Aerosol Size Distribution in Rural Areas: the Influence of Ambient Conditions and Wildfires

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Abstract — In rural areas, during the summer, a passive cavity aerosol spectrometer probe PCASP-X was installed to measure number of particles and size distributions. The study focuses on the wildfires registered in the province and their influence onto particle size distributions. The transport of the particles generated by biomass combustion to the sampling site has enabled us to carry out comparative studies of total measurements and non-contaminated measurements. It was found that the number of particles with sizes ranging from 0.2 to 0.7 μm (fine mode) increased more than 500% in measurements influenced by wildfires. A more detailed analysis of this influence was carried out in three different moments: before, during and after the smoke plume crossed the study zone. The results show that the geometric diameter of the fine mode in the measurements affected by a fire decreases with respect to the previous measurements and reaches average values of 0.11 μm. However, when the relative humidity, is high (more than 80%), the aerosol sizes increase 50%. Consequently, we argue that wildfires not only influence the number of particles, with a clear increase in the number of aerosols in the atmosphere, but they also alter their size distributions.

Keywords — Aerosol size distribution, Fine mode, Weather types, Wildfires

1 INTRODUCTION

The size of atmospheric aerosols depends to a great extent on the sources and sinks, as well as on the meteorological processes occurring during their life time [1], [2], [3], [4]. Biomass burning is one of the most important contributors of aerosols to the atmosphere releasing large amounts of particles and gases, and causing alterations in atmospheric composition at a local and at a global scale. The effects aerosols may trigger in the atmosphere depend on the type of material burnt, the combustion phase, the relative humidity and the wind conditions [5]. A growing number of studies focus on these changes, since aerosols undergo complex interactions in the atmosphere and condition global energy balance [6].

Many studies find an increased number of aerosols in the fine or accumulation mode during wildfires [7], [8], [9]. An in-depth analysis of the emissions released by biomass burning can be found in Reid et al. [5]. This study showed that out of all the particles released during these phenomena, between 80% and 90% correspond to this mode, with a count median diameter (CMD) of 0.13 μm. These particles are mainly composed of organic material; inorganic elements account for approximately 10% of the mass only [7].

The water-absorption capacity of these particles varies greatly. The hygroscopicity of aerosols originated by biomass burning depends on the internal composition of the organic and inorganic material of the sub-micrometer particles [10], [11], [12]. In consequence, studies on aerosol size distributions must take into account hygroscopicity. The ability of aerosols to absorb water vapor from the atmosphere is an important feature, but one difficult to study [13]. The water content in the particles affects factors such as size, total mass, acidity, the amount of water-soluble substances they contain, light-dispersion properties, chemical reactivity, the ability to function as condensation nuclei in a cloud and their permanence in the atmosphere.

The ability of aerosols generated by biomass burning to form condensation nuclei has been the focus of several studies. Authors such as Warner and Twomey, [14], Eagan et al. [15] and Roberts et al. [16] have found that, at supersaturations higher than 0.5%, the particles generated by wildfires function as condensation nuclei (CCN). More recently Petters et al. [17] have studied the hygroscopic properties of aerosols freshly emitted from laboratory biomass burning experiments. They conclude that at the point of emission, most particles are CCN active and do not depend upon conversion in the atmosphere to more hygroscopic compositions before they can participate in cloud formation and undergo wet deposition.

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In the case of wildfires, the source of the particles is well-defined and located, but in the study of aerosol size distributions we must take into account the origin and the type of air mass that carries the particulate matter when it reaches the sampling site.

In this paper we will analyze the characteristics of particle size distributions in the summer months and the changes in these distributions caused by the arrival of aerosols from biomass burning, mainly wildfires affecting lower vegetation, but sometimes also trees. These wildfires are particularly frequent in the rural study zone. The wildfires considered are all the fires registered in the province of León. The distance with the probe installed at the sampling site varies between a few kilometres and a maximum of 70 km.

2 STUDY ZONE

The measurements have been carried out in the district of Carrizo de la Ribera, in the middle of the province of León, Spain, north-west of the capital. This rural area lies at 873 m above sea level and has 2,554 inhabitants. Farming is the main economic activity, and hop is the most widely grown crop. The weather is controlled by a station from the Spanish National Weather Agency (AEMET) installed in that town (42° 35′ N, 5° 39′ W).

The study zone lies on the left bank of the River Órbigo, over a fluvial terrace. The climate is of the continentalized Mediterranean type, with a marked seasonality. Precipitation is scattered irregularly along the year, with minimum values in the summer and the highest values in spring and autumn.

Temperatures are fresh, with an annual average of 10.5°C. The winters are cold, with frequent frosts. The summers are warm, with maximum temperatures that may be over 35°C, although nocturnal temperatures are moderate, as it is an area of irrigated farming systems where land is mainly watered by flooding.

In the summer the province of León is one of the areas most severely affected by wildfires, both in the number of fires and in the land area burnt. Wildfires occur mainly in August and September, and affect mostly forest vegetation not including trees.

