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Influence of the trajectory of air masses on the in situ measurements of aerosol properties in Valencia

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Abstract

We analyzed the influence of the trajectory of air masses on the in situ measurements of aerosol properties (scattering and backscattering coefficients and scattering Ångström exponent) in Valencia (Spain) for the March 2006 to December 2009 period. The scattering (σ_{sp}) and backscattering (σ_{bsp}) coefficients were measured with a TSI 3563 integrating nephelometer. From the σ_{sp} and σ_{bsp} measurements, it is also possible to calculate the scattering Ångström exponent (α_s). By classifying the source regions of air masses by sectors, it has been possible to establish the dependence of aerosol properties on the type of dominant air mass, proving that in situ measurements of aerosol properties are parameters that are sensitive to the type of air mass. The highest values of scattering and backscattering coefficients are obtained under the influence of masses from North Africa and the European continent, while the lowest are obtained for Arctic-type air masses. The scattering Ångström exponent presents its highest values under the influence of European-type masses, while the lowest values are obtained for Arctic and Polar type masses.

Key words: aerosol properties, air masses, in situ measurements

1 Introduction

An air mass is usually defined as a volume of air whose physical properties (mainly temperature and humidity) remain relatively constant across surfaces of hundreds to thousands of square kilometers (Barry and Chorley, 1998). Air masses acquire their characteristics when interacting with the surfaces above which they are located (source regions), where they must remain stationary for a certain time. The source regions should also be sufficiently large and homogeneous to be able to print their characteristics on the air mass.

Although the traditional definition of air mass does not take into account the properties of the particles suspended in it, it is reasonable to assume that the continuous interaction between the surface of the source region and the air mass will also determine the nature and concentration of its aerosols. This is especially true when considering that the average life of particles in the troposphere is 5 to 10 days, thus an air mass that remains for several days over the same region will have much of its previous aerosol load replaced by that of the new corresponding source region. Actually, it is likely that air masses are not pure masses, either because of having undergone a homogeneous or heterogeneous mixture with masses of other features, or because they have spent some time traveling over other regions.

A way to study the relationship between the type of air masses and aerosols is to calculate the historical or back trajectory followed by the air parcel that is over a region at a given moment. Some studies have used back trajectories to analyze the origin of air masses prevailing in a given time, and thus understand the origin of the carried particles (Grousset et al., 2003; Niemi et al., 2004; Estellés et al., 2007a; Toledano et al., 2009). In a more limited number of cases, back trajectories have been classified in order to understand the role of transportation in local aerosol climatology (Birmili et al., 2001; Gerasopoulos et al., 2003; Slater and Dibb, 2004). Other studies have used cluster analysis as a method of classifying air masses (Dorling et al., 1992; Mattis et al., 2000; He et al., 2003; Rozwadowska et al., 2010).

In this paper, we have analyzed the influence of the trajectory of air masses on the *in situ* measurements of atmospheric aerosol properties made in Valencia (Spain) with a TSI 3563 integrating nephelometer for the March 2006 to December 2009 period. By classifying the source regions of air masses into sectors, and with the help of a simple model (Estellés et al., 2007a) that quantifies the relative influence of each sector on the final character of air masses reaching the measuring station, it is possible to classify and understand the dependence of aerosol properties on the dominant air mass type. The climatology of some aerosol properties measured in the atmospheric column and *in situ* in this town have been presented in previous studies (Estellés et al., 2007b; Esteve et al., 2012).

The knowledge of the physical and chemical properties of atmospheric aerosols is essential to understand their effect on the Earth's climate, both regionally and globally (Charlson and Heintzenberg, 1995). The Mediterranean is one of the most interesting regions of the planet in this sense, since its atmosphere is subject to different influences, both from local and remote sources. The specific region of this study, in the western Mediterranean extreme, is subject to the influence of particles from regions as diverse as the desert regions of North Africa, the continental shelf of Western Europe and the Atlantic Ocean. The characteristics of the aerosols from each of these regions are very different and will determine the properties of the atmospheric aerosols measured over the region. The combination of the atmospheric dynamics and the geographical location and the topography of the study area favors the development of mesoscale circulations (sea and mountain breezes), especially during the spring and summer months, meaning the aerosol transportation mainly takes place inland from the coast. The physical processes that determine air quality in this area have been previously documented (Millán et al., 1996, 1997, 2000).

