



Experimental determination of short- and long-wave dust radiative effects in the Central Mediterranean and comparison with model results



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ABSTRACT

Downward and upward irradiance measurements, in the short-wave (SW) and long-wave (LW) spectral range, have been used in combination with simultaneous aerosol optical depths (AODs) to experimentally determine the instantaneous and clear-sky aerosol Direct Radiative Forcing (DRF) at the surface, during a desert dust outbreak which affected the Central Mediterranean from 9 to 13 July 2012. AODs were retrieved from AERONET (AErosol RObotic NETwork) sun/sky photometer measurements collocated in space and time. The importance of downward and upward radiative flux measurements to properly account for both the surface albedo dependence on the solar zenith angle, and the land surface temperature (T_{LS}) has been highlighted. Measured radiative fluxes were in reasonable agreement with the CERES (Clouds and the Earth's Radiant Energy System) and AERONET corresponding ones collocated in space and time. SW and LW downward fluxes at the surface decreased up to 9% and increased up to 13%, respectively, as a consequence of a factor 5 increase of the AOD at 675 nm (AOD_{675}). This is due to the cooling and warming effect of desert dust in the SW and LW spectral range, respectively. In fact, we have also found that the T_{LS} increased at a rate of about 250 K per unit increase of the AOD_{675} . The aerosol DRF at the surface varied from -8 to -74 $W m^{-2}$ and from $+1.2$ to $+9.6$ $W m^{-2}$ in the SW and LW spectral domains, respectively. In particular, we have found that the LW-DRF on average offsets 14% of the related SW component. It is shown that a two-stream radiative transfer model can reproduce the experimental findings at the surface by replacing the refractive indices typical of dust particles with the ones obtained for a mixture made of dust and soot particles. The dust contamination by anthropogenic particles during its transport to the monitoring site located several hundred kilometers away from the source region was responsible for this last result. We have also found by model simulations that the LW-DRF increased linearly with T_{LS} both at the surface and at the top of the atmosphere.

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1. Introduction

Desert dust aerosols play an important role in the Earth's radiation budget and significantly influence the climate system through a direct effect (scattering and absorption of solar and terrestrial radiation) (Miller and Tegen, 1998) and an indirect effect (acting as cloud and ice condensation nuclei) (Santos et al. 2013). The IPCC's Fifth Assessment Report on Climate Change (2013) recently stated that the contribution of aerosol particles represents the largest uncertainty to the total radiative effect estimation. This can be attributed to the great variety of sources and transformation processes leading to a considerable uncertainty about concentration, size distribution, and chemical composition of aerosol particles, which mainly affect their light-absorbing and -scattering properties (Kondratyev et al. 2006). The Mediterranean basin represents one of the most important areas for the study of mineral dust due to its proximity to the Sahara desert,

which represents the main source of dust worldwide with an estimated global emission of 670 Mt. year⁻¹ (Rajot et al. 2008). Dust particles captured by wind action at the surface of arid regions are blown into the atmosphere up to high altitudes by strong convective regimes and then are transported by prevailing winds over long distances up to intercontinental scale (Blanco et al. 2003; Chin et al. 2007). Therefore, desert dust particles may change their optical and physical properties during transport, since they can interact and/or mix with other aerosol particles (Gómez-Amo et al. 2011). Perrone and Bergamo (2011) implemented a methodology to estimate the contribution of anthropogenic particles during dusty days monitored over south-eastern Italy. The aerosol direct radiative effect can be quantified by the Direct Radiative Forcing (DRF) and the Aerosol Forcing Efficiency (AFE). The DRF results strictly related to the radiative properties and concentration of the aerosol particles, while the AFE is mainly dependent on the particle composition and size (Tafuro et al. 2007). The SW- and LW-DRFs by dust are generally of opposite sign. In fact, desert aerosols cause the decrease of the incoming SW radiation at the surface due to their scattering and absorption properties inducing a cooling effect and a negative value of SW-DRF. Conversely, the absorption and scattering of dust particles in

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the LW-domain enhance the greenhouse effect by trapping the outgoing terrestrial radiation and therefore the LW-DRF is positive at the surface (Hansell et al. 2010). The magnitude and the sign of the dust DRF are also dependent on other parameters such as the particle refractive index, the altitude of the dust layer, the single-scattering albedo, the asymmetry parameter, and the surface albedo (Osborne et al. 2011; Perrone and Bergamo, 2011). The dust DRF affects the atmospheric dynamics by changing the thermal vertical profile and the atmospheric stability (Kishcha et al. 2003).

Several works on the SW (e.g. Valenzuela et al. 2012) and LW (e.g. Sicard et al. 2014) desert dust radiative effects over the Mediterranean are based on radiative transfer models. Some papers reported desert dust DRFs calculated from experimental findings and model simulations to determine the radiative flux with and without aerosol, respectively (e.g. Antón et al. 2012; García et al. 2014). Conversely, few studies have estimated the desert dust radiative effect both in the SW (e.g. Obregón et al. 2015) and in the LW (e.g. Antón et al. 2014) spectral range by only using experimental observations, as it is done in this paper. Furthermore, most of the previous works were only based on experimental measurements of the downward radiation. In fact, the upward radiation required for the DRF calculation was usually evaluated by using surface albedo values commonly retrieved from satellite measurements.

