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

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Abstract

This study is focused on the analysis of the sub-micron aerosol characteristics at rural and coastal environment in Southwestern Spain. Particle number size distributions were measured in the size range (14–673) nm using a Scanning Mobility Particle Sizer (SMPS, Model 3936-TSI), from 15 July 2004 to 31 July 2006 at El Arenosillo Station. 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. Mean geometric diameters of the four modes particles, which characterized the mean size distribution per month, were about 16 nm, 42 nm, 103 nm and 237 nm. Two kinds of episodes produced a maximum of the total concentration around noon: the new particle formation and the regional recirculation such as the sea-land breeze. Two types of nucleation events (called N\(_1\) and N\(_2\)) were observed. Events N\(_1\) were an example of the influence of regional sources and Events N\(_2\) showed the weight of local industries over the rural and coastal background levels. The 60% of nucleation events were related to NE and NW wind sectors (N\(_1\) and N\(_2\) respectively), a \(\Delta T\) higher than 12 \(^\circ\)C, a wind speed higher than 2.3 m s\(^{-1}\) and a total surface area for the accumulation mode particles below of 11 190 \(\mu\) m\(^2\) cm\(^{-3}\). The influence of the sea-land breeze processes has been analyzed, observing increases of up to 50%, 110% and 90% of the particle concentration for the nucleation, Aitken and accumulation modes. Annual evolution of monthly averages allowed to conclude that the increase or decrease of 1 cm\(^{-3}\) of the concentration for nucleation mode particles was related to opposite trend of 0.5 cm\(^{-3}\) of the concentration for accumulation mode. This anti-correlation produced a weak seasonal evolution of the total particle concentration.

1 Introduction

The atmospheric aerosol plays an important role in the global understanding and regional climatic behaviour through their direct and indirect effects. According to Charl-
son and Wigley (1994), the concentration of natural aerosol has remained roughly the same since the industrial revolution, but human activity has increased the concentration of anthropogenic aerosol. The report of the International Panel of Climate Change (IPCC, 2007) claims that the effect of anthropogenic aerosol is uncertainly concerning their influence on the climate change. The direct effect of aerosol particles on climate is related to their absorption and scattering of solar radiation. The ratio of the scattering to the extinction coefficients describes the single scattering albedo and thus the influence of the aerosols on the radiation balance of the atmosphere (Takemura et al., 2002). The indirect mechanisms show the importance of the submicrometer particles as a potential source of cloud condensation nuclei (CCN), (Charlson et al. 1987).

During the last decade, the particle number size distribution has been measured in many places around the world (Heintzenberg et al., 2000; Kulmala et al., 2004) under different atmospheric conditions. Those measurements have been taken on different platforms (ground, ships or aircraft) and during short periods of time such as campaigns (Antilla et al., 2005; Dinggenen et al., 2005; Mahajan et al., 2010), or long-term monitoring (Birmili et al., 2003; Tunved et al., 2003; Venzac et al., 2009). In this context, continuous background aerosol monitoring has been performed since 1996 at “El Arenosillo”-Atmospheric Sounding Station, which is located in the southwest coast and rural area of Spain. To begin with, the efforts were focused in the characterization of the aerosol optical properties by means of remote sensing methods, using spectroradiometer and photometric measurements, (Vergaz et al., 2005; Toledano et al., 2007a, b, Cachorro et al., 2008; Prats et al., 2008). In order to quantify the incidence of the tropospheric aerosol over columnar aerosol, in-situ monitoring of particle number size distribution was started in 2004. This paper presents the first conclusions of this aerosol variable within this framework. PM$_{10}$ and PM$_{2.5}$ mass concentrations have been widely analyzed in this area (Querol et al., 2004, 2008; Sánchez de la Campa et al., 2009) but there are not studies of the particle number size distributions.

This paper has the following sections: first, the area of study, the sampling station and the aerosol instrumentation, including its intercomparison with a GAW standard.
are presented. At the result and discussion section, a statistical analysis of the size distribution over the course of two years is investigated, by means of the diurnal, seasonal and annual cycles of the total and modal concentration. The manual inspection of this evolution has evinced the presence of representative episodes with high particle concentration such as the new particle formation and the particle accumulation during sea-land breeze days, whose main characteristics are presented in terms of meteorological and aerosol physical parameters. This analysis is going to achieve the objective of this paper, since aerosol sources in a rural and coastal area such as the background environment of Doñana National Park will be assessed. Moreover, it will allow to quantify the influence of urban areas on the background monitored values.

2 Site, measurements and methods

2.1 Sampling station site

“El Arenosillo” – Atmospheric Sounding Station is a platform to investigate some topics about atmospheric sciences. One of the main research subjects at this station is the surface and remote sensing aerosol study: from particulate mass levels (gravimetric) and chemical composition (Sánchez de la Campa et al., 2009), micrometer and sub-micrometer particle number size distributions (Sorribas, 2008) and integrating scattering and absorbing coefficients (Mogo et al., 2005, 2010), to columnar aerosol physical and radiative properties (Toledano et al., 2007a). This station was equipped with instrumentation to measure other different atmospheric parameters, such as stratospheric ozone and UV solar radiation together with their incidence on biological ecosystem, and presently is also capable to monitor gaseous pollutant concentrations (O₃, NO₂ and NOₓ) and meteorological parameters.

This observatory is located at the Province of Huelva (37.1° N, 6.7° W, 40 m a.s.l.), among the Atlantic Ocean, Mediterranean Sea and North African, (see Fig. 1). It is situated at a rural protected environment, (Doñana National Park), which covers an
extensive area with a homogeneous Mediterranean pine forest, and a coastal area at the south-west where the Atlantic Ocean seashore is at a distance less than 1 km. The nearest population is a small village, named Mazagón, about 8 km west direction. The biggest and closest population is the City of Huelva (160.000 inhabitants) at 35 km to the northwest. Emissions from industrial areas around Huelva could contribute to increase the background levels at the measured site for northwest winds.

