IOP Conf. Series: Earth and Environmental Science **28** (2015) 012001 doi:10.1088/1755-1315/28/1/012001

Atmospheric Particle Size Distributions in the Spanish Network of Environmental DMAs (REDMAAS)

E Alonso-Blanco[1], F J Gómez-Moreno[1], B Artíñano[1], S Iglesias Samitier[2], V Juncal[2], M Piñeiro Iglesias[2], P López Mahía[2], N Pérez[3], M Brines[3], A Alastuey[3], X Querol[3], B A de la Morena[4], M I García[5,6], S Rodríguez[5], M Sorribas[7,8], G Titos[7,8], H Lyamani[7,8] and L Alados-Arboledas[7,8]

[1] Department of Environment Associated Unit CSIC-CIEMAT on Atmospheric Pollution, CIEMAT, Madrid, Spain.
[2]Grupo Química Analítica Aplicada, Instituto Universitario de Medio Ambiente (IUMA), Centro de Investigaciones Científicas Avanzadas (CICA), Departamento de Química Analítica, Facultade de Ciencias, Universidade da Coruña, A Coruña, Spain.
[3]Institute of Environmental Assessment and Water Research (IDAEA-CSIC), Barcelona, Spain.
[4]Atmospheric Sounding Station "El Arenosillo", INTA, Mazagón-Huelva, Spain.
[5]Izaña Atmospheric Research Centre, (IARC/CIAI), AEMet, Santa Cruz de Tenerife, Spain.
[6]Department of Chemistry (T.U. Analytical Chemistry), Faculty of Science, University of La Laguna, Tenerife, Spain.
[7]Andalusian Institute for Earth System Research, IISTA-CEAMA, University of Granada, Granada, Spain.
[8]Applied Physics Department, University of Granada, Granada, Spain.

E-mail: elisabeth.alonso@ciemat.es

Abstract. The present work is a first approach to the study of the spatio-temporal variability of the submicrometer atmospheric aerosol in Spain. The aerosol measurements have been obtained simultaneously at seven monitoring stations that compose the REDMAAS network during two measurement campaigns corresponding to summer and winter seasons.
In both summer and winter periods those measurement stations with a direct influence of anthropogenic emissions recorded the highest concentrations of particle number. In the summer campaign, the average daily pattern of the aerosol size distribution in the traffic and background urban stations was conditioned by the traffic emissions and secondary aerosol formation through photochemical reactions (new particle formation events, NPF). However, the secondary aerosol had a higher contribution to the aerosol total number concentration in the rural background and high-altitude stations. In the winter campaign, in all sampling sites with the exception of Izaña station, the traffic and domestic activity emissions had a greater contribution than secondary aerosol formation on particle number total concentration.
New particle formation events were identified at all sites during the summer period, and at sites without direct influence of anthropogenic emissions during the winter campaign. Some aerosol shrinkage processes were also observed at the Madrid and El Arenosillo stations.

1. Introduction

Aerosol size distribution plays a crucial role on climate forcing and therefore the knowledge on its spatial and temporal variability has important implications in the understanding of climate impact.

Compilations of aerosol measurements with spatio-temporal dimension are scarce in the literature. Typically, these works are framed within aerosol instrumentation networks such as [1] focused on submicrometer atmospheric aerosol measurements in Germany obtained from the German Ultrafine Aerosol Network (GUAN, 2008) or [2] presenting submicrometer atmospheric aerosol measurements in Europe carried out within the Aerosols, Clouds, and Trace gases Research InfraStructure (ACTRIS) Network (www.actris.net).

The strong climate variability and geographical features diversity confer unique characteristics to the study of the atmospheric aerosol in Spain. Works that characterize the submicrometer atmospheric particles in different areas of the Spanish territory are numerous, as those developed by [3], [4], [5], [6], [7] and [8]. Nevertheless, to the moment there is no study on the variability of the characteristics of submicron aerosol size distributions in Spain on a temporal and spatial scale basis.

With this aim, measurements at different sites of the Spanish Network of Environmental DMAs (REDMAAS: http://www.redmaas.com) have been analysed. The aerosol measurements were simultaneously obtained at seven monitoring stations corresponding to the six research groups that form the REDMAAS. Two measurement periods, under different meteorological conditions, a summer period (1-30 June 2012) and a winter period (13 December 2012-15 January 2013) were selected for the study.