Although the classification of air masses done by Estellés et al. (2007a) has only been applied to this specific Mediterranean region, the results can be considered valid for the entire Mediterranean coast of the Iberian Peninsula. Also, considering that the properties of aerosols at a certain time in a region are the result of contributions from local sources and distant sources, these results may also be valid for southwest Europe. Although the contribution of local sources largely depends on the specific location of the study (for example, proximity to urban sources), the contribution of distant sources will be more spatially homogeneous and will be determined by the air mass, being therefore representative of a broader region.

2 Methodologies and materials

2.1 Measurement season and period

The measurements of atmospheric aerosol properties were made with a TSI 3563 integrating nephelometer during the March 2006 to December 2009 period, at the measuring station installed on the terrace of the Faculty of Physics at the Burjassot Campus of the Universitat de València (39.507° latitude, longitude -0.420°, approximately 60 meters above sea level). Due to technical problems, measurements were interrupted between February 2007 and October 2008.

Burjassot is a city of 38,400 inhabitants located northwest of the metropolitan area of Valencia, with a total population of 1,832,000 inhabitants. Given its proximity to the main population center (5 km to the southeast), the station is under the direct influence of the urban and industrial pollution typical of a metropolitan area. Its proximity to the western coast of the Mediterranean Sea (10 km to the east) also determines the type of aerosols and their dynamics in this region.

2.2 Instruments and measurements

The aerosol scattering (σ_{sp}) and backscattering (σ_{bsp}) coefficients were measured at three wavelengths ($\lambda = 450$, 550, 700 nm) with a TSI 3563 integrating nephelometer (Anderson et al., 1996; Heintzenberg and Charlson, 1996). The measurements were performed at ambient relative humidity at 15 m above the ground. The averaging time of the measurements was 1 minute, and the zero signal of clean air was measured every hour for 5 minutes. The flow rate was set at $30 \, \mathrm{l \, min^{-1}}$. The nephelometer was calibrated at least twice a year (every 4 to 6 months) using CO_2 and air as high and low span gases, respectively. The uncertainty in the nephelometer measurements considering the angular truncation errors, the non-Lambertian nature of the light source, the wavelength non-idealities and the calibration errors is about 7% (Anderson et al., 1996; Anderson and Ogren, 1998).

The nephelometer measurements were adjusted to standard pressure (1013 hPa) and temperature (0°C) conditions. Due to the design of the instrument, the measurements of the scattering and backscattering coefficients do not cover the entire angular range (0 - 180°), so that a truncation correction for particles with "no size cut" was applied to them (Anderson and Ogren, 1998). Due to the lamp heating, the relative humidity inside the nephelometer during the study period ranged from 30 to 60% with a mean value of $45 \pm 10\%$, and we can therefore consider that the measurements presented in this paper correspond to dry particles (Targino et al., 2005).

The measurements of the scattering coefficients at wavelengths of 450 nm (σ_{sp}^{450nm}) and 700 nm (σ_{sp}^{700nm}) can be used to calculate the scattering Ångström exponent (α_s) with the expression:

$$\alpha_s = -\frac{\ln\left(\frac{\sigma_{sp}^{450nm}}{\sigma_{sp}^{700nm}}\right)}{\ln\left(\frac{450}{700}\right)} \tag{1}$$