3 MATERIALS AND METHODS

A laser spectromer (Passive Cavity Aerosol Spectrometer Probe, PMS Model PCASP-X) was installed in a field 2 km from the town (42° 35′59.90′′ N, 5° 50′50.92′′ W). This device measures particle sizes ranging between 0.1 and 10 μm, considering the light-dispersion properties of the particles in a wave length of 633 nm between the angles of 35° and 135°. The spectrometer measures 31 channels, i.e., 31 discreet particle size intervals. The probe was calibrated by the manufacturer using polystyrene latex particles of a known size. The refractive index of latex beads (1.59 – 0i) is different from that of atmospheric particles, resulting in an aerosol size distribution that is "latex equivalent".

This study presents PCASP-X size distributions corrected using Mie theory and implemented with a computer code developed by Bohrenm and Huffman [18], according to the refractive index typical in rural aerosols. The value varies depending on the relative humidity [19]. The refractive indices were calculated interpolating the real part and the imaginary part according to the relative humidity at the time of measurement (variable between 17% and 100%).

It was also necessary to carry out a number of corrections on the number of counts sampled by the spectrometer in each channel. First, the flow measurement value was fixed according to the altitude of the sampling point; the probe was installed at 834 m introducing a correction factor of 0.905. In addition, each measurement was corrected according to the activity registered. Finally, the last fit is determined by the duration of the samplings, which was of 600 seconds. These corrections are described in more detail in Calvo et al. [6].

The study period covers the 123 days of the months of June, July, August and September of the year 2000. Eight measurements were carried out daily of the ambient particle size spectrum in that rural area, with 10-minute intervals every 3 hours.

The probe was installed next to a weather station to register automatically data on precipitation, pressure, temperature, relative humidity and wind speed and direction. A wind profiler was also used (Sodar SR1000), with a pulse frequency of 5 tones, around 2,150 Hz, pulse power of 300 W, a pulse-repetition period of 8 s and a maximum range of 1,250 m. This device registered automatically data from the three wind components. The data registered by the weather station and the data registered by the Sodar were all stored on a computer every 30 minutes.

The thermal inversions have been calculated using data from soundings in La Coruña (43.36°N, 8.41°W, altitude 67 m) and Madrid (40.50°N, 3.58°W, altitude 633m), provided by the University of Wyoming. Access to data is on the website http://weather.uwyo.edu/upperair/sounding.html.

A Circulation Weather Type classification

(CWTs) was set up based on Jenkinson and Collison [20] and Jones et al. [21] to identify the type of weather associated to each particular synoptic situation. These procedures were developed to define objectively Lamb Weather Types [22] for the British Isles. The daily circulation affecting the Iberian Peninsula is described using a set of Indices associated to the direction and vorticity of the geostrophic flow. The Indices used were the following: southerly flow (SF), westerly flow (WF), total flow (F), southerly shear vorticity (ZS), westerly shear vorticity (ZW) and total shear vorticity (Z). These Indices were computed using sea level pressure (SLP) values obtained for the 16 grid points scattered over the Iberian Peninsula. This method allows for a maximum of 26 different CWTs. Following Trigo and Cámara [23] in their study for Portugal, this study did not use an unclassified class, but rather opted for disseminating the fairly few cases with possible unclassified situations (<2%) among the retained classes. This classification has already been used in the Iberian Peninsula for different applications, such as the study of lightning [24], splash erosion [25] or aerosol size distribution in rain events [26].

The Department for the Environment of the regional government (Consejería de Medio Ambiente de la Junta de Castilla y León) provided the database with the number of wildfires, the district where each fire occurred, the date of detection and extinction (with exact date and time), the land area affected in hectares (ha), and the type of vegetation burnt during the summer months (June, July, August and September). This information was used to draw for each day a map of the province of León highlighting all the districts affected by wildfires.

This material and the data on wind direction at surface level and at a certain altitude, registered by the weather station and the Sodar, was used to identify the measurements that could have been affected by the transport of the smoke plumes from any nearby wildfires. The changes in the number of particles revealed the arrival of a plume at the sampling site. In general, a plume affected only 1 out of the 8 measurements carried out daily, as these measurements were carried out every 3 hours. However, in the case of large fires several measurements may be affected in one day or even over several subsequent days. The particle measurements of the 8 daily registers were compared with the time interval of the wildfire, from detection to extinction. The measurements with very large numbers of particles were thus identified. In some cases there were average increases of 2,000% with respect to the other measurements registered on that same day. After identifying the measurements that were possibly affected by the arrival of a smoke plume to the probe, the data were confirmed with an analysis of the wind direction and speed and the distance between the wilfire and the probe. The increases in the measurements that could not be explained by these variables were excluded from the study. Two different samples were generated from the information obtained: one group corresponds to measurements affected by the smoke plumes of wildfires (41 measurements in 31 different days out of the 123 study days); the other group includes non-affected measurements (943 measurements in 122 different days out of the 123 study days).

The aerosol size distribution and the daily CWT were analyzed separately in both groups.

In the measurements that were not affected by the wildfires the study focused on the influence of the type of weather onto the number of particles and the count median diameter of the fine mode. Soundings provided data on thermal inversions, both radiative and subsidence inversions, on the days where the number of particles was higher than average. In addition, a detailed analysis of several factors was carried out, including the geometric diameter, the geometric deviation of the diameter, the number of particles and the relative humidity at the time of measurement.

