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 #2.

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 first 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.

General Comment

R.2.1 - This paper was difficult to read due to the poor quality of English throughout. I may have missed some important points because of that. Please have someone edit it for grammar and clarity prior to re-submitting it. I've made a pdf of my scribbled on copy but I'm not sure how useful it will be to the authors.

We greatly appreciate the Reviewer's edition. In addition, the manuscript has been carefully reviewed regarding the English language. Many paragraphs have been edited to clarify the results of the study.

Science comments:

R.2.2 - The abstract mentions 3 modes: nucleation, Aitken and accumulation mode, but then presents mean geometric diameters for four modes – need to clarify in abstract.

Following this comment and also according to a suggestion from Referee #3, the sentence "Mean total concentration was 8660 cm⁻³ and mean concentrations for the nucleation, Aitken and accumulation modes particles were 2830 cm⁻³, 4110 cm⁻³ and 1720 cm⁻³, respectively." was modified by "Mean total concentration was 8660 cm⁻³ and mean concentrations of the size limits for the nucleation, Aitken and accumulation modes particles were 2830 cm⁻³, 4110 cm⁻³ and 1720 cm⁻³, respectively."

In section 2.3 – Dataset

R.2.3 - loss calculations for 16.5 nm particles were done – what about losses for larger particles? – explain why periods of rain influence are separated out.

- Page 3819, Lines 24:26 – The sentence "... resulting an efficiency close to 85% for 16.5 nm particles" was modified by: "... resulting an efficiency from 85% for 16.5 nm particles to 94% for 604 nm particles".

- Page 3820, Lines 7:8 – The sentence "All the spectra were checked to detect incorrect measurements and periods of rain influence." was modified by "All the spectra were checked and inaccurate measurements were removed, including the periods of rain influence (about 2% of the entire data-base). Rain process has a great effect on scavenging particles and it was not considered in this study."

In section 3.1 – Mean levels

R.2.4 - note I would change section name to 'mode descriptions' – the concentrations measured at ARN are compared with whose from 1 site in China, a site in Italy and a site in Finland with the conclusion that 'particle concentrations at ARN are closer to measured levels at rural areas in similar latitudes in Europe'. This is a pretty sweeping generalization based on very few points. I believe EUSAAR data or the WDCA/EBAS could provide more aerosol concentration data for Europe to back up this claim. Alternatively, Spracklen has a paper in ACP comparing modeled and measured CN concentrations at a variety of sites around the world – including seasonal variability. This would be a better point of comparison.

The name of the section has been changed. Moreover, the point of comparison has been modified and the new information reported is:

"Comparisons with other size distributions measured at similar latitudes sites has been based on previous and recent studies which used observations (Asmi et al., 2011) and results from global aerosol models (Spracklen et al., 2010) of total particle number concentration, for better understanding the diurnal, weekly and seasonal variability at different stations around the world. From measurements performed at mid-latitudes coastal locations, mean total particle number concentrations were between 1000 cm⁻³ and 2000 cm⁻³ while greater concentrations were observed at Mace Head with 3000 cm⁻³ ³ (Spracklen et al., 2010). Given that El Arenosillo station is at mid-latitude of a coastal site, it is possible to expect that mean particle levels can be similar than in the other coastal mentioned areas. But, the influence of continental, desert dust and anthropogenic aerosol sources may increase the coastal background particle level and therefore, the mean total concentration at El Arenosillo station. Using as reference the study of Asmi et al., (2011), $(N_{AIT} + N_{ACC})$ observed at El Arenosillo Station is the second highest value measured at Central Europe; behind Ispra Station with 7188 cm⁻³ and before Cabauw and K-Puszta Stations with 5126 cm⁻³ and 4648 cm⁻³ respectively. Referring to N_{NUC} at El Arenosillo Station, this value is higher than the particle concentration at Ispra (Rodríguez et al., 2005) and Melpitz Stations (Birmili et al., 2003). So, the total and modal particle concentration at El Arenosillo was closer to the levels in rural areas, located in Central Europe, than in the coastal rural sites of the mid-latitudes.

The following references have been included:

- Asmi et al., 2011. Number size distributions and seasonality of submicron particles in Europe 2008-2009. Atmos. Chem. Phys. Discuss., 11, 8893-8976.

- Spracklen et al., 2010. Explaining global surface aerosol number concentrations in terms of primary emissions and particle formation. Atmos. Chem. Phys., 10, 4775-4793.