Simultaneous downward and upward, SW and LW irradiance measurements have been used in this study to calculate the surface albedo (SA) and the land surface temperature (T_{LS}), respectively, which are of peculiar importance to properly determine the SW- and LW-DRF, respectively. The experimental determination of the aerosol DRF at the surface in the SW and LW spectral domain during a desert dust outbreak represents one of the main aims of this study. Experimental aerosol DRFs have been compared with the corresponding ones obtained from model simulations, based on experimental SA and T_{LS} values. Then, the impact of the refractive index values on the simulated DRF values has been investigated to improve the agreement between model and experimental data. Irradiance measurements performed at a Central Mediterranean site during 8–14 July 2012 have been utilized in this study. The Saharan dust outbreaks affect the Mediterranean region mostly in the summer confirming that the maximum desert dust load occurs in warm seasons (Choobari et al. 2014) and contributing to one of the highest radiative effects in the world (Lelieveld et al. 2002). The experimental site and instrumentation are described in Section 2. The characterization of the studied Saharan dust outbreak is provided in Section 3. The results on surface flux measurements are discussed in Section 4. The methodology and the main results about the aerosol direct radiative forcing are presented and discussed in Section 5. The comparison with model results is provided in Section 6. Final remarks are highlighted in Section 7.

2. Experimental site and instrumentation

2.1. Experimental site description

The measurements analyzed in this study have been performed at the Mathematics and Physics Department of the Salento University (40.33°N; 18.11°E) during 8–14 July 2012. The monitoring station is located 6 km away from the town of Lecce (~95,000 inhabitants), on a flat peninsular area of south-eastern Italy, and is representative of coastal sites of the Central Mediterranean away from large sources of local pollution (e.g. Basart et al. 2009; Perrone et al. 2014a). The studied area is affected by a great variety of aerosol types: mineral dust from the Sahara desert (~1500 km away) and surrounding arid regions, polluted particles from urban and industrial areas of Northern and Eastern Europe, sea salt and spray from the Mediterranean Sea itself or from the Atlantic Ocean, and biomass burning particles produced by forest fires mostly in summer (Perrone et al. 2012). Thus, the monitoring station of the present study, which is located at the center of the Mediterranean

basin, represents a good site to characterize the dust properties and corresponding radiative effects several hundred kilometers away from the source regions (e.g. Santese et al. 2008; Perrone et al. 2013).

2.2. Instrumentation description

Two Kipp & Zonen pyranometers (CMP 21 model) and two pyrgeometers (CGR 3 model) have been used to measure upward and downward radiative flux at the surface in the solar (0.31–2.8 μm) and the terrestrial (4.5–42 μm) spectral range, respectively. In particular, the Kipp & Zonen CMP 21 model is classified as a “secondary standard instrument”, which represents the best pyranometer class according to the ISO 9060 standard adopted by the WMO (World Meteorological Organization). It presents a non-linearity <0.2% in the range 100–1000 Wm^{-2} , a directional response <10 Wm^{-2} (up to 80° with 1000 Wm^{-2} beam), and a temperature response <1% (from –20 °C to +50 °C). The CGR 3 pyrgeometer has a non-linearity <1% and a temperature response <5% (from –40 °C to +40 °C). A recent calibration of both pyranometers and pyrgeometers assures the good quality of the analyzed irradiance measurements. Collected data have been averaged over 2 min and stored for subsequent analysis. The total uncertainty associated with the CMP 21 pyranometer and the CGR 3 pyrgeometer measurements is 2% and 3%, respectively, taking into account temperature, calibration, and cosine error of the devices. The pyranometers and the pyrgeometers are located at 1.5 m above ground level so as to have the horizon free of significant obstacles.

A CIMEL sun/sky photometer operating within NASA AERONET (Aerosol RObotic NETwork; Holben et al. 1998) since 2003 has been used to characterize the columnar aerosol parameters. It is located a few hundred meters away from the pyranometer's and pyrgeometer's site. The sun/sky photometer uses two automatic detectors to measure direct solar flux in 8 channels between 340 and 1020 nm and the diffuse-sky flux in 4 channels between 440 and 1020 nm. The AERONET database (<http://aeronet.gsfc.nasa.gov>) provides several products including the Aerosol Optical Depth (AOD) and the Ångström exponent (Å) from direct sun measurements. Asymmetry parameter, single-scattering albedo, real and imaginary refractive indices, and aerosol volume size distribution (for particle radii within the 0.05–15 μm range) are derived from an inversion algorithm Version 2 (V2) (Dubovik et al. 2006) based on both direct sun and diffuse-sky measurements. Model retrieved SW radiative fluxes and DRFs (e.g. García et al. 2008) are also provided. The broadband (0.2–4.0 μm) SW fluxes are calculated by a module of the radiative transfer model GAME (Global Atmospheric ModEl; e.g. Dubuisson et al. 2006), which was integrated into the operational AERONET inversion code. This module takes into account the molecular scattering and gaseous absorption effects by using the correlated k -distribution (Lacis and Oinas, 1991), which allows short computational time for providing broadband fluxes within the AERONET operational procedure. In particular, the spectral integration was done in the range from 0.2 to 4.0 μm using more than 200 spectral sub-intervals. For each of these sub-intervals, the extinction, single scattering albedo, and phase function were recalculated using the AERONET aerosol model driven by retrieved size distribution, complex refractive index, and fraction of spherical particles. The AERONET DRF at the surface is defined as the difference between downward flux with aerosol and the one without aerosol. García et al. (2012) indicated that the AERONET DRF at the surface overestimates the real values since the upward fluxes are not taken into account. Therefore, the AERONET DRF at the surface is usually corrected by the term (1-SA), according to García et al. (2012). SA is obtained from the spectral average of satellite-based surface albedo values at 440, 675, 870, and 1020 nm. Only cloud-screened and quality-assured AERONET retrievals (level 2.0) have been considered in this study.

Measured SW and LW fluxes at the surface have been compared with the ones provided by CERES (Clouds and the Earth's Radiant Energy System) to investigate the correlation between the two datasets

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