The main aerosol types present at El Arenosillo Station are coastal marine, continental and desert dust. Air mass back trajectories with starting heights of 500 m a.s.l., were used to analyze the aerosol character and showed that the main component is represented by the coastal marine conditions, which present 44% of cases followed by continental aerosol air masses with an occurrence of 38% and finally desert dust air masses, that have a relevant frequency of 18% of data (Toledano, 2005). Moreover, desert dust events are more frequent during February–March and summer months (Toledano et al., 2007b; Prats et al., 2008; Córdoba-Jabonero et al., 2010).

The wind rose in the period (15/07/2004–31/07/2006) is presented in Fig. 1. The winds blew preferably from NE and SW, indicating air came perpendicular to the Atlantic coast line. It is seen the influence of atmospheric processes, such as the sea-land breeze, more common in spring and summer time with an occurrence probability of 30% in May and a maximum of 70% in August (Adame et al., 2008). Because of the sampling area is very flat, they can be considered as a mesoscale phenomenon. Furthermore, winds from Huelva City (NW) and its surroundings could be observed with lower occurrence.

About the main meteorological characteristics, the average ambient temperature oscillated between 26°C in August and 12°C in January (annual average = 19°C). Wind speed ranges from less than 2.5 ms\(^{-1}\) in June to as high as 3.2 ms\(^{-1}\) in December (annual average = 3 ms\(^{-1}\)) (Adame et al., 2008).
2.2 Scanning mobility particle spectrometer intercomparison

The dry ambient sub-micron particle number size distribution was monitored with a TSI Scanning Mobility Particle Sizer (Knutson and Whitby, 1975). In order to assess the quality of the measurements, an intercomparison of our system was carried out in the WCCAP (World Calibration Center for Physical Aerosol Research) in Leipzig (Germany). This Institute (IfT) is endorsed by the WMO within the framework of the GAW programme and it has designed a calibration program for maintaining and comparing the measurement instrumentation. In this investigation, the limitations of our system were analyzed with the aim to perform a correct interpretation of the results. It was achieved thanks to an intercomparison campaign held at the IfT from 8 to 10 December 2003. Some instrumental restrictions are the low efficiency for measuring the nucleation and Aitken modes due to the losses in the DMA inlet (Birmili et al., 1997) and in the CPC (Su et al., 1990).

The instrument of the IfT was a DMPS (Differential Mobility Particle Sizer), which is capable to measure the particle mobility diameter in the range of 7–400 nm. In the Electrostatic Classifier, the aerosols pass through a Kr-85 Bipolar Charger and a DMA type short-Hauke with a central electrode which length is 18 cm. The CPC Model used was the 3010 of TSI. The aerosol and sheath flows were 1.0 l min$^{-1}$ and 10 l min$^{-1}$ respectively.

The SMPS of El Arenosillo (TSI Model 3936) is made up of a differential particle size classifier (TSI Model 3080, Electrostatic Classifier), a Condensation Particle Counter (TSI Model 3022A) for particle detection and the AIM software (version 4.3, TSI INC., St. Paul., MN, USA) for data reduction and analysis of the SMPS output. The polydisperser aerosol flow was 0.3 l min$^{-1}$ and the sheath flow was 3 l min$^{-1}$, which was dried with silica gel in a closed loop.

Both spectrometers were connected to the same sampling system of atmospheric aerosol. Then, both instruments characterized the same air volume and the losses in the inlet and transport lines were the same. The measurements were carried out...
simultaneously every four minutes.

Hourly averages were calculated using both measurements and the DMPS-IfT size distributions were calculated in the SMPS-El Arenosillo diameters. The correction factor (CF) was calculated using the ratio of the same hourly averages of particle size distribution given by the SMPS-El Arenosillo and the DMPS-IfT, Eq. (1).

\[
\text{Correction Factor } (D_i) = \frac{dN_i/d\log D_i \text{ (El Arenosillo)}}{dN_i/d\log D_i \text{ (IFT)}} \tag{1}
\]

In Eq. (1) the diameter dependence of this factor is observed. This CF allowed to make up for the losses that appeared in the SMPS, assuming as ideal instrument the characteristics of the DMPS of IfT. Figure 2 shows the CF average and its standard deviation. For diameters smaller than 70 nm, a decrease in SMPS efficiency was observed, with a value of 50% in 20 nm. CF was 1 for particle sizes greater than 70 nm. Using this correction factor a good coincidence between both systems was observed.

2.3 Dataset

The surface aerosol measurements were carried out in a little home-laboratory, 75 m from the main building. The inlet was placed at 3 m above the forest and about 8 m above ground level. The aerosol sampling was obtained following the recommendations of Sheridan et al. (2001) with a flow rate of 330 l min\(^{-1}\) and Reynolds Number (\(Re\)) 9100, using a vertical stainless steel tube (48 cm inner diameter and 550 cm length). Outer tube was placed concentrically around the inner tube (1 cm inner diameter and 120 cm length), which introduced the sampling aerosol in the home-laboratory with a broad curve into the flow splitter. In this tube, the flow rate was 13.3 l min\(^{-1}\) and \(Re\) 1820. A conductive flexible tube with 0.6 cm inner diameter connected to the flow splitter transported the aerosol flow up to the SMPS instrument. Sampling system efficiency was calculated according to Willeke and Baron (1993) resulting an efficiency close to 85% for 16.5 nm particles. Sampling effects were corrected for data processing.
The sub-micron particle number size distributions were measured continuously with a time resolution of 10 min from 15 July 2004 to 31 July 2006. During this period there was one longer gap, from 25 July to 11 October 2005, due to some instrumentation failures. The SMPS was operational for 604 days (83%) and its maintenance was made according to the standard procedures. The accuracy of DMA size selection was tested with PSL, obtaining a size resolution lower than 2%.