The distributions found in the measurements affected by wildfires were studied in detail and compared with total measurements and measurements not affected by fires. The evolution of the acummulation mode and of the coarse mode has been analyzed too, as well as the relative humidity at three different stages: in the measurements before the ones affected by the plumes from the fires, in the ones affected by the fires, and in the ones after the fires.

4 RESULTS AND DISCUSSION

4.1 Meteorological study and Circulation Weather Types

During the study period (June, July, August and September 2000) the average monthly temperatures are typical of the summer months. The highest value is reached in July with 17.1°C. The maximum temperatures are high in all four months, higher than 30° C, and the minimum temperatures are around 2°C.

The monthly precipitation accumulated in the study period is low. The highest value was reached in September with 26.8 mm. The average relative humidity is around 65%, with 71% in September. The average wind speed was low, with a monthly average of less than 2 m/s. The high relative humidities registered, mainly during the night, are due to the irrigation system used in the fields close to the town and also to the fact that the River Órbigo flows close by at a distance of around 2.5 km.

The CWT classification shows that during the months from June to September, the dominant CWTs were the following: the purely north-eastern type (NE), the anti-cyclonic type (A) controlled by the geostrophic vorticity, and the purely northern type (N), with 24, 19 and 15 days, respectively. The purely directional weather types are also relatively frequent: the Eastern type (E), the Western type (W) and the North-western type (NW), two hybrid types with northern direction (AN) and north-eastern direction (ANE), and one non-directional type, the cyclonic type (C). During the summer of the year 2000 the anti-cyclonic weather type and the flows with a northern component were dominant over the Iberian Peninsula, following the same trend as the one found in other studies on weather types and precipitation in the whole province of León [27].

4.2 Study of aerosol size distributions and influence of the weather type

4.2.1 Analysis of weather type

In order to study the influence of the weather types onto the number of aerosols in the study zone we analyzed the measurements that were not contaminated by aerosols from the wildfires. For each weather type we studied the average number of total particles and the standard deviation, as well as the number of days with those weather types (Fig. 1). 30% of the study days (32 out of 122) present an average content of particles.cm^{-3} higher than 2,000 and the weather types registered were the anti-cyclonic type (A), northern anti-cyclonic (AN), western anti-cyclonic (AW), south-western anti-cyclonic (ASW), the north-western cyclonic type (CNW) and the purely western type (W). However, the standard deviation observed in the number of particles varies greatly in the anti-cyclonic type (A), the northern hybrid (AN), and the purely western type (W). In other words, the anti-cyclonic situations favor the presence of a higher number of aerosols, with the arrival of maritime air masses. On the other hand, the following weather types have little influence in the number of aerosols, as they register mean values of less than 1000 particles.cm^{-3}: eastern anti-cyclonic (AE), north-eastern and north-western anti-cyclonic (ANE & ANW), eastern, south-eastern and south-western cyclonic types (CE, CSE & CSW), and the purely directional southern, south-eastern and south-western types (S, SE & SW). The standard deviations are also very low, except in the north-eastern anti-cyclonic weather type (ANE). These weather types account for 18% of the days and favor the arrival of continental air masses to the Iberian Peninsula. In the remaining weather types the average values lie between 1,000 and 2,000 particles.cm^{-3}.

A comparative analysis was carried out on the relationship between the count median diameter of the fine mode (CMD_f) and the weather types (Table 1). Four size ranges have been defined to enable us to interpret the results more easily. The particles with diameters of less than 0.13 μm are mainly detected in cyclonic weather types with southern and western wind components. The air masses that reach the Iberian Peninsula in these situations come from the north of Africa (Saharan air masses) or from the Atlantic Ocean, carrying with them marine aerosols. The particles with CMD_f sizes between 0.13 and 0.14 μm show a dominant component from the north and the west, although anti-cyclonic situations generate particles in this size range too. These weather types show a clear influence of maritime and continental European air masses. In the case of diameters between 0.14 and 0.15 μm, the weather types found are the north-eastern anti-cyclonic type and the north-western cyclonic type, i.e., the air masses that arrive at the Iberian Peninsula are both maritime and continental European air masses. The largest diameters (> 0.15 μm) correspond to continental air masses (CNE), and the Eastern anti-cyclonic weather type (AE) with maritime aerosols from the Mediterranean.

Table 1. Count median diameter of the fine or accumulation mode (CMD_f) for each Circulation Weather Type.

