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Overview of the Chemistry-Aerosol Mediterranean Experiment/Aerosol Direct Radiative Forcing on the Mediterranean Climate (ChArMEx/ADRIMED) summer 2013 campaign

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Abstract

The Chemistry-Aerosol Mediterranean Experiment (ChArMEx; <http://charmex.lsce.ipsl.fr>) is a collaborative research program federating international activities to investigate Mediterranean regional chemistry-climate interactions. A special observing period (SOP-1a) including intensive airborne measurements was performed in the framework of the Aerosol Direct Radiative Forcing on the Mediterranean Climate (ADRIMED) project during the Mediterranean dry season over the western and central Mediterranean basins, with a focus on aerosol-radiation measurements and their modeling. The SOP-1a took place from 11 June to 5 July 2013. Airborne measurements were made by both the ATR-42 and F-20 French research aircraft operated from Sardinia (Italy) and instrumented for in situ and remote-sensing measurements, respectively, and by sounding and drifting balloons, launched in Minorca. The experimental set-up also involved several ground-based measurement sites on islands including two ground-based reference stations in Corsica and Lampedusa and secondary monitoring sites in Minorca and Sicily. Additional measurements including lidar profiling were also performed on alert during aircraft operations at EARLINET/ACTRIS stations at Granada and Barcelona in Spain, and in southern Italy. Remote sensing aerosol products from satellites (MSG/SEVIRI, MODIS) and from the AERONET/PHOTONS network were also used. Dedicated meso-scale and regional modelling experiments were performed in relation to this observational effort. We provide here an overview of the different surface and aircraft observations deployed during the ChArMEx/ADRIMED period and of associated modeling studies together with an analysis of the synoptic conditions that determined the aerosol emission and transport. Meteorological conditions observed during this campaign (moderate temperatures and southern flows) were not favorable to produce high level of atmospheric pollutants nor intense biomass burning events in the region. However, numerous mineral dust plumes were observed during the campaign with main sources located in Morocco, Algeria and Tunisia, leading to aerosol optical depth (AOD) values ranging between 0.2 to 0.6 (at 440 nm) over the

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western and central Mediterranean basins. Associated aerosol extinction values measured on-board the ATR-42 within the dust plume show local maxima reaching up to 150 Mm^{-1} . Non negligible aerosol extinction (about 50 Mm^{-1}) was also been observed within the Marine Boundary Layer (MBL). By combining ATR-42 extinction, absorption and scattering measurements, a complete optical closure has been made revealing excellent agreement with estimated optical properties. Associated calculations of the dust single scattering albedo (SSA) have been conducted, which show a moderate variability (from 0.90 to 1.00 at 530 nm). In parallel, active remote-sensing observations from the surface and onboard the F-20 aircraft suggest a complex vertical structure of particles and distinct aerosol layers with sea-salt and pollution located within the MBL, and mineral dust and/or aged north American smoke particles located above (up to 6–7 km in altitude). Aircraft and balloon-borne observations show particle size distributions characterized by large aerosols ($> 10 \mu\text{m}$ in diameter) within dust plumes. In terms of shortwave (SW) direct forcing, in-situ surface and aircraft observations have been merged and used as inputs in 1-D radiative transfer codes for calculating the direct radiative forcing (DRF). Results show significant surface SW instantaneous forcing (up to -90 W m^{-2} at noon). Associated 3-D modeling studies from regional climate (RCM) and chemistry transport (CTM) models indicate a relatively good agreement for simulated AOD compared with measurements/observations from the AERONET/PHOTONS network and satellite data, especially for long-range dust transport. Calculations of the 3-D SW (clear-sky) surface DRF indicate an average of about -10 to -20 W m^{-2} (for the whole period) over the Mediterranean Sea together with maxima (-50 W m^{-2}) over northern Africa. The top of the atmosphere (TOA) DRF is shown to be highly variable within the domain, due to moderate absorbing properties of dust and changes in the surface albedo. Indeed, 3-D simulations indicate negative forcing over the Mediterranean Sea and Europe and positive forcing over northern Africa.

line and vegetation cover of the region are well known to modulate the regional climate signal at small spatial scales (e.g. Millan et al., 1997; Gangoi et al., 2001; Lionello et al., 2006).

