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Atmospheric boundary layer and ozone-aerosol interactions under Saharan intrusions observed during AMISOC summer campaign



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HIGHLIGHTS

- Selection of dusty and clean conditions in an experimental campaign.
- Arrival of Saharan dust in the upper levels (from 750 to 6000 m) in SW Spain.
- ABL structure and evolution: comparison during dusty and clean conditions.
- · Aerosol profiles using Lidar for non-dusty and dusty cases.
- O₃-aerosol interactions for a dusty scenario: particles increase and no O₃ destruction.

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ABSTRACT

A research campaign was performed for the AMISOC (Atmospheric Minor Species relevant to the Ozone Chemistry) project at El Arenosillo observatory (southwest Spain) in May-June 2012. The campaign focused on the impact of Saharan dust intrusions at the atmospheric boundary layer (ABL) and ozoneaerosol interactions. In-situ and remote-sensing techniques for gases and aerosols were used in addition to modelling analyses. Meteorology features, ABL structures and evolution, aerosol profiling distributions and aerosol-ozone interactions on the surface were analysed. Two four-day periods were selected according to non-dusty (clean conditions) and dusty (Saharan dust) situations. In both scenarios, sea-land breezes developed in the lower atmosphere, but differences were found in the upper levels. Results show that surface temperatures were greater than 3 °C and humidity were lower during dusty than non-dusty conditions. Thermal structures on the surface layer (estimated using an instrument on a 100 m tower) show differences, mainly during nocturnal periods with less intense inversions under dusty conditions. The mixing layer during dusty days was 400-800 m thick, less than observed on nondusty. Dust also disturbed the typical daily ABL evolution. Stable conditions were observed in the early evening during intrusions. Aerosol extinction on dusty days was 2-3 times higher, and the dust was confined between 1500 and 5500 m. Back trajectory analyses confirmed that the dust had an African origin. On surface, the particle concentration was approximately 3.5 times higher during dusty events, but the local ozone did not exhibit any change. The arrival of Saharan dust in the upper levels impacted the meteorological surface, inhibited the daily evolution of the ABL and caused an increase in aerosol loading on the surface and at higher altitudes; however, no dust influence was observed on the surface

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1. Introduction

Climate change and air pollution are two strongly related major

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issues in atmospheric research. Photochemical pollutants cause major damage to human health and the environment. There are several issues that are weakly studied or poorly understood, e.g., the interaction between species in a gas phase and aerosols, and the influence of dust in the atmospheric boundary layer (ABL). Both issues should be addressed in detail, paying special attention to

surface meteorological changes in dust-influenced zones.

Desert dust contributes to 40% of the mass of aerosol injected to the troposphere, half of which is from the Sahara desert (Miller et al., 2004). Additionally, African dust can be transported to regions far from its sources, even to the Caribbean or America (Prospero, 1999). Dust outbreaks are mainly contained to the Subtropical Eastern North Atlantic Region (Alonso-Pérez et al., 2012), Europe (Pey et al., 2013) and Africa (Engelstaedter et al., 2006; Drame et al., 2011).

Saharan particles have a large surface area, which strongly absorbs shortwave solar radiation, resulting in photolysis rate reduction. Additionally, dust particles may play a significant role as a reactive surface on which heterogeneous chemistry can take place. Hanisch and Crowley (2003) showed that ozone loss can be caused by decomposition, catalytic destruction or absorption on mineral oxides. Several publications have discussed the relationship between high dust concentrations and ozone reduction (De Reus et al., 2005; Bonasoni et al., 2004; Jenkins et al. 2012a,b; Andrey et al., 2014).

The concentrations of chemical species in the lower atmosphere, however, are governed by surface exchanges, chemical processes and particular dynamics occurring in the ABL (Garrat et al., 1994). Synoptic conditions affect the surface meteorology and ABL development, influencing the ABL chemical species. Few works have investigated the Saharan dust influence on the ABL (Carlson and Prospero, 1972; Marsham et al., 2008; Messager et al., 2010) and its implications in gas—aerosol interactions.

The study area, located in SW Spain, is affected by maritime and continental air masses. Mesoscale processes can be developed and were investigated by Adame et al. (2010). Two sea-land breeze patterns, pure and non-pure, were identified at that time. Pure breezes show the typical behaviour of coastal areas, where flows run perpendicular to the coastline. Non-pure breezes occur under synoptic forces, causing a nocturnal regime with air flows not perpendicular to the coastline. Chemical species are affected by the breeze patterns. Under pure breeze, medium—high concentrations of ozone are measured, while in non-pure breeze conditions, the ozone concentrations are lower.

Using previous experience, an experimental campaign at El Arenosillo was designed to simultaneously measure aerosols and chemistry active gases for the AMISOC (Atmospheric Minor Species relevant to the Ozone Chemistry at both sides of the Subtropical jet) project.