All the spectra were checked to detect incorrect measurements and periods of rain influence. So, more than 75000 valid size distributions were obtained. The monthly number of measurements during the period of study is presented in Fig. 3, with about 3500–4000 spectra per month. The data were prepared for processing using daily files. In this study, particle number size distributions were assumed to have a three modal structure: a nucleation mode 14–30 nm, an Aitken mode 30–100 nm and an accumulation mode 100–673 nm.

Meteorological parameters were obtained at 0.5 km southwester sampling site.

3 Result and discussion

3.1 Mean levels

In order to study the particle number size distribution, Table 1 presents the mean total concentration ($N_T$) with 8660 cm$^{-3}$ and the 10th and 90th percentiles with 3250 cm$^{-3}$ and 15 450 cm$^{-3}$ respectively. The median value with 7090 cm$^{-3}$ is indicating that the most frequent values were smaller than the mean total concentration. Another database recorded with the same size range and in rural sites presented mean levels such as: 16 300 cm$^{-3}$ in Pearl River Delta (China) (Liu et al., 2008), 2210 cm$^{-3}$ in Hyytiälä Station in southern central Finland (Laakso et al., 2003) and 9280 cm$^{-3}$ in Northern Italy (Ispra) (Rodríguez et al., 2005). Thus, the particle concentration at El Arenosillo was closer to the measured levels in rural areas, located to similar latitudes in Europe.
About modal concentrations, the mean values were 2830 cm$^{-3}$, 4110 cm$^{-3}$ and 1720 cm$^{-3}$ for the nucleation ($N_{\text{NUC}}$), Aitken ($N_{\text{AIT}}$) and accumulation ($N_{\text{ACC}}$) modes respectively, (see Table 1). Standard deviation of $N_{\text{NUC}}$ represented a variation of 100%, while $N_{\text{AIT}}$ and $N_{\text{ACC}}$ was higher than 65%. There is not a long-term monitoring of the aerosol particle size distribution conducted in rural areas in Spain. But if levels at El Arenosillo Station were compared to other rural sites in Europe, $N_{\text{NUC}}$ was higher than those over continental area such as Northern Italy (Rodríguez et al., 2005) while it is lower than levels observed during spring time in Hyytiäla Station (Laakso et al., 2003). On the other hand, similar concentrations for three modes were observed at the Melpitz Station, when air mass type was Southern continental (Birmili et al., 2001).

In Figure 4a the relative occurrence frequency for mode diameter (size corresponding to maximum concentration) for 1-h data-base is presented. In this way, the variability due to change for the number density was removed and the shape of the mean distribution could be analysed. The main features of the Fig. 4a show a high variability of the mode diameter within a wide range, 16.5–143 nm, with two maximums of 22% for 16.5 nm and 11% for 69.8 nm. $N_T$ was distributed around mean value of 8660 cm$^{-3}$ (see Table 1) as Fig. 4b shows, finding the higher frequency with 18% at 6000 cm$^{-3}$ and there was only 1% of total concentration above 24 000 cm$^{-3}$.

The biggest forest fire of the last decades happened at the North of Huelva Province, from 27 July to 4 August 2004. The smoke plume reached up to El Arenosillo Station, adding fine aerosol particles and affecting atmospheric conditions. During these days, the area also suffered the most intense desert dust intrusion (22 July–4 August), ever registered during the last years with modern photometric techniques (for details see Cachorro et al., 2008; Prats et al., 2008). In this present study the union of these two events is named “mixed event”. Unfortunately, problems occurred with the SMPS system during the period 24–27 July, made impossible to realize a detailed analysis of his beginning.

In order to understand the annual behaviour of the total and modal particle number concentration and the impact of this special event over mean values, Table 1
lists the statistical parameters during the entire period of study and only during the “mixed event”. During this episode, mean total particle number concentration was about 3.2 times higher than that usually encountered and its maximum was more than 188,000 cm\(^{-3}\).

In general, the particle emissions from forest fires are dominated by an accumulation mode, (Niemi et al., 2004; Janhäll et al., 2010) and occasionally also by a nucleation mode caused for the gas-to-particle conversion of inorganic and organic species released during the plume aging (Reid et al., 2005). In the “mixed event”, the mean concentration for nucleation, Aitken and accumulation modes increased by the factors of 4.7, 2.9 and 1.3 respectively and then, nucleation mode was the most influenced. Because of ultrafine mode changes fast and is not transported in the atmosphere as far as accumulation mode particles, these results can also indicate the closeness of the place where this event occurred (only 70 km away). The frequency of some aerosol physical properties for the mixed event is clearly seen in Fig. 4b, with \(\sim 30\%\) of data below 9000 cm\(^{-3}\), and \(\sim 43\%\) above 24 000 cm\(^{-3}\). On the other hand, Fig. 4a shows the highest incidence over the smallest diameters with \(\sim 40\%\) for 16.5 nm.

### 3.2 Monthly and seasonal total and modal concentrations

The daily number concentration average was evaluated using the hourly averages and the 75% quality criteria. For example, a day that represented a high daily average value was 11 April 2005 with 16,532 cm\(^{-3}\), during which the presence of a nucleation event increased the particulate levels at midday. The predominance of the accumulation mode during all day, allowed to chosen the 20 January 2005 as example of a day with a similar concentration to the mean level, with 7940 cm\(^{-3}\). Finally, 11 May 2005 was selected as representative of a low concentration, with 2520 cm\(^{-3}\). During the day before, it was drizzling and thus, the daily concentration average during 11 May was representative of the wet deposition conditions.

Figure 5 shows the monthly averages of the total and modal particle number concentrations, calculated using daily averages. 10th and 90th percentiles are indicated in
subplots, showing a high daily variability for the number concentration. The maximum was obtained in July 2006 with 11,300 cm$^{-3}$ and the minimum in January 2005 with 6,830 cm$^{-3}$. The mean total and modal concentrations are also represented (dashed grey line in Fig. 5 and also see Table 1).