CMD_f (μm)	**CIRCULATION WEATHER TYPE**
< 0.13	ANW-ASW-AW-C-CE CS-CSE-CSW-E-S-SW
0.13 – 0.14	A-AN-CW-N-NE-NW-SW-W
0.14 – 0.15	ANE-CNW
> 0.15	AE-CNE

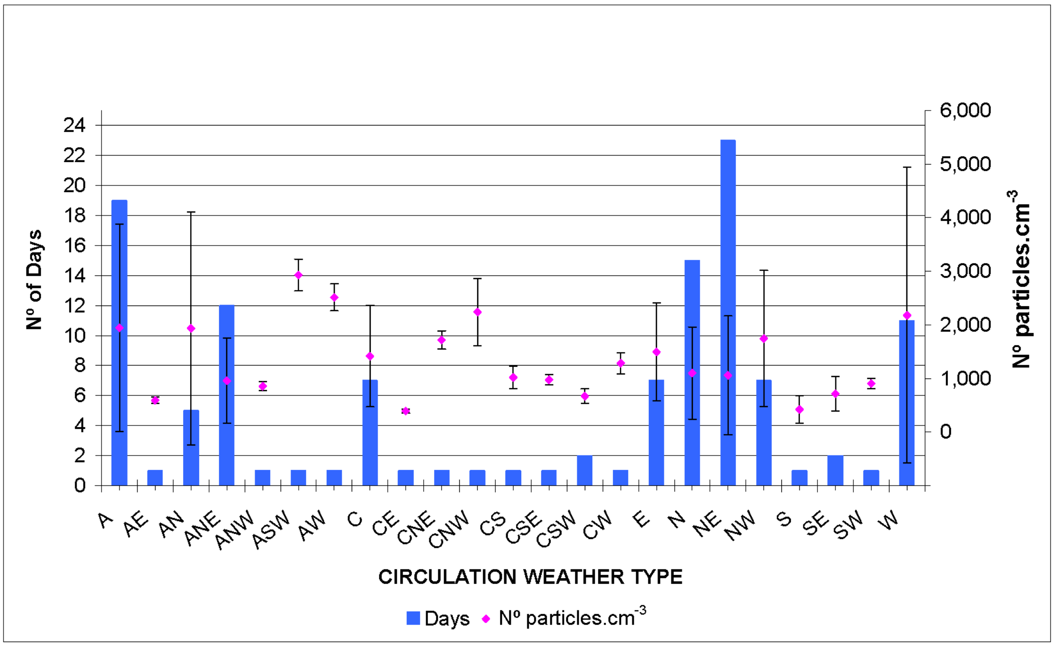

Fig. 1. Number of particles registered and standard deviation for each Circulation Weather Type in 122 days in the months of June, July, August and September in accordance with the days with each particular weather type.

Air masses from the north of Africa bring a higher number of particles, but of smaller sizes (smaller than 0.13 µm) than the ones brought to the Iberian Peninsula by air masses coming from continental Europe (larger than 0.13 µm). With the weather types typical of maritime air masses the sizes registered show that, in general, the aerosols are smaller than 0.14 µm. In other words, even though continental air masses contribute fewer particles than maritime air masses, the aerosols they carry are larger. The arrival of European air masses has been studied by authors such as Alonso et al. [28], Gangoiti et al. [29], Viana et al. [30] or, more recently, by Escudero et al. [31].

4.2.2 Temporal evolution of the number of particles

Fig. 2 shows the daily evolution of the number of particles and the standard deviation during the whole study period. In general it was found that the average number of particles registered daily is lower than 2,000 particles.cm^{-3}, with big differences in the average numbers. There were two periods at the end of the summer with oscillations between 2,000 and 7,000 particles.cm^{-3} (second half of August and second half of September). This situation leads to a gradual increase in the number of particles, as well as in the standard deviation. During the 27 days comprised by these periods there were frequent thermal inversions at altitudes lower than 1,000 meters (AGL), both radiative and subsidence inversions. Both types of inversion often occurred the same day. These inversions hinder the dispersion of aerosols, so their number remains high in the lower layers of the atmosphere, increasing up to 70% when compared with the other study days. The days with more than 2,000 particles cm^{-3} correspond mainly to the anti-cyclonic weather type (A), registered in six study days, followed by the cyclonic type (C), the purely directional northern (N) and north-easter types (NE), with three days each. The northern anti-cyclonic type (AN) and the purely eastern (E), northern (N), western (W) and north-western (NW) types occurred in two days each. Finally, the south-western anti-cyclonic type (ASW), the northern anti-cyclonic type (AN) and the north-western cyclonic type (CNW) were found in one day each. It can therefore be argued that the increase in the daily number of particles in the months of August and September was not only motivated by the thermal inversions those days, but there was a clear influence of the weather types too, since the anti-cyclonic and the northern type register high numbers of aerosols.