The scattering and backscattering coefficients are indicators of the amount of aerosol in the atmosphere, while the scattering Ångström exponent reflects the nature of aerosols. Aerosol properties that depend on the amount of particles present are often called extensive properties, whereas those
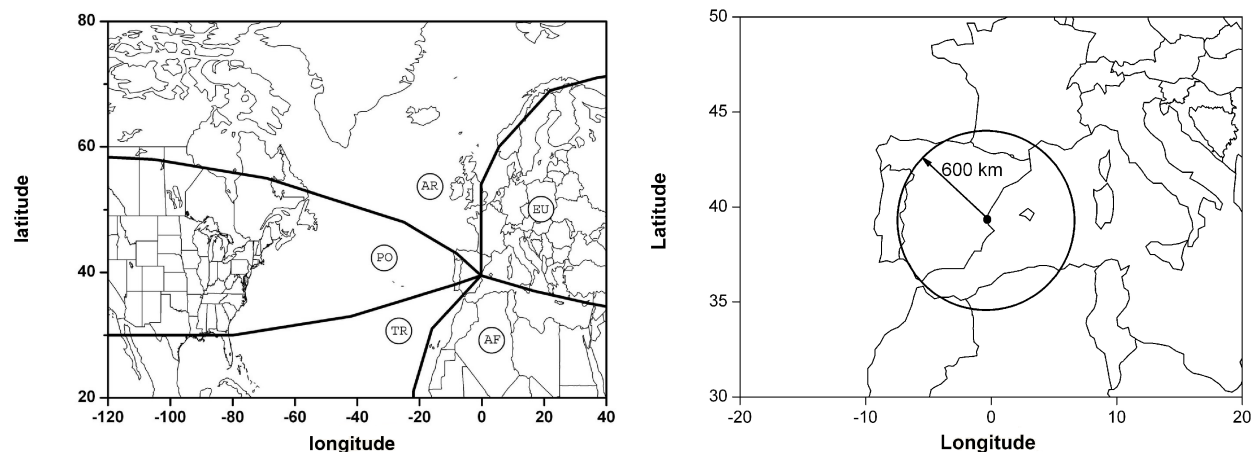
Figure 1. Main areas for backscattering trajectory classification: European (EU), African (AF), Tropical (TR), Polar (PO), Arctic (AR) and Regional (O).

related to the nature of the aerosol are called intensive properties (Ogren, 1995).

This study only shows the results obtained for the 550 nm wavelength, as the scattering and backscattering coefficients on the other wavelengths behave quite similarly. This 550 nm wavelength is especially important because the sun emits its maximum intensity at around 500 nm, so it is used for atmospheric correction of satellite images as well as for radiative transfer models.

2.3 Air masses classification

The HYSPLIT model (HYbrid Single-Particle Lagrangian Integrated Trajectory), developed by NOAA (National Oceanic and Atmospheric Administration) (Draxler and Rolph, 2003), offers the possibility to obtain the trajectory of the air parcel (and some key meteorological parameters) quantitatively in text format files. Estellés et al. (2007a) designed a simple classification model for describing the back trajectories using basic indices, thereby giving the air mass a primary or secondary feature. The applied vertical transportation model is that directly obtained from the vertical speed fields of the meteorological file. The flying time is limited to 5 days (120 hours), reaching the study area at 12:00 GMT. There are five areas selected by Estellés et al. (2007a) to classify the origin of the different back trajectories: European (EU), African (AF), Tropical (TR), Polar (PO) and Arctic (AR). Complementarily, they also define a regional class (O) (Figure 1). These sectors were defined considering both definitions based on temperature and surface type of the source region of air masses and other aspects related to aerosol physics.

In this simple classification model, some percentage indices ζ_i are defined, which describe the nature of an air mass in terms of the basic classes, and which are obtained by dividing the number of hours that the air mass needs to move through a particular sector i (t_i), and the total travelling time
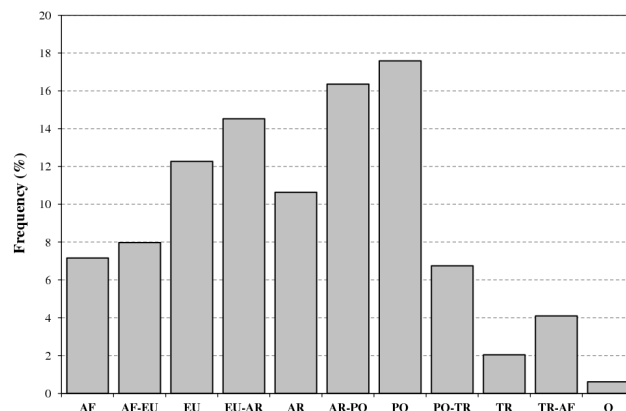
Figure 2. Incidence of each type of air mass in Valencia in the March 2006 to December 2009 period. AF: African, EU: European, AR: Arctic; PO: Polar, TR: Tropical, O: Regional.