In Fig. 5b and d, the monthly N$_{NUC}$ and N$_{ACC}$ are plotted and some periods are chosen to study in detail the behaviour between both concentrations (see grey areas). As it is showed the Periods A (January–March 2005) and B (April–June 2005) are representative of a clear anti-correlation between N$_{NUC}$ and N$_{ACC}$, observing an increase/decrease of N$_{NUC}$/N$_{ACC}$ respectively during the Period A and an opposite behaviour during the Period B.

The Period A showed an increase of N$_{NUC}$ with a rate of 570 cm$^{-3}$ month$^{-1}$ and a decrease of N$_{ACC}$ with 230 cm$^{-3}$ month$^{-1}$. Also, 60% of the particles in the nucleation mode grew to Aitken size range, increasing N$_{AIT}$ with a rate of 340 cm$^{-3}$ month$^{-1}$. This behaviour may be related to the new particle formation, a process which is promoted with a reduction of N$_{ACC}$ and an increase of N$_{NUC}$ and N$_{AIT}$, as it has been observed.

And during the Period B, N$_{NUC}$/N$_{ACC}$ decreased/increased with a rate of 570/300 cm$^{-3}$ month$^{-1}$ respectively. It could be explained to the presence of large particles from desert dust air mass and from an increase of atmospheric stagnation which resulted in an aerosol aging and therefore an increase for N$_{ACC}$. Higher N$_{ACC}$ produced a large surface area concentration which favoured the condensation of gases onto pre-existing particles and suppressed the new particle formation (decreasing N$_{NUC}$). Both processes are present over the Iberian Peninsula during the Period B and during spring-summer time in general (Toledano et al., 2007b; Querol et al., 2008).

If the trends of N$_{NUC}$ and N$_{ACC}$ during the Periods A and B are compared, it is possible to conclude that the decrease/increase in the accumulation mode of 0.5 cm$^{-3}$ is related to an increase/decrease in the nucleation mode of 1 cm$^{-3}$.

On the other hand, the great variability among the same months on several years is striking, such as the comparison between the concentration evolutions during Period B and Period C (April – June 2006) evinces. Thus, in April 2005 and 2006 there were
a mean values with $4040 \, \text{cm}^{-3}$ and $2710 \, \text{cm}^{-3}$ (1.5 times lower) respectively for $N_{\text{NUC}}$ and with $1220 \, \text{cm}^{-3}$ and $1940 \, \text{cm}^{-3}$ (1.6 times higher) for $N_{\text{ACC}}$. It can be related to the trend toward the increment of the desert dust episodes observed since 2002 (Toledano et al., 2007b).

From the evolution of the data-base, a trend toward the increment of $N_{\text{AIT}}$ with a rate $1150 \, \text{cm}^{-3} \, \text{year}^{-1}$ was observed, such as the correlation line shows in Fig. 5c. It is not possible to indicate the reason for this behaviour, since it would take many more years with measurements. The anti-correlation between $N_{\text{NUC}}$ and $N_{\text{ACC}}$ produced a weak annual cycle of $N_T$, marked principally by increasing $N_{\text{AIT}}$ (see correlation line in Fig. 5a). The non-dependence of $N_T$ with the season is a behaviour not observed in other rural background areas, which usually shows a clear maximum during summer time (Laakso et al., 2003; Ström et al., 2003; Venzac et al., 2009). In contrast, other in-situ compounds such as tropospheric ozone at El Arenosillo station exhibits an increase of 50% for its concentration from winter to summer times, (Adame et al., 2010).

An often applied approach is the use of equations to describe the size distribution as separated modes, i.e., as lognormal aerosol size distribution. This technique was applied over the mean size distribution per month which was lognormal fitted in four modes. The average geometric mean diameter (GMD) observed at El Arenosillo, was about 16 nm, 42 nm, 103 nm and 237 nm, respectively for the nucleation, Aitken, accumulation 1 and 2 modes. Typical continental environments show normally only one accumulation mode which presents a GMD greater than 150 nm (e.g. Birmili et al., 2001; Tunved et al., 2003; Venzac et al., 2009). This accumulation 2 mode was made up to larger particles and it could be related to the presence of the desert dust events over the Iberian Peninsula.
3.3 Diurnal evolution of the modal size ranges

The diurnal evolution of the total and the modal particle number concentrations were calculated from hourly averages for each month. The mean diurnal cycles of \( N_T \), 10th and 90th percentiles are indicated in Fig. 6a and of \( N_{NUC} \), \( N_{AIT} \) and \( N_{ACC} \) in Fig. 6b. Higher values for \( N_T \) were found during day time because of an increase for \( N_{NUC} \) and \( N_{AIT} \), being the maximum concentration around noon. As Fig. 6a shows, the 90th percentile is the most appropriate statistic parameter to observe this tendency.

The highest values for \( N_{NUC} \) were observed in April 2005 and in July 2006 with \( 10^{200} \text{ cm}^{-3} \) at 12:00 GMT and 10:00 GMT respectively. The maximum for \( N_{AIT} \) was found in July 2006 with \( 10^{850} \text{ cm}^{-3} \) at 11:00 GMT. The lowest values for \( N_T \) were reached during the early morning hours with round \( 5500 \text{ cm}^{-3} \). They were observed as much winter (January 2005) as summer (July 2005) months. This again shows the enormous variability of the ultrafine particle concentration founded at El Arenosillo. Their main reasons will be presented in Sect. 3.4.

Figure 7 shows the daily mean cycles per season for the total and modal concentrations which were calculated from the daily mean cycles per month (more details in Fig. 6). Nucleation and Aitken modes governed the daily behaviour during spring-summer times. During winter- autumn times the daily evolutions were determined only for the nucleation mode. The maximum for \( N_{NUC} \) were reached at 11:00 GMT and 10:00 GMT and for \( N_{AIT} \) at 11:00 GMT during spring and summer times respectively. Then, during summer time there was a delay of 1-h between the maximum for \( N_{NUC} \) and \( N_{AIT} \).

