On the sub-micron aerosol size distribution in a coastal-rural site at El Arenosillo Station (SW-Spain)

Sorribas et al.

Response to review by Anonymous Referee #1.

We thank the referee for comments and suggestions on the manuscript. Here we detail the response to all questions. In addition, as a consequence of responding the Reviewer's comment, several Figures have been eliminated and replaced with a sentence or two of text. And also, new Figures and Tables have been added with the aim to: first, provide information valuable to the scientific community for regional comparisons, model initiation or testing, and second, describe the size distribution by means of airmass type and trajectory analysis. Then, the Figures and Sections have been renumbered and the text, abstract and conclusions have been accordingly modified, incorporating new results. A revised manuscript with these modifications is also available.

R.1.1a - The presentation of two-year, grand average number concentration is not valuable to the community for regional comparisons, model initiation or testing. The results must, at a minimum be sorted by air mass type and trajectory analysis over a reasonable transport distance and time. The local vertical structure of the atmosphere at the site should be included in the meteorological analysis.

In the revised manuscript, SMPS observations, back-trajectory analysis and meteorological parameters have been used for characterization of the submicron size range. The structure of the first part of the manuscript has not been modified. So, the area of study, the sampling station and the aerosol instrumentation, including its intercomparison with a GAW standard are again described in Sect. 2. The results and discussions are given in Sect. 3, showing again in Sect. 3.1, 3.2, and 3.3, a statistical analysis of the size distribution over the two-year period by means of the diurnal, seasonal and annual cycles of the total and modal concentration. The new information is provided from Sect. 3.4.

Section 3.4

Section 3.4 has been titled (Day's segregation by means of regional and synoptic-scale patterns). The introduction of the former Sect. 3.4 is considered as a separate Section (also called Sect. 3.4) in the revised manuscript. Also, other new information has been incorporated.

The following paragraph has been included in Page 3827, Line17. "The long-term monitoring carried out during the 498 days without influence of the sea-land breeze pattern (see Fig. 5), will allow to study the particle size distribution in terms of airmass type (Birmili et al., 2001; Tunved et al., 2003; Shen et al., 2011). Different airmass have been identified according to the long-range transport of aerosol by back-trajectories analysis using the HYSPLIT model (Draxler and Hess, 1998). Toledano et al., (2009) classified the back-trajectories on the basic of cluster analyses to determine the influence of airmass origin on column-integrated optical properties of aerosol over El Arenosillo. The cluster algorithm and the mean back-trajectories arriving at three different heights, 500 m, 1500 m and 3000 m above sea level (hereinafter referred to as centroids) were shown in the work of Toledano et al., (2009). In that study, the relation between airmass type and aerosol optical properties was only restricted at 1500 m level, being this height selected like representative of the column-integrated optical properties measured by sun – photometry technique. In our case, the 500 m altitude level was assumed the most representative for in-situ measurements. An analysis of the flow patterns that represent clusters at 500 m level and the influence of these synoptic-scale patterns over the particle size distribution will be presented in Sect. 3.5."

Also, a new Figure 6 has been included in the revised manuscript, which illustrates the annual evolution of the monthly relative frequencies for days under non-pure and pure breezes and synoptic-scale flows together with the number of days per month during the studied period.

Section 3.5

Section 3.5 has been titled (Size distribution in relation to air masses) and it is totally new. 4-day airmass back-trajectories (at 00:00 GMT, 06:00 GMT, 12:00 GMT and 18:00 GMT) were computed daily at 500 m, during the 498 days without regional circulation (see Fig. 6) and they were classified using the seven clusters obtained in Toledano et al., (2009). The new Fig. 7 shows the seven centroids of the cluster classification. A new Table 2 shows the monthly frequencies of the airmass at 500 m during the two-year period. The methodology to evaluate the median particle number size distribution per cluster is presented. The new Fig. 8 shows the median particle number size distribution per cluster and 10^{th} and 90^{th} percentiles. The new Fig. 9 shows the median surface area and volume size distributions per cluster. The resultant median size distribution has been log-normal fitted and the modal parameters are summarized in the new Table 3. Finally, the differences and similarities observed between the median size distribution per cluster are discussed.

