Spatial and Temporal Variability of Aerosol Properties at Different Altitudes in Sierra Nevada Using In-Situ Techniques

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Abstract. This work aims to describe the spatial and temporal variability of aerosol properties over the Southeast Iberian Peninsula using in-situ techniques and ancillary tools. The study was perfomed at Granada (37.16ºN, 3.61ºW) at three different altitudes: IISTA-CEAMA (680 m above sea level, a.s.l.), Hoya de Pedraza (1950 m a.s.l.) and Albergue Universitario (2500 m a.s.l.). The last two station were emplaced at Sierra Nevada Natural Park. The study period comprises from July to September 2015, in a summer campaign. In-situ aerosol optical properties were analyzed during this period, showing behaviors characteristic of mineral dust presence. Different satellite models were used to confirm the character of the Northern African origin air masses affecting the study region. The interest of this study is characterizing the events occurred in Granada and Sierra Nevada, observe the differences between altitudes, gaining an insight into the different impacts of the dust events at the measurements sites.

Keywords: aerosol, altitude, absorption Ångström exponent, absorption coefficient, black carbon, desert dust, mountain, Saharan dust intrusion.

INTRODUCTION

Atmospheric aerosol particles can modify the Earth's radiative balance: scattering aerosols with negative radiative forcing (e.g., mineral dust, sulfate, nitrate, and organic carbon) and absorbing aerosols with positive radiative forcing (e.g., black carbon) affect the radiation budget in the opposite way (Kopp and Mauzerall, 2010; IPCC, 2013). Additionally, atmospheric aerosols have a great impact on human health (Lightly et al., 2011) as well as in ambient air quality (e.g. Akimoto, 2004). Aerosol particles originate from different sources, are exposed to different kinds of processes in the atmosphere, and are removed from it by a variety of sinks. The source origin of aerosols plays an important role in their characterization. Anthropogenic aerosols like black carbon (BC) are primary products of incomplete combustions of carbonaceous fuels exhausted by diesel engines (e.g. Reche et al., 2011). Biomass burning and domestic heating based on fuel-oil are also BC aerosol sources. Talking about natural

aerosols, desert dust is one the major aerosol components in the global atmosphere that affects the Earth's climate through interacting with both solar and thermal infrared radiation. The North African dust is the most important source of mineral dust in the Northern Hemisphere (e.g., Liu et al., 2008). Because mineral desert dust can be transported over thousands of kilometers, it is not only a regional phenomenon, but has a global climate effect. In fact, the Iberian Peninsula is frequently affected by North African dust episodes with large aerosol load that can modulate the aerosol climatology in different areas, especially in southern Spain (Lyamani et al., 2005).

Most in-situ surface aerosol measurement sites are within the boundary layer at low elevation sites, however there is a small subset of high elevation sites. For instance, atmospheric aerosol optical properties have been widely investigated in the city of Granada at surface level during the past years (e.g. Titos et al., 2012) but there is lack of information about aerosol properties in Sierra Nevada national park. This emplacement is a unique natural laboratory to investigate aerosol properties due to its isolated location in the far south of Europe and its proximity to the African continent and to the western Mediterranean. Its high altitude allows characterizing lofted aerosol layers in the free troposphere associated to long-range transport processes (i.e. dust transport from the Sahara desert or pollution coming from Europe) as well as detect transport processes associated to the growth of the planetary boundary layer (i.e. pollution transport from Granada urban area to Sierra Nevada). To perform such characterization, three measurement stations were deployed in southern Spain: one in Granada urban area (IISTA-CEAMA, 680 m a.s.l.) and two sites at different heights in Sierra Nevada slope (Hoya de Pedraza, 1950 m a.s.l. and Albergue Universitario, 2500 m a.s.l.). The main purposes of the present study are to investigate the impact of dust episodes in aerosol properties at three different altitudes and to investigate the differences between data obtained during dust outbreaks in comparison with data in normal summer conditions.

EXPERIMENTAL SITE AND INSTRUMENTATION

The measurements used in this work were performed over the Southeaster Spain at Granada (37.16ºN, 3.61ºW) at three different heights: the Andalusian Institute for Earth System Research (IISTA-CEAMA) at 680 m a.s.l., Hoya de Pedraza at 1950 m a.s.l. and Albergue Universitario at 2500 m a.s.l. The last two measurement stations are located over Sierra Nevada mountain range. The main external aerosol source regions affecting the study area are: Europe as a major source of anthropogenic pollution and North Africa as a principal source of natural dust.

Among the instruments used in this study, Aethalometer measures the rate of change of optical transmission through a spot on a filter on which aerosol is being collected. From the increase in optical attenuation due to the increment of collected aerosol, absorption coefficient can be calculated at 7 different wavelengths as well as BC mass concentrations can be estimated. Two Aethalometer models were used: AE33, located at IISTA-CEAMA and two AE31, locating one at each mountain station. The MAAP (Multi-Angle Absorption Photometer) measures the aerosol light absorption coefficient, $\sigma_{ap}(\lambda)$, at 637 nm at IISTA-CEAMA station. Number size

distributions were provided by APS (Aerodynamic Particle Sizer, model 3321, TSI) at IISTA-CEAMA and two GRIMMs (model EDM 164) at Hoya de Pedraza and Albergue Universitario respectively. As ancillary instrument, CHM15k-Nimbus (Jenoptik S.A., Germany) ceilometer measurements has been used, located at IISTA-CEAMA. It includes a pulsed Nd:YAG laser, emitting in 1064 nm of wavelength. Quicklooks obtained by this instrument were used to identify aerosol layers over the study area.