This observation asserts that the particle growth rate was higher during the spring months. This may be due to during spring time, the pollination of the forest around El Arenosillo was at the height of the VOC’s emissions to the atmosphere. Moreover, the particle area surface was lower in spring than in summer time, because of the number of desert dust episodes was also lower. They could allow the condensation process over the new particles was more efficient and thus, the particle growth rate in spring
was higher than summer time. On the other hand, the colder seasons (autumn and winter) had theirs maximums for $N_{\text{NUC}}$ at 14:00 GMT and 13:00 GMT respectively and for $N_{\text{AIT}}$ was during night-time.

The decline for $N_{\text{NUC}}$ and $N_{\text{AIT}}$ in the afternoon was faster during summer time than spring time. Moreover, summer time presented in the late afternoon an increase of concentration for all of the particle sizes. Maybe, at night time the VOC’s concentrations were higher than during day time, in spite of lower emissions, due to the shallower and more stable nocturnal boundary layer (Ieda et al., 2006). It could produce a particle growth by coagulation of small particles and by condensation of volatile organic components over pre-existing particles (Kulmala et al., 2001; Rodríguez et al., 2005), and then to explain the increase for $N_{\text{NUC}}$ and $N_{\text{AIT}}$ in the late afternoon.

Moreover at night, the minimum levels for $N_T$ during the cold seasons were a 15% lower than warm months and it could be due to an increase of the atmospheric mixing and then the dilution processes with respect to spring and summer times. About the absent of the diurnal evolution of $N_{\text{ACC}}$, its composition by long-lived particles must be taken into account and then it can explain that it not disappear and reappear in a diurnal behaviour, (Rodríguez et al., 2005; Venzac et al., 2009).

With the aim to study the relation between different particle size ranges, the linear correlation coefficients between hourly $N_{\text{NUC}}$ and $N_{\text{AIT}}$ ($R = 0.40$), hourly $N_{\text{AIT}}$ and $N_{\text{ACC}}$ ($R = 0.38$) and hourly $N_{\text{NUC}}$ and $N_{\text{ACC}}$ ($R = 0.09$) were calculated. As expected, concentration for $N_{\text{NUC}}$ and $N_{\text{AIT}}$) and $N_{\text{AIT}}$ and $N_{\text{ACC}}$ were statistically dependent on each other in a daily behaviour (hourly tendency), by the previous mentioned processes such as nucleation, condensation and coagulation. What is surprising is when the correlation $N_{\text{NUC}}$ and $N_{\text{ACC}}$ is evaluated from daily and monthly averages since increasing to $R = 0.33$ and $R = 0.66$ respectively. Then, the relationship between these two modes is observed again, as was showed in Sect. 3.2, when long-term scale was used.
3.4 Study of representative episodes

If there is no strong synoptic forcing, the coastal areas are influenced by mesoscale processes because of the diurnal heating and cooling differences between the land and sea. In an area as El Arenosillo, it determines the regional atmospheric conditions which affect the re-circulation of aerosol particle emitted by local and long-transport sources. The manual inspection of daily evolution of the particle size distribution and the wind direction allowed to distinguish two episodes of interest with high particle number concentration: the first, related to the sea-land breeze days and the accumulation of the pollution in the wind flow; and the second, to the new particle formation by nucleation processes. These last ones were observed as much under breeze re-circulation as synoptic forcing.

With the aim to present the main properties of the particle size distribution, initially each day was classified on the basis of the sea-land breeze phenomenon and for which, the methodology used in Adame et al. (2010) was applied. Then, all data-base (604 days) was separated in 106 days (18%) and 498 days (82%) with and without atmospheric re-circulation, respectively. The classification can be observed in Fig. 8. Adame et al. (2010) evinced the existence of two different sea-land breeze patterns, called pure-breeze and non-pure breeze. In our work, the differences in both patterns will not explain because they were presented in great detail in the previous mentioned paper. Just with the aim to show the importance of the wind direction depending of sea-land breeze type, Fig. 9 can be observed. So, as long as 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. Figure 1 situates better these different particle sources. In our data-base, the 106 days influenced by sea-land breeze can be segregated in 51 days (8%) for pure breeze type and in 55 days (9%) for non-pure breeze type. The percentages showed in brackets in this section are referred to the 604 days of the entire data-base.
With regard to nucleation events, which will be characterized in more detail in later sections, have been classified into two types depending on the wind direction during the previous hours of the nucleation burst particles: Event N$_1$ if the wind was coming from NE and N$_2$ if was from NW. The nucleation events were not observed in the remaining wind sectors. N$_1$ and N$_2$ episodes were detected under the breeze and non-breeze patterns and then, its occurrence has been evaluated for each one (see Fig. 8). As no differences were observed between both, these have been combined and Event N$_1$ occurred for 48 days (8%) and Event N$_2$ for 42 days (7%), as Fig. 8 shows.

If the days with Events N$_1$ and N$_2$ are removed on the classification of non-breeze and breeze days, 48 days (8%) were representatives of pure breeze pattern (PB days), 36 days of non-pure breeze pattern (N-PB days) and 430 days of no breeze pattern (71%). These will be discussed in detail below.

### 3.4.1 New particle formation events (N$_1$ and N$_2$)

Figure 10 shows some selected examples of the different short time events. The evolution of N$_{NUC}$, the total area surface concentration ($S_T$), as well as the wind direction can be also observed. The first short time event analysed was due to the new particle formation by gas-to-particle conversion. Three nucleation events with different characteristic are showed in Fig. 10a, 10b and 10c which appeared during 11 April 2005, 8 June 2005 and 18 December 2004 respectively. Theirs main properties were calculated using the methodology presented in Birmili et al. (2003). The first one started at 09:30 GMT and it had a total duration of 19.2 h, a particle formation rate of 3.5 cm$^{-3}$ s$^{-1}$ and a growth rate of 3.8 nm h$^{-1}$. But, the second one started at 08:50 GMT and it was a lower duration and formation rate with 6.8 h and 2.4 cm$^{-3}$ s$^{-1}$ respectively, being its growth rate higher with 5.3 nm h$^{-1}$.