The aim of this paper is to examine the effects of Saharan dust on surface meteorology and ABL structure, and their influence on ozone and aerosols on the surface. Experimental data on surface and at upper height-levels were compared using two aerosol scenarios corresponding to dusty and non-dusty conditions.

Section 2 shows the AMISOC-ARN campaign framework, including the measurement area description, instrumentation and modelling tools. Section 3 presents the main results. Section 3.1 shows the selection of non-dusty and dusty events, Section 3.2 shows the associated weather conditions, Section 3.3 analyses the ABL structure and daily evolution, Section 3.4 presents the vertical aerosol features, and Section 3.5 discusses the ozone and aerosols interactions. Finally, Section 4 summarises this work.

2. AMISOC-ARN campaign framework, experimental, models and ancillary information

2.1. Campaign and area description

The AMISOC-ARN campaign was performed at "El Arenosillo" (37.1°N, 6.7°W, 40 m a.s.l.) at the Institute for Aerospace Technology (INTA). Observations were performed from 15 May to 15 June 2012

using in-situ and remote sensing instrumentation to study particles, ozone and meteorology features.

El Arenosillo observatory is located in the SW Iberian Peninsula (hereafter IP) in the Doñana National Park, 1 km from the Atlantic Ocean coastline (Fig. 1). Surface meteorology, solar radiation, surface chemistry species (ozone, NO and NO₂), aerosol size distribution, are routinely monitored. Additional instrumentation was deployed: lidar and sun-photometry for, respectively, heightresolved and columnar measurements, in addition to daily atmospheric soundings. In addition, a 100-m-high tower was used with two meteorological stations at 50 and 100 m.

2.2. Experiment

2.2.1. Surface in-situ instrumentation

Micrometer-sized particles (0.54–10 μm) were monitored with an Aerodynamic Particle Size (APS) Spectrometer (TSI Mod. 3321). This instrument is a time-of-flight spectrometer measuring the velocity of particles in an accelerating air flow though a nozzle. Suspended mineral dust range from 0.1 to 20 μm in diameter. In previous works on desert dust detected at ground level performed at this observatory (Córdoba-Jabonero et al., 2011), the medium-distance dust transport from the Saharan desert showed diameters ranging from 0.07 to 10 μm (Sorribas et al., 2011). The total particle concentration with sizes ranging from 0.05 to 10 μm was selected to identify dust on the surface. A time resolution of 30 min was used.

Surface ozone were collected with an ozone analyser (Dasibi 1008 RS) based on the absorption of ultraviolet radiation at 254 nm using a flow rate of $2 \, l \, min^{-1}$ and a detection limit of 1 ppb. This instrument is calibrated weekly and monthly. An ozone average is collected every 10 min. The inlet is 8 m above the ground and 2 m above the canopy.

2.2.2. Remote sensing instrumentation

Columnar-integrated measurements were collected with a CIMEL. This sun-photometer measures direct sun and sky radiation at four wavelengths: 440, 670, 780 and 1020 nm. The calibration of the solar direct irradiance was performed by transference with an AERONET instrument (site: Granada), which was recently calibrated by the RIMA-AERONET network. The irradiance calibration uncertainty is 1–2%. The spectral aerosol optical depth (AOD) and principal plane inversion methods described by Olmo et al. (2008) were used to retrieve the AOD and Ångström wavelength exponent (AEx), which are the main CIMEL parameters used for dust intrusion evidence. Spectrally resolved AOD and AEx were derived at visible wavelengths close to the lidar laser wavelength.

The Micro Pulse Lidar v.3 (MPL-3) system used is a single-wavelength (523 nm), high-repetition (2500 Hz), low-power (~7 µJ), eye-safe commercially available backscatter lidar, for observations of the vertical distribution of aerosols and clouds while fully un-attended. MPL is currently in operation within MPLNET (Micro Pulse Lidar NETwork, mplnet.gsfc.nasa.gov) at Sta. Cruz de Tenerife. This MPL-3 was deployed at El Arenosillo for the campaign. More instrumental details are found in Campbell et al. (2002).

Vertical aerosol extinction coefficient profiles were obtained using a Klett-Fernald-Sasano iterative algorithm (Klett, 1985; Fernald, 1984; Sasano and Nakane, 1984) in addition to the AOD values reported by Cimel data registered during the campaign. Parameters, such as the Lidar Ratio (LR), and the top of the aerosol layer were calculated for each profile. No inversion was performed under cloudy conditions. MPL-3 measurements were collected on a daily basis, and raw profiles were acquired with a 1 min resolution-time and a vertical resolution of 15 m. These profiles were averaged

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