(in our case, T = 120 h). Both values are weighted by two factors that take into account the average lifetime of aerosols and the height of the air parcel from the ground, which in our case have been set at 120 h and 2,000 m. To assign a definite character to each set of back trajectories, in practice we say that an air mass is pure i class if the index ζ_i has a value of more than 80%. Otherwise, the mass will be considered as mixed, and its composition will include all classes whose indices are larger than 20%. Henceforth, we will only work with pure masses, or mixed masses consisting of only two pure mass types.

To improve the temporal consistency between the back trajectories used in the classification of air masses and the measurements of aerosol properties, hourly average values have been used, which were calculated from data recorded every minute between 12:00 GMT and 13:00 GMT, in a manner that minimizes the effect of daily variability due to local sources.
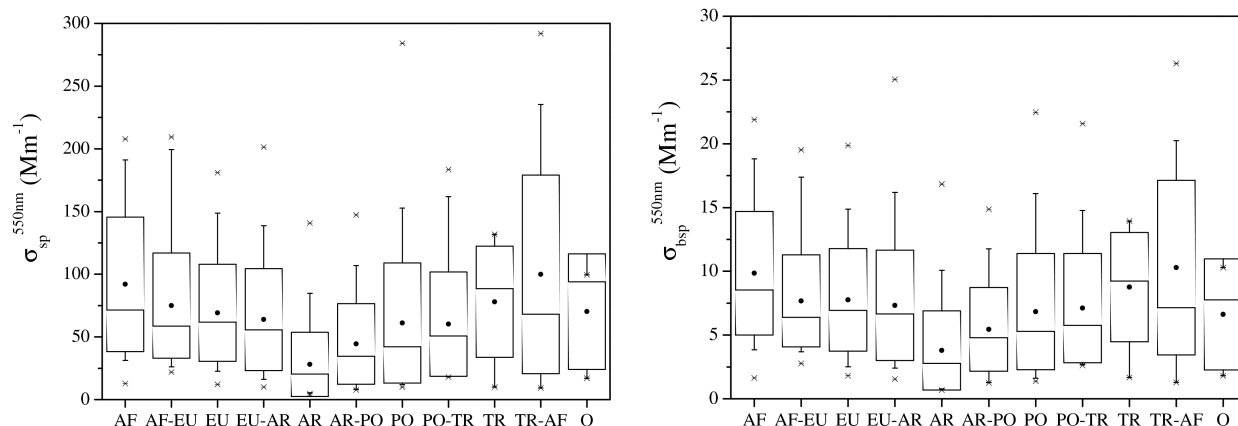
Figure 3. Dependence of the scattering (σ_{sp}) and backscattering (σ_{bsp}) coefficients at 550 nm on the air mass. AF: African, EU: European, AR: Arctic, PO: Polar, TR: Tropical, O: Regional.

3 Results

The incidence of each type of air mass, pure and mixed of two classes, in Valencia during the study period is shown in Figure 2. It can be observed that the most common air masses are PO (17.6%), AR-PO (16.4%) and EU-AR (14.5%) types, while TR-type and O-type masses are the least frequent (2% and 0.6% respectively). These results agree in part with those obtained by Estellés et al. (2007a) for the same region, with AR-PO and AF types being the most frequent air masses and TR-types being the least frequent. In our case, the AF-type air masses are not as frequent, as the period considered in this study contains more winter months than summer months, and AF-type air masses show a frequency maximum in the summer months and a minimum in the winter months (Estellés, 2006).

3.1 Scattering and backscattering coefficients

Scattering and backscattering coefficients are indicators of the attenuation of solar radiation due solely to aerosol dispersion on an angular range of 0 - 180° and 90 - 180°, respectively.

Figure 3 shows the classification of the scattering (σ_{sp}) and backscattering (σ_{bsp}) coefficients at 550 nm with the air mass in a box-plot. In these box-plots, the mean is indicated by a dot. The upper and lower limits of the box indicate the mean plus and minus the standard deviation. The vertical bars of the box represent the P_5 and P_{95} percentiles. The dividing line of the box indicates the median. The crosses refer to the P_1 and P_{99} percentiles, which in most cases are indistinguishable from the minimum and maximum, represented as a horizontal segment.