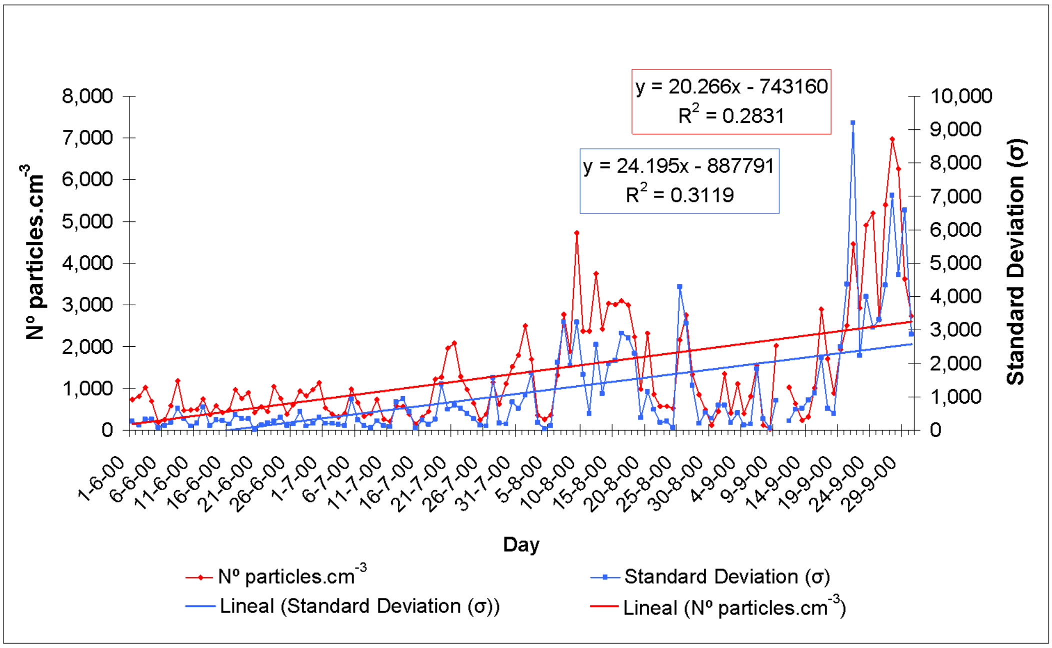

Fig. 2. Average number of particles and standard deviation in the months of June, July, August and September of the year 2000.

Fig. 3 shows the evolution of the geometric mean diameter (D_g) of the particles along the hours of measurement, relating this parameter to the geometric deviation (σ_g), the relative humidity registered (HR) and the total number of particles at those times. The trends found illustrate a clear difference during the summer between night (between 1900 UTC, beginning of dusk, and 0400 UTC, beginning of dawn) and day (from 0400 UTC to 1900 UTC). The geometric mean of the diameter during the night presents a growing trend reaching 0.16 μm at 0400 UTC. During this time interval, the geometric deviation of the diameter is high, i.e., during the night large and small particles coexist. However, from 0400 UTC the geometric mean of the diameter decreases until a size of around 0.12 μm at 1300 UTC and then remains stable until 1900 UTC, when it rises again. The geometric deviation of the diameter is low, so the particles registered during the day present homogeneous sizes.

It was found that the relative humidity follows a trend parallel to particle growth. During the night the relative humidity values registered are higher than 85% due to the irrigation system in the area. Many canals transfer water from the River Órbigo to the different farming areas and this fact seems to be influencing particle sizes. From around 0400 UTC the relative humidity decreases to 45% and there is a parallel decrease in the size of the aerosols. Humidity and particle sizes started increasing around 1600 UTC.

During the days studied no sudden changes have been noticed in the air masses that could explain these oscillations in size, nor was there any

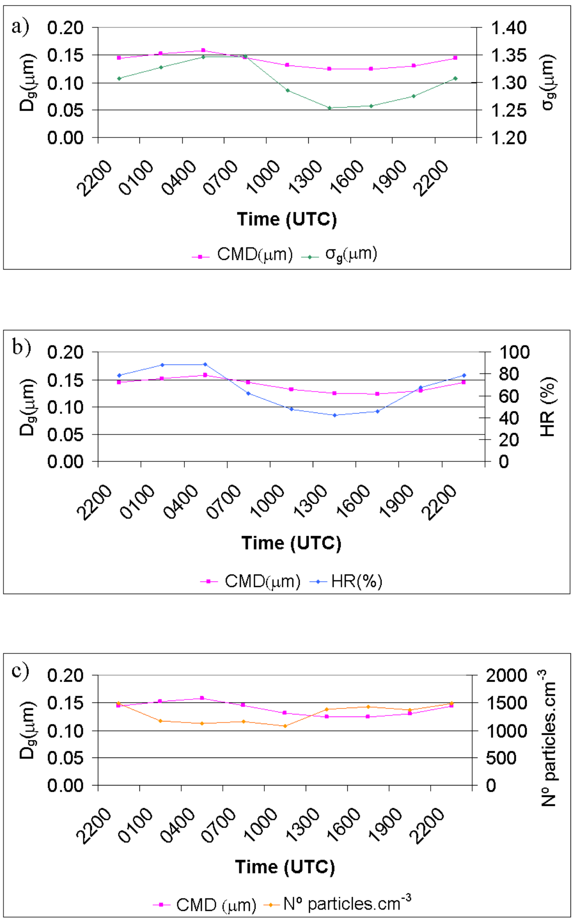

Fig. 3. (a) Geometric mean diameter (D_g) of the particles during the 8 daily hours of measurements and its geometric deviation (σ_g), (b) relative humidity (HR) and (c) total number of particles at those times.

anthropogenic contribution of new particles with larger sizes because there is no industry in the area. The measurements affected by wildfires were not considered either. The increase in particle size is therefore conditioned by the relative humidity in the ambient atmosphere, and there is a hygroscopic growth of these particles caused by the absorption of water vapor from the air.