According to the comments of Reviewer 2, the following reference has been eliminated:

- Liu, S., Hu, M., Wu, Z., Wehner, B., Wiedensohler, A., and Cheng, Y.: Aerosol number size distribution and new particle formation at a rural/coastal site in Pearl River Delta (PRD) of China, Atmos. Environ., 42, 6275-6283, 2008.

R.2.5 - Should not combine percentiles and means in figures or discussion. Use either means and standard deviations together or medians and percentile together. Otherwise are mixing statistics.

According to the Reviewer's comment, this suggestion has been used in various sections of the manuscript. Some of the main changes are:

- Figure 5 (now Figure 3) has been modified. The standard deviation has been included and the 10^{th} and 90^{th} percentiles have been deleted.

- Figure 14 (now Figure 10) has been modified. The median daily of the particle size distribution has been included and the standard deviation has been deleted. The ratio (Mean/Median) has been also included.

R.2.6 - In discussion of figure 5b and d it is unclear where the values come from (e.g., 570 cm3, 230 cm3, 60%). Are these numbers for the whole 2 year period or an average over a month(s). Likewise, it us unclear, where the 0.5 and 1 a/cm3 values for concentration changes in accumulation and nucleation mode come from.

- Page 3823, Lines: 11:17 – In order to avoid the confusion noted by the Reviewer and for clarity in the presentation of the results, we have used the total particle increase or decrease during Periods A and B, instead of the average over the months.

- Page 3823, Lines 14:26 - The sentence "If the trends of N_{NUC} and N_{ACC} during the Periods A and B are compared, it....in the nucleation mode of 1 cm⁻³)" was modified by "The linear correlation coefficients (R) and the slope (A) between monthly mean $N_{ACC} \& N_{NUC}$ (R = 0.7, A = -0.4) were calculated. As expected, these monthly means were statistically dependent on each other. Thus, it is possible to conclude that every decrease/increase in the accumulation mode of about 0.4 cm⁻³ (slope value) was related to an increase/decrease in the nucleation mode of 1 cm⁻³. It is also corroborated from monthly means of N_{NUC} and N_{ACC} shows previously during Periods A and B."

R.2.7 - earlier in manuscript April is not mentioned as a dust-influenced month but here it is specifically discussed as such. Do you see the dust influence in the AERONET measurements (i.e., does Angstrom exponent decrease?)

The following sentence has been incorporate in the manuscript to resolve the confusion noted by the Reviewer:

"In Toledano et al (2007b) on the basic of photometric measurements, April is not mentioned as a month influenced by desert dust episodes. In April 2005, mean aerosol optical depth (AOD – 440 nm) of 0.15 and mean Ångström exponent (α) of 1.09 were reached, (AERONET Level 2.0) and in April 2006 these mean values were 0.25 and 0.76 respectively, (AERONET Level 1.5 - no data in Level 2.0 were achieved). Taking into account these mean values and those obtained by the inventory of African desert

dust events over El Arenosillo (Toledano et al., 2007b), the number of observations produced by the desert aerosols is higher in April 2006 than in April 2005. It is also corroborated by the back-trajectory analysis at 500 m presented in Sect. 3.7."

R.2.8 - 'It is not possible to indicate the reason for this behaviour, since it would take many more years with measurements'. But you could suggest possibilities or indicate what measurements you would need. Were no changes in meteorology or trajectory path observed?

Page 3824, Lines: 5-8 – This paragraph has been modified by: "From the two-year period of data presented here, a trend in increasing N_{AIT} with a rate 1150 cm⁻³ year⁻¹ was observed, such as the correlation line shows in Fig. 3c. This behaviour may be related to the impact of sea-land breeze patterns, when the land breeze flow is blowing from NE (called pure breeze in the following sections). Days under this regional pattern, which were more frequent in 2006 (see Sect. 3.4), evinced an increase of N_{AIT} in comparison with those without incidence of regional circulation (see, Sect. 3.6, Table 4). But, it is desirable to take many more years of in-situ measurements, (e.g. of particle size distributions and extinction coefficients), to know the significance and more deeply the reason of this behaviour.

R.2.9 - If nucleation and accumulation modes are anti correlated shouldn't they show opposite trend lines? Figure 5b and d both show increasing trends.

- In our discussion we wanted to say that one of the variables was directly proportional with the multiplicative inverse of the other, during a specific periods (e.g. Periods A and B). For clarity, we have changed the word 'anti-correlated' for 'inverse variation'.