The homogeneous nucleation process enhanced the particle concentration at diameters below the detection limit of our particle spectrometer. The growth toward greater diameters, due to the condensation and coagulation processes, caused an increase of number concentration, at first for the nucleation mode and later, for Aitken and
accumulation size ranges, such as the evolution of mode diameter evinces in Fig. 10a and b. The “banana” shape representative for these two events allows determining that these events were produced simultaneously in a large area (at least 100 km). So, new particles which are formed in other point are growing during the transport to El Arenosillo site and theirs sizes were measured within of the ultrafine mode. Due to the sampling size started at 14 nm, it is not possible to conclude that the formation processes took place in the sampling site. These two nucleation events were characterized by a slow increase of $S_T$ and by a fast increase and later decrease of $N_{NUC}$ with the maximum around midday.

Meteorological analysis showed that they occurred when the wind was blowing from the NE direction during the prior hours of its apparition (see Fig. 10a and b). They have been called Event $N_1$.

This is followed by a study on the place where this new particle formation was produced, choosing as example the day 11 April 2005. Given that the initial size of the new particles was 1 nm, the lowest diameter measured in El Arenosillo was 14 nm and the growth rate was 3.8 nm h$^{-1}$, the formation occurred 3.7 h before that the particle arrived to the sampling site. Then, it happened at 05:50 GMT. During that time, while the new particles arrived at El Arenosillo, the average wind speed was 4 m/s, which allows to conclude that the first monitored particles were formed as very close to 55 km away in NE sector. Due to the changes in the wind direction occurred after 12:00 GMT, the particles that previously passed on El Arenosillo during the morning are again measured in the afternoon. Given that they had grown through the condensation and coagulation processes, it was possible to observe a nucleation event so long.

In Fig. 11a the median diurnal evolution of the particle number size distribution for the 48 days (see Fig. 8) with presence of Event $N_1$ is presented.

Selecting the PB days and those with the wind blowing from the NE direction during all day, in Fig. 12 are presented the main characteristic of the daily mean of the particle concentration by means of some aerosol physical properties and some meteorological parameters. The daily mean properties for all days are in black colour and the days...
with Events $N_1$ are presented in red colour. Figure 12a shows an anti-correlation between $N_{NUC}$ and total area surface concentration for accumulation mode ($S_{ACC}$), while in contrast, $N_{ACC}$ and $S_{ACC}$ and $S_T$ and $V_T$ are highly correlated (see Fig. 12c and d). The Events $N_1$ (red spots) are associated to low or medium values of $S_{ACC}$, $N_{ACC}$ and $V_T$. It is due to the presence of pre-existing particles provides enough area surface to use the precursor gases in the condensation process and then to increase the size but no the particle number in the atmosphere. Then, these gases are not used in the nucleation processes.

$N_{NUC}$ is also represented by means of some meteorological factors such as $\Delta T$ (temperature difference between minimum and maximum every day) and wind speed, (see Fig. 12e and 12f respectively). $\Delta T$ is related to the mixing status of the atmosphere. Since, higher values of $\Delta T$ are representatives of better mixing. In Fig. 12e, it is evinced that the Events $N_1$ were produced with $\Delta T$ above 10°C. This observation was already realized in Buzorius et al. (2003) where, it was concluded that days with new particle formation appeared with advection of cold air toward surface and then with high values of $\Delta T$.

Finally, Fig. 12f showed Events $N_1$ occurred when the wind speed was higher than 2 m s$^{-1}$ and the highest $N_{NUC}$ was reached during the highest speed averages.

In order to propose a methodology for the quantification of the aerosol physical properties and meteorological parameters which are necessary for predicting the Events $N_1$ at the southwest Spain, some statistical parameters have been evaluated. Then, the conclusions have been that 60% of Events $N_1$ can be predict if the wind is blowing from the NE direction in the morning and there are a $S_{ACC}$ below of 11 190 µm$^2$ cm$^{-3}$ (30th percentile), a $\Delta T$ higher than 12°C (10th percentile), and a wind speed higher than 2.3 m s$^{-1}$ (10th percentile).

Particle number size distribution evolution during 18 December 2004 is chosen as an example (see Fig. 10c) to display the second type of nucleation event. These events happened when the wind was blowing from NW-N directions. They could be explained as a secondary particle formation which was produced by the emissions
from the industrial areas situated around Huelva City. Because of the wind effects, the anthropogenic pollution with a continuous high level of precursor gases can be able to cause an increase of the concentration for ultrafine size range over the sampling site during an elapsed time between 2 and 8 h.

It was called Event N₂ and it is also different than the analyzed previously, because the highest plateau concentration of $N_{\text{NUC}}$ was maintained over time. So, $N_{\text{NUC}}$ in Fig. 10a and b present a slight diurnal trend toward the larger particles sizes, which was characteristic of the Event N₁, while in Fig. 10c the maximum of $N_{\text{NUC}}$ stays longer during the Event N₂. It can be explain because the particle source of the Event N₁ could be a large area and, in function of the time that the particle took to arrive at El Arenosillo, it could have grown up to different diameters. However, in the case of the Event N₂, it could be a punctual source and then all particle emitted simultaneously were arriving at El Arenosillo at the same time and they were grown up to similar size.

Otherwise, they were characterized by an increase of $S_T$ only during the last hours of the episode. About the relation of Event N₂ with the aerosol properties and the meteorological factors, they were similar to those observed for the case of Event N₁ in Fig. 12. Median diurnal evolution of the particle number size distribution for the 42 days with presence of Event N₂ (see Fig. 8) is presented in Fig. 11b.