The main objective of this study was to characterize, for the first time in Spain, the influence of the meteorological conditions and the characteristics of the sampling sites on the features of submicrometer atmospheric aerosol size distributions.

2. Measurement campaigns

2.1. Measurement sites

The seven monitoring stations are located throughout the entire Spanish territory being representative of the climate and territorial variability of the country. A summary of the characteristics of each measurement station is shown in Table 1.

Table 1. Characteristics of the seven observation sites within Spain: research group/city, type of measurement station (Urban Traffic (UT), Urban Background (UB), Rural Background (RB) and High-Altitude (HA)), location (geographical coordinates and altitude) and climate.

GEOGRAPHICAL AREA AND CLIMATOLOGY		
Research group/City	**Measurement station type/Location**	**Climate**
IUMA-UDC: A Coruña	UB: 43.3 N, 8.4 W, 45 m a.s.l.	Atlantic
CIEMAT: Madrid	UB: 40.5 N, 3.7 W 657 m a.s.l.	Continental Mediterranean
IDÆA-CSIC: Barcelona	UB: 41.4 N, 2.1 E 68 m a.s.l.	Mediterranean
IDÆA-CSIC: Montseny	RB: 41.8 N, 2.4 E 720 m a.s.l.	Mediterranean
INTA: El Arenosillo (Huelva)	RB: 37.1 N, 6.7 W, 40 m a.s.l.	Ocean Mediterranean
CEAMA-UGR: Granada	UT: 37.2 N, 3.6 W, 680 m a.s.l.	Continental Mediterranean
IARC-AEMET: Izaña (Tenerife)	HA: 28.3 N, 16.5 W, 2367 m a.s.l.	Temperate Mountain

2.2. Measuring Instruments

During the campaigns, continuous measurements of aerosol size distributions have been performed with Scanning Mobility Particle Sizers (SMPS) in all sampling stations, with the exception of Granada station. For this site, an Ultrafine Particle Monitor (UFPM) was used.

SMPS equipments are composed by a Differential Mobility Analyser (DMA) connected to a Condensation Particle Counter (CPC), while UFPM is composed by a DMA and an aerosol

electrometer used as detector. Detailed information on the equipments used in the aerosol monitoring campaigns can be found in table 2.

Table 2. Characteristics of the measurement equipment on each sampling station.

Equipment	Neutralizer/charger	Flow rate	Measurement size range/ Temporal resolution	Nº Channels (Decade)
IUMA-UDC: A Coruña				
TSI-SMPS: DMA 3081 CPC 3785	Kr-85 (2 mCi)	10:01	7.37/7.64-299.6/289 (4.5 min)	104 (64)/ 102 (64)
CIEMAT: Madrid				
TSI-SMPS: DMA 3081 CPC 3775	Kr-85 (2 mCi)	10:01	20.2-661.2 (4.5 min)	107 (64)
IDÆA-CSIC: Barcelona				
TSI-SMPS: DMA 3081 CPC 3772	Kr-85 (2 mCi)	5:01	10.9-478.3 (5 min)	106 (64)
IDÆA-CSIC: Montseny				
SMPS-IfT: DMA CPC 3772	Kr-85 (2 mCi)	5:01	10.9-478.3 (5 min)	71 (38)
INTA: El Arenosillo (Huelva)				
TSI-SMPS: DMA 3081 CPC 3776	Kr-85 (2 mCi)	10:01	14.1-736.5 (10 min)	111 (64)
CEAMA-UGR: Granada				
TSI- UFPM 3031	Corona-jet charger	5	20->200 (10 min)	5
IARC-AEMET: Izaña (Tenerife)				
TSI-SMPS: DMA 3081 CPC 3010	Kr-85 (10 mCi)	5:01	10.6-495.8 (5 min)	108 (64)

The instrument intercomparison within the REDMAAS network carried out prior to measurement campaigns [9, 10] guaranteed the quality of measurements.

In order to compare SMPS measurements, the data have been corrected for diffusion losses and multiple charges with the TSI AIM (Aerosol Instrument Manager, TSI Inc.) software. Furthermore, aerosol data obtained at Madrid, El Arenosillo and Izaña stations have also been corrected for particle diffusion losses in the entry sampling lines to the SMPS [11].