- For more information about this behaviour, it is advised to consult the R.3.12 answer, corresponding to the Reviewer 3.

R.2.10 – do geometric mean diameters of dust months look different from non-dust months?

According to suggestions from the Reviewers 1 and 3, the median size distribution for each of the seven main classes of the atmospheric flows was evaluated, using the clustering algorithm presented in Toledano et al., (2009). This methodology was only applied over the days influenced by synoptic-scale pattern and the results were showed in the new Section 3.5 (Entitle: Size distribution in relation to airmasses). Median size distribution was log-normal fitted and the modal parameters were summarized for each of the seven clusters. Based on this analysis, we can answer to the Reviewer 2 his question, using e.g. the following paragraph, which has been incorporated to the manuscript.

"Significant differences were found between the median size distributions for marine and continental or desert dust airmasses. So, while maritime flows showed three lognormal modes, the continental and desert dust airmasses evinced four modes. Median size distributions for desert dust and continental aerosol were dominated by the Aitken and accumulation modes and maritime airmasses were by nucleation and Aitken modes." - This reference has been included:

Toledano, C., Cachorro, V.E., De Frutos, A.M., Torres, B., Berjón, A., Sorribas, M., and Stone, R.S. Airmass classification and analysis of aerosol types at El Arenosillo (Spain). J. Appl. Meteorol. Clim., 48 (5), 962-981, 2009.

In Section 3.3

R.2.11 - Please comment on N_{ACC} - do the seasonal peaks in N_{ACC} correspond to dust?

The Figure 6, where the seasonal peaks in N_{ACC} were observed, has been deleted because similar information was provided in Figure 7 (now Figure 4) and according the suggestions of the Reviewer 2. But, the higher levels corresponded to desert dust and continental airmasses, as was commented in R.2.10.

R.2.12 - "This observation suggests that the particle growth rate was higher during the spring months." I am missing something I think.

- Why does peaking at the same time mean the growth rate is higher?

According to the comment of the Reviewer (see, 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. So, this comment will be included in the new article and now, it was deleted of the revised manuscript.

- Is growth rate the same as nucleation rate?

- No, it is not. Particle formation rate (or particle nucleation rate) was evaluated by dividing an increase in number concentration for nucleation mode by the elapse time. And the particle growth rate was evaluated considering a closely linear fashion of the mode diameter of the total distribution (size corresponding to maximum concentration) with time.

- For more clarity in the presentation of the results, the term 'nucleation rate' has been replaced by 'formation rate'.

R.2.13 - "could be due to an increase of the atmospheric mixing and then the dilution processes with respect to spring and summer times" I would think there was less mixing/dilution in the winter due to lower boundary layer heights.

- Page 3826, Line 13 - The sentence "... it could be due to an increase of the atmospheric mixing and then the dilution processes with respect to spring and summer times" was modified by "It could be due to meteorological scenarios which produce situations of atmospheric stagnation during the warm months, characterized by the lack of renewal of the air masses. In these episodes, the particle transport to rural areas takes place through a regional atmospheric circulation (as e.g. breeze phenomenon) and its effect over rural areas is to increase the particle and gases concentration (Gangoiti et al., 2002; Querol et al., 2008; Adame et al., 2010b)."

- These references have been included:

Gangoiti, G., Alonso, L., Navazo, M., Albizuri, A., Perez-Landa, G., Matabuena, M., Valdenebro, V., Maruri, M., García, J.A., Millán, M.M. 2002. Regional transport of pollutants over the Bay of Biscay. Analysis of an ozone episode under a blocking anticyclone in west-central Europe. Atmospheric Environment, 36, 1349-1361.

Adame, J.A., Bolívar, J.P., De la Morena, B.A. 2010. Surface ozone measurements in the southwest of the Iberian Peninsula (Huelva, Spain). Environ. Sci. Pollut. Res., 17, 355-368.

R.2.14 - need to be careful comparing with mountain sites – they have different drivers of diurnal cycles.

Page 3826, Line: 17 - The reference (Venzac et al., 2009) was modified by (Shen et al., 2011).

Shen, X.J., Sun, J.Y., Zhang, Y.M., Wehner, B., Nowak, A., Tuch, T., Zhang, X.C., Wang, T.T., Zhou, H.G., Zhang, X.L., Dong, F., Birmili, W. and Wiedensohler, A. First long-term study of particle number size distributions and new particle formation events of regional aerosol in the North China Plain. Atmos. Chem., Phys., 11, 1565-1580, 2011.