METHODOLOGY

The data processed in this study was the part relative to in-situ measurements, i.e. data from Aethalometers, MAAP, APS and GRIMMs. For these instruments, the removal of the rest of 99th percentile as a signal of bad quality data was applied.

The aerosol attenuation coefficient, $b_{atn}(\lambda)$, was used to estimate the BC mass concentration using mass absorption efficiencies recommended by the manufacturer. In this work, the change in optical attenuation at 660 nm wavelength has been used. At this wavelength the absorption by other absorbing compounds (e.g. organic particles and dust) is very small relative to that by BC and thus the aerosol absorption at 660 nm can be attributed to BC alone. In this study, Aethalometers measurements were compensated by means of C constant by comparing with MAAP data. This constant makes a correction between absorption coefficient and attenuation coefficient at the same wavelength.

The Absorption Ångström Exponent (AAE) describes the wavelength dependency of the absorption coefficient and it can give information about the type of aerosol or source. However, we observed differences in the AAE retrieved from the different instruments during co-located measurements. Thus, to assure comparability between AAE calculated from different Aethalometers we have referenced the spectral dependence of the AE31 instruments to that of the AE33, which is considered more accurate.

GRIMMs spectrometers determine the aerosol number size distribution, $dN/dlog\ D_p$, based on light-scattering measurements. As these instruments were factory calibrated with an refractive index (RI) different from ambient aerosol, GRIMM diameters have been recalculated following Pio et al. (2014) methodology that is based on Mie theory. Different RIs were used for dust particles and "clean continental" conditions. In order to compare APS and GRIMM measurements, geometric diameters (D_{pgeo}) are converted to aerodynamic diameter (D_{paer}) with the Stokes Law approximation. Particle number, N, was directly obtained from the measured number size distribution in both instruments and the aerosol mass size distribution, $dM/dlog\ D_p$, can be obtained from the measured number size distribution, the diameters and a different density depending on the predominant aerosol type. PM_x (particles with diameters below x µm) mass concentrations were calculated from $dM/dlog\ D_p$ up to x µm.

Ancillary tools as CALIMA (Caracterización de AerosoLes originados por Intrusiones de Masas de aire Africanas), HYSPLIT4 model (HYbrid Single Particle Lagrangian Integrated Trajectory; Draxler et al., 2009) version 4.9, NAAPs model

(Navy Aerosol Analysis and Prediction System) and MODIS (Moderate Resolution Imaging Spectroradiometer), were used to characterize the air masses arriving at Granada in the measurement period.

RESULTS

An overview of the in-situ variables measured during the study period at Granada (IISTA-CEAMA) and Sierra Nevada (Hoya de Pedraza and Albergue Universitario), showed an important increase during two periods (01/08/15-15/08/15 and 26/08/15-02/09/15). The results suggested that the air masses affecting the stations were enriched in mineral dust particles. These dust episodes were confirmed by the models.

Backscatter signal from ceilometers confirmed the intrusions of Saharan dust plumes. In-situ variables allowed for accurately determine the times of maximum dust concentrations. High AAE values pointed to an increase in UV-absorbing aerosols during dust events at the mountain stations while this enhancement was reduced at the surface station due to the predominance of BC particles. Additionally, volume distribution were studied at both events showing maximums at different diameters, presenting particles with bigger size at Albergue Universitario than at Hoya de Pedraza.

In order to study the differences between dusty and non-dusty conditions at the three measurement stations, a statistical analysis of the main variables was performed. The periods with Saharan dust events were called as *dust conditions* and non-dusty conditions as *summer conditions* (all data excluding dust events). During both conditions, BC mass concentration varied with altitude, being Albergue Universitario the station that presents lower BC mass concentration followed by Hoya de Pedraza and finally IISTA-CEAMA. Also, the spatial variation related to the proximity of the stations to the source was crucial in the analysis because IISTA-CEAMA is the station more affected by pollution due to its urban background character and thus, it is reflected in the higher BC mass concentrations reached.

Finally, diurnal evolutions of in-situ variables were studied excluding from the analysis desert dust events, in order to investigate the “normal conditions” in summer. The averaged diurnal evolution of AAE at IISTA-CEAMA was characterized by two minima in coincidence with traffic rush hours indicating fresh fuel oils emissions. AAE daily averaged pattern at Hoya de Pedraza shows one maximum possibly related with the thermally driven pollutants upslope from the city and therefore aged BC aerosols. Averaged diurnal BC (σ_{ap}) at IISTA-CEAMA showed two maxima in coincidence with traffic rush hours as well. At the mountain sites one maximum was reached at Albergue Universitario similar to the central peak at Hoya de Pedraza, possibly related with the entrance of the stations within the Planetary Boundary Layer (PBL). Two additional peaks are present at Hoya de Pedraza that could be due to traffic activity since there is a larger influence of NIR-absorbing particles that can be seen for the lower AAE values and higher BC and $\sigma_{ap}(660)$ values at that time.

CONCLUSIONS

The spatial and temporal variability of aerosol properties have been investigated at different altitudes in Sierra Nevada using in-situ techniques. This study represents an in-depth analysis of major Saharan dust events during the study period (a summer campaign that comprises from July to September 2015). The relevance of this analysis was the characterization of these events at Sierra Nevada and Granada by means of in-situ parameters and ancillary tools.

Future analysis at Sierra Nevada and Granada should be done during long-periods in order to characterize the spatial and temporal variability of aerosols optical properties at different seasons. Additionally, a better understanding of the planetary boundary layer dynamics, vertical wind field and transported processes will be crucial for quantify the impact of different air masses at different altitudes in Granada.

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