Although the scattering (σ_{sp}) and backscattering (σ_{bsp}) coefficients at 550 nm were measured *in situ* at ground level, these parameters are observed to be sensitive to the type of predominant air mass. Days that show higher σ_{sp} and σ_{bsp} values are under the influence of mixed and pure masses of
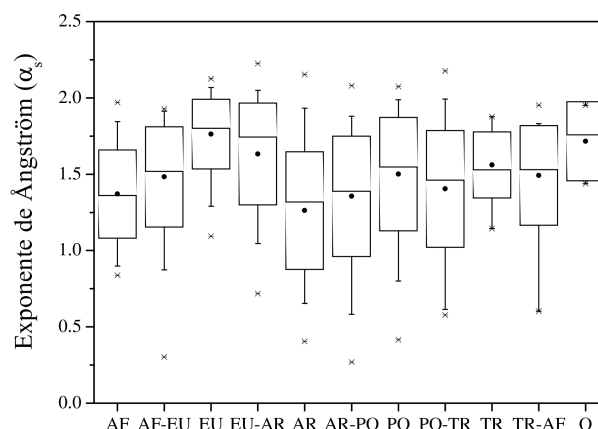
Figure 4. Dependence of the scattering Ångström exponent (α_s) on the air mass. AF: African, EU: European, AR: Arctic, PO: Polar, TR: Tropical, O: Regional.

AF and EU types. TR-type masses also present high values of σ_{sp} and σ_{bsp}, probably due to the presence of traces of mineral dust from a desert source added to the effect of relatively high temperatures and humidity that favor hygroscopic growth of sea salt and other aerosols resulting from gas-particle conversion. O-type masses show high σ_{sp} and σ_{bsp} values because they are stagnant in the study area due to low pressure gradient in anticyclone situations, facilitating the accumulation of various anthropogenic pollutants which, in the Mediterranean basin, are added to possible dust of peninsular or African origin. The minimum of σ_{sp} and σ_{bsp} are found in AR-type masses, which are the cleanest masses. These results agree with those obtained by Estellés et al. (2007a) for aerosol optical thickness in the same region, with maximum values for air masses of pure AF type and mixed with masses of EU and TR types, and minimum values for those of AR and PO types.

Table 1. Average and standard deviation of the scattering and backscattering coefficients at 550 nm and of the scattering Ångström exponent according to the dominant air mass (N: number of days; air mass type, AF: African, EU: European, AR: Arctic, PO: Polar, TR: Tropical, O: Regional).

	σ_{sp}^{550nm} (Mm^{-1})	σ_{bsp}^{550nm} (Mm^{-1})	α_s	N
AF	90 ± 50	10 ± 5	1.4 ± 0.3	35
AF - EU	80 ± 40	8 ± 4	1.5 ± 0.3	39
EU	70 ± 40	8 ± 4	1.8 ± 0.2	60
EU AR	60 ± 40	7 ± 4	1.6 ± 0.3	71
AR	30 ± 30	4 ± 3	1.3 ± 0.4	52
AR PO	40 ± 30	5 ± 3	1.4 ± 0.4	80
PO	60 ± 50	7 ± 5	1.5 ± 0.4	86
PO TR	60 ± 40	7 ± 4	1.4 ± 0.4	33
TR	80 ± 40	9 ± 4	1.6 ± 0.2	10
TR AF	100 ± 80	10 ± 7	1.5 ± 0.3	20
O	70 ± 50	7 ± 4	1.7 ± 0.3	3

3.2 Scattering Ångström exponent

The Ångström exponent is a parameter which is directly related to the aerosol size distribution and can be used to analyze in a simple but compact way the relative proportion of fine and coarse particles in the size distribution. When the distribution consists mainly of fine particles, this parameter has values in the range of 1.5 to 2.0, while it has values close to zero (even negative in some cases) when the distribution consists of particles of a larger radius (dust, hygroscopic or aggregated particles) (Seinfeld and Pandis, 1998).