Figure 13 shows the number of the events per month (N₁ and N₂). With the aim to evident the representatively of the results, the number of measured months is also indicated. The Event N₁ presented a clear seasonality with two maximum (in March and in December) of 5 and 4 events per month respectively. The minimums were reached during summer time, being its occurrence zero during July and August. Just as, it has commented at some opportunities throughout this report, this behaviour could be related to the increment of the desert dust episodes during summer time. On the other hand, the monthly evolution of the Event N₂ (see Fig. 13), did not show a clear behaviour.
3.4.2 Sea-land breeze days

The presence of regional re-circulations as the sea-land breeze are very typical over the coastal areas during spring and summer time and they play an important role in transporting air pollution from and towards the urban areas, (Ma and Lyons, 2003; Sorribas et al., 2007; Adame et al., 2010; Wright et al., 2010). As noted above in Sect. 3.4, two types of sea-land breeze over our study area have been observed: pure breeze and non-pure breeze. Wind blowing at night from the land (NE for pure breeze and NW for non-pure breeze) towards the sea as a result of the nocturnal cooling of the land surface is called land breeze. This local wind produced an accumulation of particles over the sea at offshore distance. Then, the wind direction changed at round 07:00 GMT and 08:00 GMT for pure breeze and non-pure breeze respectively (Fig. 9a). It was blowing during the day from the sea surface (SW sector for both land patterns) towards the land as a result of diurnal heating of the land surface (sea breeze). Then, the accumulated particle was transported over the land, increasing the background levels.

Figure 14 shows the mean daily particle number size distribution and mean daily evolution of modal concentrations corresponding to 430 NB-days, 48 PB-days and 36 N-PB-days (see the day segregation in Fig. 8). 10th and 90th percentiles are also represented.

To quantify the effect of each type of breeze, due to the accumulated particles at offshore distance, NB days were chosen as reference of the modal concentration reached. For NB days, the maximum concentration of \( N_{NUC} \) (\( N_{NUC_{max}} \)) was 3450 cm\(^{-3}\) (Fig. 14b.1). It was a 30% for PB and a 50% for N-PB days higher than for NB days (Fig. 14b.2 and b.3). In the case of \( N_{AIT_{max}} \) during NB days its level was 4280 cm\(^{-3}\) and for PB and N-PB it was 30% and 110% higher respectively (Fig. 14c.1, c.2 and c.3). And finally, \( N_{ACC_{max}} \) was 2000 cm\(^{-3}\), increasing by 90% for PB days and decreasing by 5% for N-PB days (Fig. 14d.1, d.2 and d.3). Then, it is concluded that the two breeze patterns were a particle source within the ultrafine size range and only PB scenario...
within the accumulation mode. Some factors that has to be taken into account are that PB days were influenced by continental air mass (see Figs. 1 and 9a) and then, the size range more influenced was the accumulation mode. On the other hand, marine and industrial aerosol had an influence on the concentrations measured during N-PB days, which have a size within the ultrafine range.

For PB days, the daily evolution of $N_{\text{NUC}}$ presented three minimums (Fig. 14b.2), which were reached at 08:00 GMT, 18:00 GMT and 02:00 GMT. This behaviour was only observed in this scenario. The first minimum may be caused by the mixing processes that could begin after sunrise and therefore the breakdown of the mixed layer, which increased the particle dilution. And the second and third minimums and the maximum concentration at 20:00 GMT had already been discussed earlier in Sect. 3.3 due to the evolution of $N_{\text{NUC}}$ and $N_{\text{AIT}}$ observed in Fig. 7 during the summer months. During these months, the occurrence of the breeze phenomenon is higher as the maximum gradient of temperature between land and sea is reached. Thanks to the evolution shown in Fig. 14b.2, it is reasoned that the behaviour can be observed in Fig. 7 is only caused by the PB scenario. Future works will be guided to know the reason for this behaviour, which will be based on the correlation to some meteorological parameters with vertical resolution and the aerosol data.

The evolution of $N_{\text{NUC}}$ during N-PB days presented an absolute maximum at 10:00 GMT, then two hours before that PB days (Fig. b.2 and b.3). This behaviour could be related to the transport particle speed (wind speed), which for N-PB days is generally higher (see Fig. 9b).

As long as the maximum concentration at noon for N-PB and PB days was due to the accumulated particle produced by the land breeze, the maximum concentration for NB days was because of the occurrence of episodes similar than Events N$_1$. The difference between the mean size distribution of Event N$_1$ (Fig. 11a) and the mean total particle concentration of NB days (Fig. 14b.1, c.1 and d.1) was the maximum of the total particle concentration reached. Thus, for the case of Event N$_1$, that maximum was greater than 17 000 cm$^{-3}$ and for the case of NB days was around 10 000 cm$^{-3}$.
On 15 September 2004 has been selected as example of an N-PB day (Fig. 10d). The wind direction evolution shows that land breeze propagated in offshore direction (from NW-N) during the night, the onset of sea breeze was at 09:30 GMT and then the sea breeze blew perpendicular to the coast (from SW). 40 min later, air mass with high particle concentration came to the sampling area and it was influenced during the next five hours. Concentrations were multiplied by 12, 3.8 and 2.6 for the nucleation, Aitken and accumulation modes. Total surface concentration was also increased. This event was the longest observed during the data-base analyzed. During the DAMOCLES campaign, these type of episodes were also differentiated using PM and chemical composition measurements over this area (Pey et al., 2008).

4 Conclusions

In this paper, an analysis about the sub-micron particle number size distribution in Southwest Spain is presented. It was measured continuously during 604 days at El Arenosillo Station which is located in a rural and coastal background environment. Mean total concentration was 8660 cm\(^{-3}\) and mean modal concentrations were 2830 cm\(^{-3}\), 4410 cm\(^{-3}\) and 1720 cm\(^{-3}\) for \(N_{\text{NUC}}\), \(N_{\text{AIT}}\) and \(N_{\text{ACC}}\) respectively. The most significant episode was a “mixed event” of a strong desert dust intrusion plus a forest fire occurred during 27 July–4 August 2004. It increased the mean modal concentrations by 4.1, 2.8 and 1.2, respectively.