Section 3.6

Section 3.6 has been titled (Size distribution according to sea-land breeze days). The following information has been incorporated into this section. The median size distribution for NB, PB and N-PB days has been log-normal fitted and the modal parameters are summarized in the new Table 4. Moreover, the clustering classification introduced in Sect. 3.5 was also applied during PB and N-PB days. So, the main airmass flows during PB and N-PB days were analyzed. The specific case of 15 September 2004 was included as an example of an episode caused by the sea-land breeze which can have an influence on health. Evolution of the size distribution during this event was shown in Figure 10d (now Figure 11)

Section 3.7

Sect. 3.7 has been titled (New particle formation events (N_1 and N_2)). This section has been divided into two sub-sections. Section 3.7.1 includes the analysis of Events N_1. Sect. 3.7.2 is used to present the most relevant results of Events N_2. Events N_2 were classified into two categories. The categorization followed the apparition of sea-land breeze during the diurnal evolution (N_2^B) or the influence of synoptic-scale pattern (N_2^{NB}). This new classification was incorporated to Fig. 8 (now Fig. 5). The resultant median size distribution for Events N_1, N_2^B and N_2^{NB} has been log-normal fitted and the modal parameters are summarized in the new Table 5. According to the suggestions made by the Reviewer 3 (R.3.9 and R.3.23), the individual cases haven deleted in the revised manuscript. And by the comments of the Reviewer 3 (R.3.23), we are going to prepare a separate paper with a longer time series to analyze in more detail the nucleation events in El Arenosillo. Therefore, Events N_1 and Event N_2 are presented in very general terms in the revised manuscript. The new Figure 12 shows the mean size distribution, N_{NUC}, S_T and wind direction for Events N_1, N_2^B and N_2^{NB}.

In Sect. 3.7.1,
the following sentences have been included:

- "These nucleation events were characterized by a mean particle nucleation rate of 0.74 cm^{-3} s^{-1}, a mean growth rate of 1.96 nm h^{-1} and a mean total duration of 9.25 h (starting at 10:55 GMT, ending at 20:10 GMT). These properties were calculated basing on the principles and methods inferred by Birmili et al., (2003) and on the mean size distribution (Fig. 12a.1)."

- "With the purpose to provide some information about the influence of airmass source region on Events N_1, the clustering classification introduced in Sect. 3.5 is now constrained to days by which the Events N_1 was observed. The airmass observed over El Arenosillo at 06:00 GMT, just in the hours before the onset of the event, was selected as representative in each of the 48 days with Event N_1 (see, Fig. 5), and then, the event frequency for each airmass types was calculated. It can be evinced that the Event N_1 frequency is strongly linked to the airmasses origin. Marine north-westerly aerosol from Atlantic and Arctic Oceans (clusters 3 and 6) had the highest probability to lead an Event N_1 with 42%, while the lowest probabilities were found in the desert dust airmasses (cluster 1) with 10%. Maritime westerly flow pathway from Atlantic Ocean (clusters 2 and 5) had the 23% and the continental airmasses (clusters 4 and 7) the 25%."

The following sentences have been deleted:

- "Figure 10 shows some selected examples of the different short time events. The evolution of N_{NUC},…, being its growth rate higher with 5.3 nm h^{-1}. "

- "This is followed by a study on the place where this new particle formation was produced, …, it was possible to observe a nucleation event so long."

In Sect. 3.7.2,
the following sentences have been included:

- "When comparing the mean daily evolutions of the particles size distribution for Events N_2^B and N_2^{NB}, shown in Fig. 12b.1 and 12c.1 respectively, the particle growth in the afternoon need to be analyzed. During Events N_2^B (Fig. 12c.1), the particles from the industrial area was arriving to El Arenosillo in the nucleation size range. But these particles were growing by condensation and coagulation processes and they were measured again in the evening in higher size ranges, due to the re-circulation of the airmass. In contrast during Events N_2^{NB}, the wind direction was almost constant (Fig. 12b.1) and particles on El Arenosillo were always coming directly from industries and thus the particle growth was not observed in the evening, (see in Fig. 12b.1 how the mode diameter remained as a constant value)."

- "As was done for Events N_1, Events N_2 have been also studied in terms of the airflow patterns, using a similar procedure that for Events N_1. Starting the analysis with the Events N_2^{NB}, marine north-westerly aerosols, from Atlantic and Arctic Oceans (clusters 3 and 6), had the highest event frequency with 48%, followed by the maritime westerly flow pathway from Atlantic Ocean (clusters 2 and 5) with 39%. The lowest event frequency was observed for continental (clusters 4 and 7) and desert dust aerosol (cluster 1) with 9% and 4% respectively. And finally, the study for Events N_2^B allowed us to conclude that the trajectories with presence of maritime westerly flows (clusters 2 and 5) presented the highest event frequency with 63%, following by maritime north-westerly flows (clusters 3 and 6) with 27%. The lowest frequency was observed for both desert dust (cluster 1) and continental (clusters 4 and 7) airmasses with 5%."

Abstract and conclusions

Abstract and conclusions have been modified accordingly.

The following references have been included:

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R.1.1b - The local vertical structure of the atmosphere at the site should be included in the meteorological analysis.