As for the number of particles registered, the study zone is a rural area and there are no anthropogenic contributions of aerosols, so the number of particles remains stable throughout the whole day, oscillating between 1,000 and 1,500 particles.cm^{-3}. The number of particles is slightly lower during the night, because the wind is weaker and aerosols are deposited on the ground. During the day, in contrast, the effect of the wind on the ground results in a slight increase in the number of aerosols due to the contribution of soil particles. However, on days with some of the measurements affected by fires, average values of more than 7,000 particles cm^{-3} have been registered.

4.3 Influence of wildfires on aerosol size distribution measurements

The influence of wildfires onto aerosol size distributions was studied by taking two samples: on the one hand, the measurements not affected by the fires, taken before and after the plume crossed the study zone; and on the other hand, a sample of measurements affected by the smoke plume from wildfires in a radius of around 70 km. Comparative analyses were carried out using these data to identify changes in atmospheric aerosols and their evolution during these events.

The comparative analysis between the monthly aerosol size distributions including all measurements and including only data not contaminated by wildfires (Fig. 4) shows a clear decrease in the number of particles smaller than 1 µm in the latter case. This decrease is more dramatic in the months of August and September, a period with more wildfires and more land area affected. In the months of June and July, especially in June, most of the fires registered were very small (less than 1 burnt) or small, affecting less than 10 ha. Wildfires contribute to the increase in the number of aerosols between 0.2 µm and 0.7 µm. There is a homogenization of the monthly data not contaminated by the registers of aerosols from wildfires: particles larger than 1 µm, corresponding to the coarse mode, remain constant. Fig. 5 shows a more detailed comparative study of the relationship between aerosol size distributions in the measurements before and after the ones affected by the wildfire, on the one hand, and the distributions in the measurements affected by wildfres over the whole study period. A significant increase of around 500% was found in

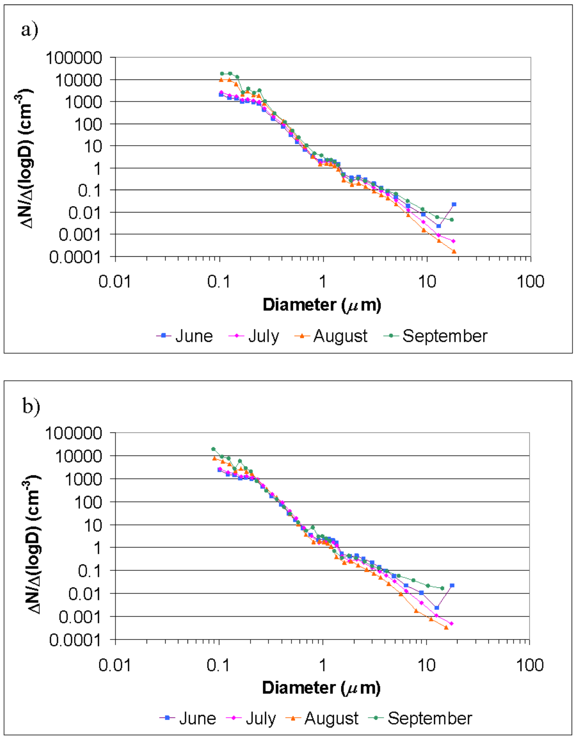

Fig. 4. Mean size distributions in the months of June, July, August and September 2000 in (a) all the measurements registered (with and without wildfires) and (b) measurements not contaminated by wildfires.

this time span in the number of particles in the fine mode (particles between 0.1 µm and 1 µm).

It is the number of particles in the fine mode that increases, so to investigate the sizes of these particles when the probe is measuring aerosols from wildfires, we carried out a comparative analysis of the mean of the CMD of the fine or accumulation mode before, during and after the arrival of the smoke plume from the wildfires. The influence of humidity on particle growth was also analyzed. The measurements carried out during the wildfire when compared with the ones immediately before show a clear decrease in the size of particles of around 18%, from average CMD_f values of 0.13 µm to 0.11 µm. However, it was found that when the relative humidity registered in this measurement affected by aerosols from wildfires is higher than in the previous measurement, around 80%, the mean of the geometric diameter rises from 0.10 µm to 0.14 µm. In these cases we argue that aerosols derived from biomass combustion undergo a hygroscopic growth in high relative humidities because of the absorption of atmospheric water vapor, thus increasing their size. Already in 1959, Orr et al. [32] demonstrated that when the relative humidity (RH) rises above 40%, even weakly soluble aerosol particles can absorb water from the air. As a consequence, this additional water increases the particle size. After the smoke plume the CMD tends to stabilize and returns to the values before the arrival of the plume.

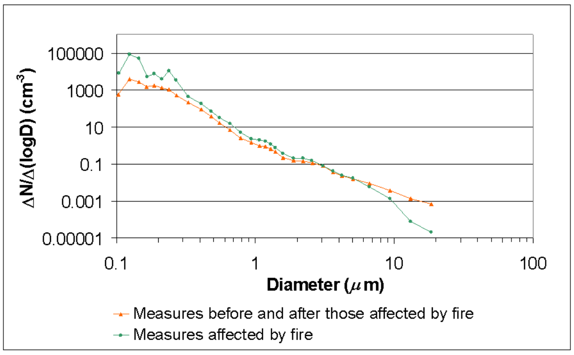

Fig. 5. Mean size distributions of the measurements carried out immediately before and after those affected by wildfires and the measurements affected during the months of June, July, August and September 2000.