About the modal parameters the mean size distribution per month was separated in four modes and the mean geometric diameters were about 16 nm, 42 nm, 103 nm and 237 nm for nucleation, Aitken, accumulation 1 and accumulation 2 modes, respectively.

Daily pattern exhibited an absolute maximum of the total concentration around noon, which was governed by \(N_{\text{NUC}}\) and \(N_{\text{AIT}}\) during the warm seasons and only by \(N_{\text{NUC}}\) during cold seasons. This maximum was produced for the new particle formation and for the accumulation of particle during the typical air mass recirculation over the coastal areas such as the sea-land breeze. By means of the wind direction, two types of
nucleation events (called $N_1$ and $N_2$) were detected. 60% of these were related to NE and NW wind directions respectively, a $S_{ACC}$ below of 11 190 µm$^2$ cm$^{-3}$, a $\Delta T$ higher than 12 °C and a wind speed higher than 2.3 m s$^{-1}$. Events $N_1$ were an example of the influence of regional sources and they were characterized by the aerosol number transfer from the nucleation to the Aitken mode due to particle growth. However, Events $N_2$ showed the weight of local industries over the rural and coastal background levels and they did not evince the particle size growth. The number of Events $N_1$ per month presented the maximum values during spring and winter with 5 and 4 events respectively and the annual evolution could be related to the frequency of desert dust episodes. The monthly evolution of the Events $N_2$ did not show a clear behaviour.

The influence of the sea-land breeze processes has been presented by means of two different patterns (pure and non-pure breezes), observing that both were particle sources within ultrafine size range. If the maximum particle concentrations for NB days are chosen as reference (3450 cm$^{-3}$, 4280 cm$^{-3}$ and 2000 cm$^{-3}$ for $N_{NUC}$, $N_{AIT}$ and $N_{ACC}$ respectively), $N_{NUC_{max}}$ was increased about 30% and 50% for PB and N-PB days respectively, and $N_{AIT_{max}}$ about 30% and 110% respectively. $N_{ACC_{max}}$ was only increased about 90% for PB days.

Because Events $N_1$, Events $N_2$ and the N-PB and PB days were well determined from the wind sectors, the horizon of the sampling site could be divided into two hemispheres. High particle number concentration hemispheric (HC) from SW to NNW in clockwise presented (9120±264) cm$^{-3}$ and lower particle number concentration hemispheric (LC) from N to SSW in clockwise (6550±780) cm$^{-3}$.

Annual cycle allowed to conclude that the increase or decrease of 1 cm$^{-3}$ of $N_{NUC}$ was related to opposite trend of 0.5 cm$^{-3}$ of $N_{ACC}$ and moreover that, the relation between monthly $N_{NUC}$ and $N_{ACC}$ was highly correlated with $R = 0.66$. It could be explained because of the presence of large particles from desert dust air mass resulted in an aerosol aging and an increase of $N_{ACC}$. It produced a large surface area concentration which favoured the condensation process onto pre-existing aerosols and suppressed the nucleation events (decreasing $N_{NUC}$). This anti-correlation between
both modal concentrations produced a weak seasonal evolution of $N_T$.

On the other hand, a trend to increase the $N_{AIT}$ is observed with a rate of $1150 \text{ cm}^{-3} \text{ year}^{-1}$. It is not possible to conclude the reason for this conduct because many more years with measurements are necessary.

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References


Table 1. Statistic of the total and modal concentration for 1-h year data series and only for the mixed event (desert dust episode plus forest fire) happened during 27 July–4 August 2004.

<table>
<thead>
<tr>
<th></th>
<th>N&lt;sub&gt;Total&lt;/sub&gt; (cm&lt;sup&gt;-3&lt;/sup&gt;)</th>
<th>Nucleation Mode (cm&lt;sup&gt;-3&lt;/sup&gt;)</th>
<th>Aitken Mode (cm&lt;sup&gt;-3&lt;/sup&gt;)</th>
<th>Accumulation Mode (cm&lt;sup&gt;-3&lt;/sup&gt;)</th>
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<tr>
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<td>15 450</td>
<td>6800</td>
<td>7730</td>
<td>3160</td>
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<td></td>
</tr>
<tr>
<td>Mean</td>
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<td>13 320</td>
<td>12 020</td>
<td>2270</td>
</tr>
<tr>
<td>STD</td>
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<td>37 310</td>
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<td>24 500</td>
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Fig. 1. Map of Iberian Peninsula highlighting the location of El Arenosillo Station, the (15/07/2004–31/07/2006) wind rose and the coastal line.
Fig. 2. Ratio of the size distribution measured by the SMPS-EI Arenosillo and the DMPS-IfT. It is defined as the Correction Factor (FC).
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Fig. 7. Daily mean cycles of the total and nucleation, Aitken and accumulation mode concentrations per season, July–September (summer), October–December (autumn), January–March (winter) and April–June (spring).
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Fig. 10. Daily evolutions of the particle number size distribution at El Arenosillo Station, during (a) 11 April 2005, (b) 8 June 2005, (c) 18 December 2004 and (d) 15 September 2004. Total surface (black line), concentration for nucleation mode (red line), wind direction (green line) and mode diameter (black diamonds) are also represented.
Fig. 11. Mean particle size distribution for new particle formation with (up) wind blowing from NE (Event N$_1$, 48 days) and (down) blowing from NW (Event N$_2$, 42 days) at El Arenosillo Station.
Fig. 12. 24-h averaged of several submicron aerosol physical properties and meteorological parameters at El Arenosillo station.
Fig. 13. Percent of days per month with Event N₁ and Event N₂.
Fig. 14. Mean daily, 10th and 90th percentiles of particle number size distribution and for modal concentration corresponding to 430 NB-days (non breeze), 48 PB-days (Pure) and 36 N-PB-days (Non-Pure) breeze patterns at El Arenosillo.