Figure 4 shows the classification of the scattering Ångström exponent (α_s) with the air mass through a boxplot. Although one would expect the minimum values of α_s to be those of AF-type masses and their mixtures with TR-types, as is the case in the extinction Ångström exponent in the atmospheric column for the same region (Estellés et al., 2007a), these correspond, on the contrary, to those of PO and AR-type masses, which are cleaner air masses but can carry a mixture of sea salts immersed in an air with high levels of humidity and mineral dust, causing a low value of the scattering Ångström exponent. The main reason for this is that the transportation of mineral dust generally occurs in the middle layers of the troposphere, so AF-type masses are not always detectable at the surface (Derimian et al., 2006; Elias et al., 2006; Pereira et al., 2011). The maximum α_s values are found in EU-type masses, which carry large amounts of polluting particles of anthropogenic origin (soot produced in the combustion of fossil fuels) which are located in the accumulation way of size distribution (particles with sizes between 0.1 μm and 1 μm). O-type masses also have high values of α_s, probably due to the existence of particles from pollution and continental coarse dust or sea salt in warm, humid environments of the Mediterranean Sea. These results for the maximum values of α_s coincide with those obtained by Estellés et al. (2007a) for the extinction Ångström exponent in the atmospheric column for the same region.

The scattering Ångström exponent values on the surface are slightly larger than the values of the extinction Ångström exponent in the atmospheric column obtained for the same region (Estellés et al., 2007a). These differences between the results obtained for the land surface and the atmospheric column have been also observed by other authors (Lyamani et al., 2010), and are mainly due to the different techniques used in obtaining the Ångström exponent: the values observed for the atmospheric column are obtained by averaging the light extinction throughout the atmospheric column, while the values of the TSI 3563 integrating nephelometer are obtained from light scattering measurements performed on the surface.

Table 1 shows a summary of the classification of the aerosol properties according to the type of air masses, basic and mixed of two components.

4 Conclusions

The influence of the trajectories of air masses has been analyzed in the *in situ* measurements of aerosol properties in Valencia for the March 2006 to December 2009 period. The scattering (σ_{sp}) and backscattering (σ_{bsp}) coefficients were measured with a TSI 3563 integrating nephelometer. The scattering Ångström exponent (α_s) was also determined from σ_{sp} and σ_{bsp} measurements.

By classifying the source regions of air masses by sectors, and with the help of a simple model that quantifies the relative influence of each sector on the final character of air masses reaching the measurement station, it has been possible to establish the dependence of these aerosol properties with the type of dominant air mass. It has been checked, therefore, that the measurements of the aerosol properties performed at ground level are parameters that are sensitive to the type of air mass.

Pure and mixed AF-type masses have maximum values of scattering and backscattering coefficients, and the values of the scattering Ångström exponent are higher than expected, given the presence of mineral dust in these kinds of air masses, which usually provide many larger size aerosols in the upper atmosphere.

Mixed and pure EU-type masses present high values of scattering and backscattering coefficients and maximum values of the scattering Ångström exponent, since they transport large numbers of polluting particles of anthropogenic origin.

Mixed and pure TR-type masses also present high values of scattering and backscattering coefficients, although the values of the scattering Ångström exponent are low, probably due to the presence of traces of mineral dust of desert origin plus the effect of relatively high temperatures and humidity that favor hygroscopic growth of sea salt and other aerosols resulting from gas - particle conversion.

Mixed and pure AR and PO-type masses have the lowest values of scattering and backscattering coefficients, since they are cleaner air masses, as well as of the scattering Ångström exponent, probably due to the presence of a mixture of marine salts immersed in an air with high humidity and mineral dust levels.

Finally, O-type masses have high values of scattering and backscattering coefficients, as well as scattering Ångström exponent, because they get trapped in the study area facilitating the accumulation of various anthropogenic pollutants, which in the Mediterranean basin are added to dust of possible peninsular or African origin.

Given that aerosol properties at a certain time over a region are the result of contributions from local sources and distant sources, the results obtained in this study can be considered valid for the entire Mediterranean coast of the Iberian Peninsula as well as for the entire southwest of Europe since the contribution of distant sources, which depends on the type of air mass, will be similar, although the contribution of local sources depends largely on the study region.

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