Total particle number concentration (N_t), and the number of particle concentrations for each of the three modes: nucleation ($N_{<30\ nm}$), Aitken ($N_{30\text{-}100\ nm}$) and accumulation ($N_{>100\ nm}$), were obtained from the aerosol size distributions. The choice of the intervals of particles size that define each of the modes is based on the classification developed by [12].

The average data coverage for each of the measurement campaigns (summer and winter periods) were 61% and 74% respectively. Figure 1 shows the data coverage for each measurement station and study period.

In addition, the local new particle formation (NPF) events have been identified based on the methodology developed by [12]. This methodology has also allowed the identification of shrinkage events.

For these events, the growth (GR) and shrinkage rates (SR) were calculated based on [13]. The calculation was made from the modes of the size distributions averaged every 15 or 10 minutes (D_{mode}), depending on the temporal resolution of each instrument (table 2). The aerosol size distributions were fitted to a lognormal function to estimate the mode.

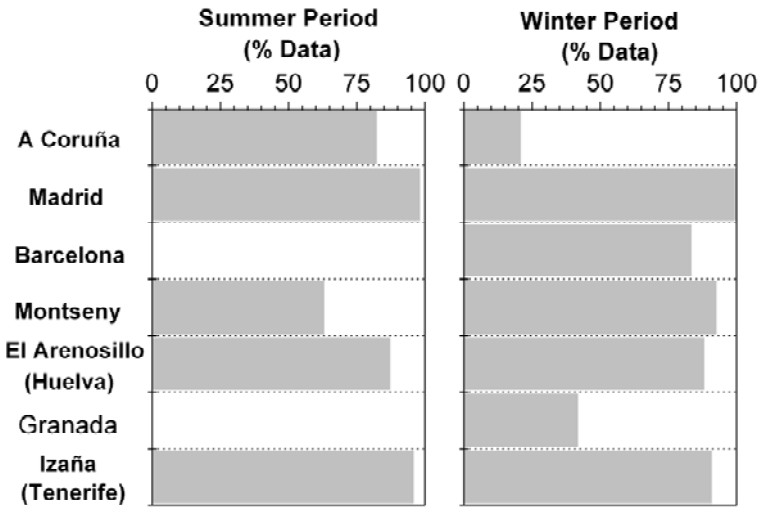

Figure 1. Data coverage of the submicron aerosol fraction for each measurement campaign and station.

3. Results and discussion

3.1. Particle number size distributions

The meteorological situation is rather different between both periods studied. In general, an active atmospheric dynamic together with an increase of the convective boundary layer facilitate the dilution processes in the warm period. However, in the cold period an increase of the anthropogenic emissions together with a greater frequency of high pressure systems over Iberian Peninsula lead to a high atmospheric stability and consequently to an increase of the atmospheric pollution load.

This situation caused differences in averaged total particle concentration between the two measurement periods in the different sampling sites.

With the exception of the Madrid station, data gaps from the traffic and background urban stations have made difficult the comparisons between the measurement periods (figure 1). In Madrid, N_t levels during the cold period were higher than during the warm period. This fact was due to the more stable conditions and reduction of the boundary layer along with the increase of pollutant emissions from traffic and domestic activities in this period [8]. On the opposite, at the rural background and high-altitude stations, with low or zero direct anthropogenic emissions, N_t was greater during the summer period because a greater atmospheric dynamics and the presence of gaseous precursors in the atmosphere facilitated the nucleation processes.

Furthermore, the emission sources in each sampling station conditioned the daily evolution of the aerosol size distribution in each measurement period.

At those stations with a clear influence of traffic emissions, such as A Coruña, Madrid, Barcelona and Granada three peaks were observed in the particle concentration in the summer period: the first peak (between 06:00 and 09:00 UTC approximately) and the third peak (between 18:00 and 00:00 UTC approximately) were attributed to traffic emissions, and the second peak (around 12:00 UTC) corresponded to secondary aerosol formation by photochemical activity. However, during the winter period only the two peaks corresponding to traffic emissions were observed. In these urban stations, the major contributor to total particle concentrations during the winter period was the Aitken mode, while, the increase in nucleation processes had a significant contribution to N_t, especially when NPF occurred, during summer period.