5 CONCLUSIONS

As for the average number of particles in each weather type, it may be said that the air masses that register more than 2,000 particles cm^{-3} in the weather types A, AN, AW, ASW, CNW and W are maritime air masses. In contrast, the weather types that register fewer than 1,000 particles cm^{-3} (AE, ANE, ANW, CE, CSE, CSW, S, SE and SW) are influenced by continental air masses.

Air masses from the north of Africa contribute a larger number of particles, but of smaller sizes (smaller than 0.13 µm), than the ones contributed by air masses from continental Europe (larger than 0.13 µm). With weather types with maritime air masses the size range registered indicates that the aerosols are usually smaller than 0.14 µm. Consequently, even though continental air masses contribute fewer particles than maritime air masses, these particles are larger.

The average number of particles (excluding the ones generate by biomass combustion) registered daily during the study period is lower than 2,000 particles cm^{-3}, with big differences between the measurements. In the months of August and September up to 7000 particles cm^{-3} were registered.

The particles registered during the night are larger than the ones registered during the day and the latter have more uniform sizes. The size increase of the particles during the night is influenced by the relative humidity registered in those hours, higher than 85%. There is a hygroscopic growth of the particles by absorption of water vapor from ambient atmosphere. The water irrigation system is partly responsible for the increase in ambient humidity. In addition, the number of particles remains constant during the day, when no wildfires are raging nearby, with values between 1,000 and 1,500 particles cm^{-3}.

With respect to the influence of wildfires onto aerosol size distributions, it may be concluded that wildfires do have an effect on the number of particles in the atmosphere not only by increasing their number, but also by altering aerosol size distributions increasing the number of particles in the fine mode (particles between 0.1 µm and 1 µm) by around 500%, mainly the ones in the size range between 0.2 and 0.7 µm. The measurements affected by wildfires present aerosols with average CMD_f around 0.11 µm, a value lower than the ones registered in measurements not affected by a fire. However, the relative humidity registered increases in the measurements affected by the smoke plume when compared with the ones not affected (HR around 80%). There is a 50% increase in the CMD of the fine mode, and this fact is due to a hygroscopic growth of the aerosols absorbing water vapor.

The authors consider it necessary to keep this research line open, since the devastating consequences of wildfires cause immediate alterations in atmospheric composition at local and/or regional scales. Moreover, the increase in the number of aerosols causes changes in the global radiance balance of the Earth, generating direct as well as indirect radiative forcings. The sequels may be latent, causing an increase in the albedo in the areas devoid of vegetation, favoring these processes.

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REFERENCES

[1] Suzuki, T. and S. Tsunogai, 1988. Daily variation of aerosols of marine and continental origin in the surface air over a small island Okushin in Japan sea. *Tellus*, 40, 42–49.

[2] Ito, T., 1993. Size distribution of Antarctic submicron aerosols. *Tellus,* 45, 145–159.

[3] Castro, A., Marcos, J.L., Dessens, J., Sánchez, J.L., Fraile, R., 1998. Concentration of Ice Nuclei in Continental and Maritime Air Mases in León (Spain). *Atmospheric Research*, 47–48, 155–167.

[4] Fraile, R., Calvo, A.I., Castro, A., Fernández-González, D., García-Ortega E., 2006. The behavior of the atmosphere in Long-Range transport. *Aerobiologia*, 22, 35–45.

[5] Reid, J.S. Eck, T.F., Christopher, S.A., Koppmann, R., Dubovik, O., Eleuterio, D.P., Holben, B.N., Reid, E.A., Zhang, J., 2005. A review of biomass burning emissions part III: intensive optical properties of biomas burning particles. *Atmos. Chem. Physic.* 5, 827–849.

[6] Calvo, A.I., Pont, V., Castro, A., Mallet, M., Palencia, C., Roger, J.C., Dubuisson, P., Fraile, R., 2010. Radiative forcing of haze during a forest fire in Spain. *J. Geophys. Res.* 115, D08206, doi:10.1029/2009JD012172.

[7] Cachier, H., Liousse, C., Buatmenard, P., and Gaudichet, A., 1995. Particulate content of savanna fire emissions. *J. Atmos. Chem.* 22, 123–148.

[8] Remer, L. A., Kaufman Y. J., Holben B. N., Thompson A. M., and D. McNamara, 1998. Biomass burning aerosol size distribution and modeled optical properties. *J. Geophys. Res.* 103 (D24), 31,879–31,891.

[9] Reid, J. S. and Hobbs, P. V., 1998. Physical and optical properties of smoke from individual biomass fires in Brazil. *J. Geophys. Res.* 103, 32,013–32,031.

[10] Cocker, D.R., Whitlock, N.E., Flagan, R.C., 2001. Hygroscopic properties of Pasadena, California aerosol. *Aerosol Sci. Technol.* 35, 637–647.

