description of this methodology was too long. It has been reduced and reformulated.
- With the aim to strength this section, new RCF of African dust was proposed to be computed for different regions of the study area, namely the western side, the central side and the eastern side of the Iberian Peninsula and the Balearic Islands, during different periods of the year. The new results would help to differentiate between specific source areas of dust across northern Africa, associated to each circulation pattern.

2.3 Estimation of the impact index

 This section was rewritten with the aim to clarify the procedure and the meaning of the impact index. Basically it accounts for each sampling site and each circulation type the average contribution of the African dust on the ambient levels of PM10 concentrations and the frequency of episodic days.

3. Results.

3.1 Circulation classifications

- As suggested by Referee#1, lines 1-2 in page 5507: "...increased gradient from the N (21% at O Saviñao and Niembro) to the S (65% at Viznar)" was rewritten as: "The episodic days occurred less frequently at northern locations (21% at O Saviñao and Niembro) than at central (30-50% at Els Torms, Monagrega, Risco llano, Bellver and Zarra) and southern locations (>50% at Barcarrota and Viznar) of the western Mediterranean basin. At most southern locations it was evident a higher frequency of episodic days due to the higher proximity to the African mainland".
- The 4 circulation patterns obtained from rotated PCA and K-means were showed in Figures 2, 3, 4 and 5 and compared. PC1, PC2, PC3 and PC4 circulation patterns resembled quite well cluster 2, 4, 3 and 1 circulation patterns, respectively. A discussion was included to demonstrate that the 4 circulation patterns obtained with PCA had similar features than those obtained with K-means, in terms of the frequency of episodic days attributed to

- each pattern and the season with a higher frequency of occurrence. Table 2 was included to highlight this finding. It was concluded that both methodologies produced the same qualitative results.
- As suggested by Referee#2, lines 13-16 in page 550, were replaced with: "When this pressure gradient between the Icelandic low and the subtropical high is more intense than normal (positive NAO) the westerly winds are stronger across northern Europe, bringing Atlantic air masses over the continent associated with mild temperatures and higher precipitation. On the opposite, dryer conditions than usual are produced at lower latitudes across southern Europe".
- Additional comments were included showing links between the occurrence of the circulation patterns described in our results and the findings showed by Pey et al. (2013) on the atmospheric patterns which governed the 2001-2011 summer periods.

3.2 Identification of potential source areas of dust

- This section was completely rewritten and improved. Figures 6, 7 and S03 in the first
 version of the manuscript were eliminated. A detailed identification of different source
 areas of dust associated to each circulation pattern, influencing the levels of particulate
 matter at specific regions of the western Mediterranean basin during specific periods of
 the year, was showed.
- Table 4 and Figures 9, 10, 11, 12 and 13 were included in the revised version, to describe the new results.
- Some comments on the chemical composition of African dust, in relation with the source areas, were added.

3.3 Influence of the different circulation types on the ADL levels in PM10

- The title of the section was changed with "Estimation of the impact of ADO produced by the circulation types over different regions of the western Mediterranean basin".
- The values of the impact index obtained for each circulation type and each region of study were interpreted taking into account the results showed in the previous sections.
- The four images in Figure 14 were numbered as a, b, c and d.

4. Conclusions:

- This section was also rewritten, avoiding the abusive use of acronyms and emphasizing in the key findings.
- New findings concerning the identification of specific source areas of dust associated to each circulation type during specific periods of the year were included.

•	As suggested by Referee#2, lines 21 in page 5516: "their prediction and analysis of aerosol properties" was replaced with "forecast and analysis of aerosol properties".