The situation was very different in those stations without a direct influence of anthropogenic emissions. The daily pattern of the aerosol size distribution was quite similar for both study periods. In the El Arenosillo and Montseny stations the particle concentration maximum appeared after 12:00

UTC. In these stations, the transport of the anthropogenic emissions from large urban areas imposed a background particle concentration throughout the day. This meant that the Aitken mode was the dominant particle mode, except during the central hours of the day when the environmental conditions were optimal for NPF development (low concentrations of preexisting particles in the atmosphere, high solar activity and low relative humidity), and consequently these processes were intense.

Only a peak was observed at the Izaña station around 12:00 UTC in the warm and cold periods, which was attributed to nucleation processes. In this station the nucleation mode was the largest contributor to the total particle concentration in both periods.

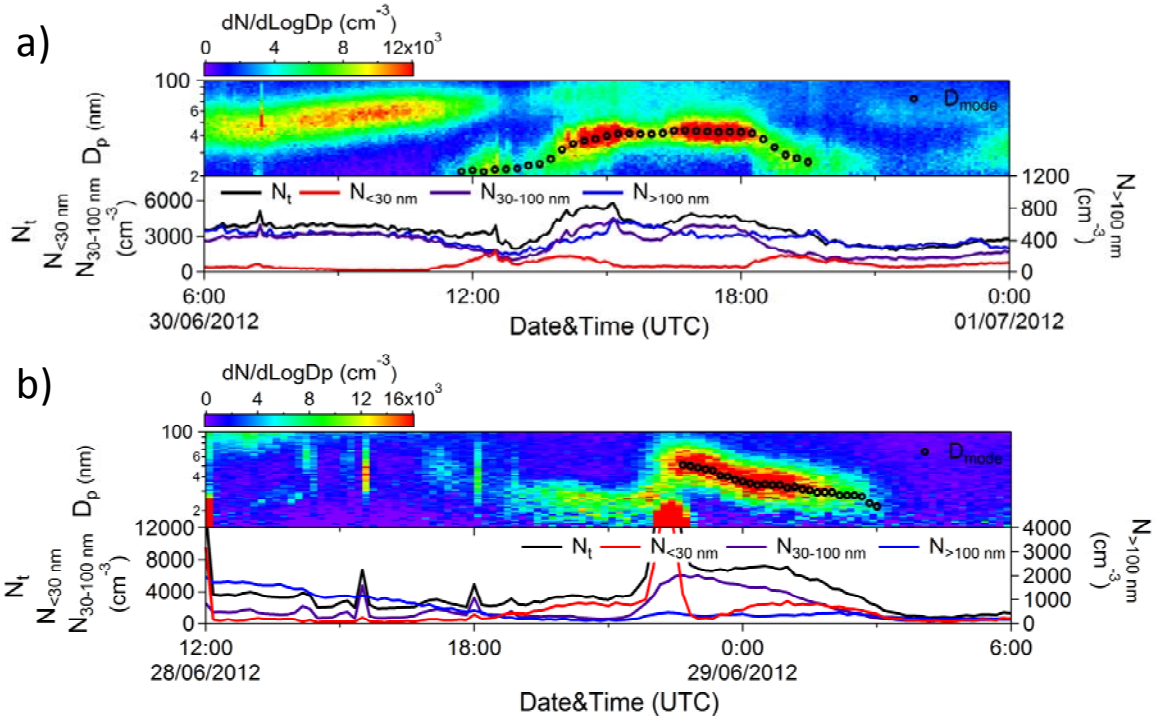

Figure 2. Example of two case studies: a) shrinkage during NPF identified in Madrid station and b) shrinkage event in absence of previous NPF identified in El Arenosillo station. Evolution of the aerosol size distributions, the total number of particles (N_t) and particle concentration for each of the three modes; nucleation ($N_{<30\ nm}$), Aitken ($N_{30\text{-}100\ nm}$) and accumulation ($N_{>100\ nm}$) as well as the particle mode diameter (D_{mode}) for each case study.

3.2. NPF and shrinkage events

NPF events have been observed during the summer period in all sampling stations, when the environmental conditions were optimal for their development.

These processes were particularly common in Izaña station associated with the transport of gaseous precursors from the boundary layer [4]. In Madrid, Montseny and El Arenosillo sites the influence of the biogenic volatile organic compounds (BVOCs) emitted by vegetated areas close to the stations played an important role in these processes [5, 7, 8]. In A Coruña and El Arenosillo the sea breezes had an important role in the NPF development [3, 6, 7].