As was shown in Sect. 1, initially, the efforts at El Arenosillo station were focused on the characterization of the aerosol optical properties by means of remote sensing methods. In order to quantify the contribution of the tropospheric aerosol on columnar aerosol, in-situ monitoring of size distribution was started in July 2004. Some of the first studies within this strategy have been published recently in Córdoba-Jabonero et al., (2011) (see, references Section in the former manuscript). But the objective of this paper is to report the first description of this aerosol parameter in an area where there is a lack of studies about the long-term particle number size variability. Future studies will use radiosonde observations, airborne and ground-based in situ measurements, together with information from a meteorological model, to complement the conclusions obtained in our work.

R.1.2 - Other moments of the size distribution in addition to the number concentration, total or by mode, should be presented.

Mean total and modal concentrations for surface and volume metrics have been included in Table I. These values of mean total and modal surface concentration were wrong in the former manuscript. The correct data have been included in the text, Table I and Figure 12 (now Figure 13) of the revised manuscript. And their averages and evolutions have been commented several times along the revised manuscript. These are some examples:

- Section 3.1: "The mean total particle surface area (S_T) and volume (V_T) concentrations were (245 ± 140) μm^2 cm^{-3} and (9 ± 6) μm^3 cm^{-3}, respectively, and the median total concentrations (with 10-90 percentiles) were 220 (80-420) μm^2 cm^{-3} and 8 (2-16) μm^3 cm^{-3}, Table I."

- Section 3.5: "About mean total surface-area and volume concentrations for desert-dust and continental airmasses, they ranged (270 - 300) μm^2 cm^{-3} and (10 - 11.5) μm^3 cm^{-3} respectively, being the highest values of both metrics for desert-dust aerosol (Cluster 1)."

- Section 3.5: "About mean total surface-area and volume concentration for marine flows pathways, they ranged (142-208) μm^2 cm^{-3} and (4.5-6.9) μm^3 cm^{-3}."

- Section 3.7.1: "when the daily mean S_{ACC} was higher than 320 μm^2 cm^{-3}, Event N_1 was no observed."

- Section 3.7.2: "Other aerosol physical properties were Events N_2^B were characterized by an increase of S_T only during the last hours of the episode from 200 μm^2 cm^{-3} to 290 μm^2 cm^{-3} (Fig. 12c.2)."

R.1.3 - Several of the figures could be eliminated or replaced with a sentence or two of text.

- Figures 2, 3, 6, 9 y 13 have been deleted and all subsequent Figures and Tables have been renumbered afterwards.

- Page 3819, Line 9 - The next sentence was eliminated "Figure 2 shows the CF average and its standard deviation."

- Page 3820, Lines 8:10 – The sentence "The monthly number of measurements,…, spectra per month" was modified by: "The number of measurements during the period of study were about 3500-400 spectra per month".

- Page 3825, Lines 2:7 – The sentence was deleted "The mean diurnal cycles of N_T, 10th and 90th percentiles are,…, statistic parameter to observed this tendency."

- Page 3827, Lines 20:23 – The sentence "Just with the aim of showing the importance of the wind direction depending on the sea-land breeze type, to highlight that while the land breeze flow for pure breeze is blowing from NE (Doñana National Park and Seville City), it is from NW (Huelva industrial areas) for non-pure breeze. Fig. 1 shows these different particle source regions."

- Page 3831, Lines 19:27 – The sentence "Figure 13 shows the number of the events per month (N_1 and N_2). With the aim,…, did not show a clear behaviour" was modified by "The number of days with nucleation events per month (as a percentage) reached the maximum value in December 2004 and April 2005, with a percentage higher than 30%, while the minimum was found predominantly during summer time with 6%. If this events frequency is compared to the daily mean cycles per season for nucleation and Aitken modes (Sect. 3.3 and Fig. 4), it is concluded that during summer time, the maximum at noon are not mainly related to nucleation events. Following the general characterization of nucleation events of this study, a more detail description of their seasonal evolution and main aerosol physical properties, as well as a comparison with observations at other sampling sites, will be described in other paper, using a large database."

R.1.4 - The result and conclusions with respect to trends and new particle production rates and processes are very weak.

According to the comments of the Reviewer 3 in R.3.23, we are going to prepare a separate paper with a longer time series to analyze in more detail the nucleation events in El Arenosillo. Therefore, Events N_1 and Event N_2 are presented in very general terms in the revised manuscript. For more details, reviewer is advised to consult the R.1.1a (Sect. 3.7).

R.1.5 - The narrative needs to be extensively edited for proper English language usage.

The manuscript has been carefully reviewed regarding the English language. Many paragraphs have been edited to clarify the results of the study.