[11] Vakeva, M., K. Hameri, T. Puhakka, E.D. Nilsson, H. Holiti and J.M. Makela, 2001. Effects of meteorological processes on aerosol particle size distribution in an urban background area. *J. Geophys Res.* 105, 9,807–9,821.

[12] Aklilu, Y., Mozurkewich, M., Prenni, A.J., Kreidenweis, S.M., Alfarra, M.R., Allan, J.D., Anlauf, K., Brook, J., Leaitch, W.R., Sharma, S., Boudries, H., Worsnop, D.R. 2006. Hygroscopicity of particles at two rural, urban influenced sites during Pacific 2001: Comparison with estimates of water uptake from particle composition. *Atmos. Environ.* 40, 2,650–2,661.

[13] Kerminen, V-M. 1997. The effects of particle chemical character and atmospheric processes on particle hygroscopic properties. *J. Aerosol Sci.* 28:1, 121–132.

[14] Warner, J. and Twomey, S., 1967. The production of cloud nuclei by cane fires and the effect on cloud drop concentrations. *J. Atmos. Sci.* 24, 704–713.

[15] Eagan, R. C., Hobbs, P. V., and Radke, L. F., 1974. Measurements of cloud condensation nuclei and cloud droplet size distributions in the vicinity of forest fires. *J. Appl. Meteor.* 13, 553–557.

[16] Roberts, G. C., Artaxo, P., Zhou, J., Sweitlicki, E., and Andreae, M.O., 2002. Sensitivity of CCN spectra on chemical and physical properties of aerosol: A case study from the Amazon basin. *J. Geophys. Res.* 107, doi:10.1029/2001JD000583.

[17] Petters, M. D., C. M. Carrico, S. M. Kreidenweis, A. J. Prenni, P. J. DeMott, J. L. Collett Jr., and H. Moosmüller, 2009. Cloud condensation nucleation activity of biomass burning aerosol. *J. Geophys. Res.* 114, D22205, doi:10.1029/2009JD012353.

[18] Bohren, C. F.; Huffman, D. R., 1983. Absorption and Scattering of Light by Small Particles. Wiley, New York.

[19] Kim, Young J. and Boatman, Joe F., 1990. Siza Calibration Corrections for the Active Scattering Aerosol Spectrometer Probe (ASASP-100X). *Aerosol Science and Technology*, 12:3, 665–672.

[20] Jenkinson AF, Collison FP., 1977. An initial Climatology of gales over the North sea. Synoptic Climatology Branch Memorándum, 62, Meteorological Office, London.

[21] Jones P.D., Hulme M., Briffa K.R., 1993. A comparison of Lamb circulation types with an objective classification scheme. *International Journal of Climatology.*13, 655–663.

[22] Lamb HH., 1972. British Isles weather types and a register of daily sequence of circulation patterns, 1861-1971. Geophysical Memoir 116, HMSO, London.

[23] Trigo R.M. and DaCamara C. C., 2000. Circulation Weather Types and their Influence on the Precipitación Regime IN Portugal. *Int. J.* Climatol. 20, 1559–1581.

[24] Tomás, C., De Pablo, F., Rivas Soriano, L., 2004. Circulation weather types and cloud-to-ground flash density over the Iberian Peninsula. *Int. J. Climatol.* 24, 109–123.

[25] Fernandez-Raga, M., Fraile, R., Keizer, J.J., Varela-Teijeiro, M.E., Castro, A., Palencia, C., Calvo, A.I., Koenders, J., Marques, R.L., 2010. The kinetic energy of rain measured with an optical disdrometer: an application on splash erosion. *Atmospheric Research*, 96, 225–240.

[26] Castro A., Alonso-Blanco E., González-Colino M., Calvo A. I., Fernández-Raga M., Fraile R., 2010. Aerosol size distribution in precipitation events in León. Spain. *Atmos. Res.* 96, 421–435.

[27] Fernández-González, S., Del Río, S., Castro, A., Penas, A., Fernández-Raga, M., Calvo, A.I., Fraile, R., 2009. NAO, weather types and precipitation in León. 5h European Conference on Severe Storms, 12–16, Landshut (GERMANY).

[28] Alonso, L., Gangoiti, G., Navazo, M., Millán, M.M., Mantilla, E., 2000. Transport of tropospheric ozone over the Bay of Biscay and the Eastern Cantabrian coast of Spain. *J. Appl. Meteorol.* 4, 475–486.

[29] Gangoiti, G., Alonso, L., Navazo, M., Albizuri, A., Pérez-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. *Atmos. Environ.* 36, 1,349–1,361.

[30] Viana, M., Querol, X., Alastuey, A., Gangoiti, G., Menendez, M., 2003. PM levels in the Basque Country (Northern Spain): analysis of a 5 yr data record and interpretation of seasonal variations. *Atmos. Environ.* 37, 2,879–2,891.

[31] Escudero, M., Querol, X. Ávila A., Cuevas, E., 2007. Origin of exceedances of the European daily PM limit value in regional background areas of Spain. *Atmos. Environ.* 41, 730–744.

[32] Orr, C., Hurd, F. K., and Corbett, W. J., 1959. Aerosol size and relative humidity. *J. Colloid Sci.* 13, 472–482.