Section 3.4.1

R.2.15 – "The banana shape observed for these two events allows determining that these events were produced simultaneously in a large area (at least 100 km)". How is this the case? Please provide a reference (also are has units of km2).

Page 3829, Lines: 2-3 – The sentence "... were produced simultaneously in a large area (at least 100 km)" was modified by "...were quite homogeneous in a larger-scale airmass, (Birmili et al., 2003; Kulmala et al., 2004)." These two references can be found in the manuscript published in ACPD.

Section 3.4.2

R.2.16 – need to define ultrafine size range – is there any indication of sub-um sea salt aerosol during PB days?

- Page 3820, Line 13 – The following sentence has been included: "The term 'ultrafine size range' was used to define particles with a diameter below 100 nm (nucleation plus Aitken modes)."

- In the new Sect. 3.6 (Entitled: Size distribution according to sea-land breeze days), the following paragraph has been included: "The clustering classification introduced in Sect. 3.5, was also applied during PB and N-PB days. It showed that 68% of PB days were influenced by continental and desert-dust airmasses (clusters 1, 4 and 7) and this may explain that the hourly concentrations for accumulation mode are higher that for N-PB days (see, the daily evolution of N_{ACC} showed in Fig. 10d.2)."

- In the new Sect. 3.6 (Entitled: Size distribution according to sea-land breeze days), the following paragraph has been included: "In contrast, maritime westerly flows (clusters 2 and 5) had an occurrence frequency of about 50% for N-PB days, and it may also increase the particle levels for ultrafine size range,"

- Based on the above two points, we can answer to the Reviewer 2 the question. Because the continental airmasses was the predominant flow during PB days, we think that the contribution of sea-salt aerosol over El Arenosillo is low. During N-PB days, the more frequent was the marine airmass, but the typical marine trajectory is coming from NW sector. Then, the airmass must travel some time over the Peninsula Ibérica and so, the contribution of sea-salt would be low.

Conclusions

R.2.17 - "This anti-correlation between both modal concentrations produced a weak seasonal evolution of Nt." I'm not sure I agree with this statement. Figure 5a shows a fairly strong increase in N_T along with weak trends in N_{NUC} and N_{ACC} and a fairly strong increase in N_T along with weak trends in N_{NUC} and N_{ACC} and a fairly strong trend in N_{AIT}. Perhaps I am misunderstanding what is meant by evolution.

The conclusion section was modified accordingly to the changes made in the paper. Then, according to the comment from the Reviewer the following sentence was included: "The evolution of the monthly mean total concentration over the two-year period showed independence with the seasons. This behaviour was because there were a clear inverse variation between monthly mean of N_{NUC} and N_{ACC} . The linear correlation between both modal concentrations showed that they were statistically dependents ob each other. Thus, it was possible to conclude that every decrease/increase in the accumulation mode of about 0.4 cm⁻³ was related to an increase/decrease in the nucleation mode of 1 cm⁻³. Moreover, monthly mean total concentration showed a trend in increasing which was primarily related to an increase in N_{ATT} . This behaviour may be related to a high impact of sea-land breeze circulation during 2006, but it is desirable to take many more years of in-situ measurements to know the significance and more deeply the reason of this behaviour."

Other comments

R.2.18 - Section 3.2. – what is the 75% quality criteria?

Page 3822, Lines 18:19 - The sentence "The daily number ...using the hourly averages and the 75% quality criteria" was modified by: The daily mean concentration was evaluated using the hourly averages and applying as statistical method that if the number of hourly averages was lower than 18 for each day (75% of the measurements), the daily mean was ignored."

R.2.19 - Section 3.4.2. – need to define ultrafine size range.

- Page 3820, Line 13 – The following sentence has been included: "The term 'ultrafine size range' was used to define particles with a diameter below 100 nm (nucleation plus Aitken modes)."

R.2.20 -

Figures 1 – put an arc on windrose representing 'pure' and 'non-pure'. Done

Figure 4 (now Figure 2) – Make major ticks more obvious so can figure out how to line up numbers in text with bars. Done

Figure 5 (now Figure 3) – Make major ticks more obvious so can line up months with points. I would either rearrange figure so the left column has the four trends plots stacked and the right column has the percentile plots. Or I would put the percentile bars on the main plots and get rid of the 'subplots'. Done

Figure 7 (now Figure 4) – add vertical lines to delineate seasons. Done

Figure 12 (now Figure 13) - say what the colors are in the caption or add a legend. Done