In addition, these events were also identified during the winter period in Monseny, Izaña and El Arenosillo sites, stations without direct anthropogenic emissions.

The shrinkage events have been observed only during the summer period in Madrid and El Arenosillo stations. Two clear examples of these processes are shown in figure 2. In Madrid station a shrinkage process was identified during the growth phase of the newly nucleated particles on 30 June 2012. However, in El Arenosillo station the shrinkage was identified in the absence of a prior growth process on 28 June 2012. Both case studies occurred in the second half of the day, after 18:00 UTC,

possibly caused by an increase in the wind speed and/or a reduction in the photochemical activity. The SR was -6.2 and -7.8 nm h^{-1} respectively, for each shrinkage process identified.

4. Conclusions

In each sampling site, the pollution sources and meteorological conditions determined the aerosol size distribution characteristics.

In both summer and winter periods, the urban area emissions were the main contributors to the total particle concentration at traffic and background urban stations, while in rural background and high-altitude stations, the nucleation processes had a higher contribution to aerosol particle number concentrations.

In general, the station location determined the behaviour of the particle concentration corresponding to each size distribution mode. At those stations with a higher influence of anthropogenic emissions the Aitken mode was the largest contributor to the total concentration of particles in opposition to less polluted sites, in which it was the nucleation mode.

NPF events occurred during the summer period in all measurement stations and also during the winter period in those with low anthropogenic influence. In addition, some shrinkage processes were identified in two measurement stations, Ciemat and El Arenosillo.

These results are a first approximation to a further study on the patterns of the submicrometer particle size distributions in Spain, which will be developed as part of the project "Atmospheric aerosol properties under different spatial and temporal scenarios and their climate influence (PROACLIM)". In addition, in a later study the influence of the geographic location, the land features and the atmospheric dynamics at regional level of each measurement site on aerosol size distribution and its processes will be examined in detail.

Acknowledgments

This work has been financed by the Ministry of Science and Innovation (CGL2011-15008-E, CGL2010-1777, CGL2011-27020, CGL2014-52877-R & CGL2014-55230-R), Xunta de Galicia (GRC2013-047 potentially cofounded by ERDF) and the European Union Seventh Framework Programme (FP7/2007–2013) ACTRIS under grant agreement no. 262254. E. Alonso acknowledges the FPI grant to carry out the doctoral thesis/PhD at the CIEMAT and M. I. García acknowledges the grant of the Canarian Agency for Research, Innovation and Information Society (ACIISI) co-funded by the European Social Funds.

References

[1] Birmili W *et al* 2009 *Gefahrstoffe Reinhaltung der Luft* **69** pp 137-145
[2] Asmi A *et al* 2011 *Atmos. Chem. Phys.* **11** pp 8893-8976
[3] Iglesias-Samitier S *et al* A14 *Proc. of 2nd Iberian Meeting on Aerosol Science and Technology (RICTA) Tarragona, Spain, 7–9 July 2014* pp 79
[4] García M I *et al* 2014 *Atmos. Chem. Phys.* **14(8)** pp 3865-3881
[5] Cusack M *et al* 2013 *Tellus B* **65** pp 1–19
[6] Sorribas M *et al* 2011 *Atmos. Chem. Phys.* **11(21)** pp 11185-11206
[7] Sorribas M *et al* 2015 *Science of the Total Environment* **511** pp 723-737
[8] Gómez-Moreno F J *et al* 2011 *Atmos. Environ.* **45** pp 3169-3180
[9] Gómez-Moreno F J *et al* B-WG01S2P30 *Proc. of European Aerosol Conference (EAC), Granada, Spain, 2–7 Sept. 2012*
[10] Gómez-Moreno F J *et al* A076 *Proc. of European Aerosol Conference (EAC), Prague, Czech Republic, 1–6 Sept. 2013*
[11] Wiedensohler A *et al* 2012 *Atmos. Meas. Tech.* **5** pp 657-685
[12] Charron A and Harrison R M 2013 *Atmos. Environ.* **37** pp 4109-4119
[13] Dal Maso M *et al* 2005 *Boreal Env. Res.* **10** pp 323
[15] Kulmala M *et al* 2012 *Nature protocols* **7** pp 1651-1667