So far, most global and regional climate simulations have investigated the impact of global warming on the Mediterranean climate without detailed considerations of the possible radiative influence and climatic feedback from the different Mediterranean aerosols (anthropogenic, marine, biomass burning, secondary biogenic and mineral dust particles). The Mediterranean region is rich in a variety of particles (natural and anthropogenic) from both continental and marine sources (Lelieveld et al., 2002). In Fig. 1, we illustrate the significant differences in aerosol loading between the eastern, central, and western sub-basins and between the North and the South of the Mediterranean shown by long-term aerosol satellite products. The aerosol optical depth (AOD), which represents the integration of the extinction by particles along the whole atmospheric column displays annual mean values (Fig. 1) from 0.2 to 0.5 (in the visible wavelengths), depending on the aerosol types observed over the Euro-Mediterranean region (Nabat et al., 2013).

Numerous studies have documented the AOD for polluted-anthropogenic Mediterranean aerosols at local scale over southeastern France (Mallet et al., 2006; Roger et al., 2006), Spain (Horvath et al., 2002; Alados-Arboledas et al., 2008), Greece (Chazette and Liousse, 2001; Gerasopoulos et al., 2003), Crete (Fotiadi et al., 2006), and Italy (Tafuro et al., 2007; Ciardini et al., 2012). Under polluted conditions, they report moderate AOD values ranging between 0.1 to 0.5 (at visible wavelengths). In parallel, multi-year TOMS and MODIS observations over the eastern Mediterranean (Hatzianastassiou et al., 2009) or the Po Valley (Royer et al., 2010) indicate the occurrence of high AOD values (up to more than 0.8 at 500 nm) over large urban areas surrounding megacities.

Up to now and contrarily to anthropogenic polluted aerosols, only few studies have been dedicated to biomass burning aerosols over the Mediterranean, which are mainly observed in July and August (driest months of the year) when the development of forest

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planetary boundary layer height (decrease up to 30 % in the presence of anthropogenic aerosols) and local air-quality. In addition to their important effects on the surface and TOA DRF, most of the Mediterranean aerosols are also able to absorb more or less effectively the solar radiations leading to a significant atmospheric forcing and associated SW heating rate. Local studies previously mentioned (Roger et al., 2006; Saha et al., 2008; Pace et al., 2005; Pere et al., 2011; D. Meloni et al., 2015) clearly report significant SW heating rate due to absorbing particles with values reaching up to 2–3 K day⁻¹, depending on the aerosol types. Finally, aerosols also have a significant effect on photolysis rates that may affect tropospheric chemistry and ozone production over the basin (Casasanta et al., 2011; Mailler et al., 2015).

In regards to such surface, TOA and atmospheric forcings, there is a need to investigate how the change in the radiative budget due to natural/anthropogenic aerosols influence the surface temperature (both over land and sea), relative humidity profiles, exchanges (latent heat fluxes) between ocean and atmosphere, cloud-cover (semi-direct effect of absorbing particles), precipitation and finally the whole Mediterranean hydrological cycle. The induced perturbations in the sea surface–atmosphere fluxes is expected to be important despite the relatively small size of the Mediterranean Sea, since this basin plays an important role at much larger scale by providing moisture for precipitation to its surroundings land region extending to northern Europe and northern Africa (Gimeno et al., 2010 and Schicker et al., 2010). Indeed and as shown by Ramanathan et al. (2001) for the Indian region or Foltz and McPhaden (2008) and Yue et al. (2011) for the Atlantic Ocean, a modification of the sea-surface evaporative fluxes, due to the dimming radiative effect of aerosols at the sea surface could significantly influence the lower troposphere moisture content and the associated precipitation distribution around the Mediterranean. In parallel, the absorbing particles over the Mediterranean (Mallet et al., 2013) could exert a semi-direct effect that could modify the vertical profiles of relative humidity and cloud cover, which has to be quantified. In that context, the main objectives of the ChArMEx/ADRI MED project were the following:

the estimated DRF at the regional scale for the period of experiment. Longer (inter-seasonal and inter-annual) aerosol-climate simulations are not described here and can be found in Nabat et al. (2012, 2014, 2015a).

2 Overview of the surface observation network

The regional experimental set-up deployed in the western and central Mediterranean during the campaign ChArMEx SOP-1a is shown in Fig. 2.

2.1 The Cape Corsica and Lampedusa surface super sites

Two super-sites were fully equipped for documenting the aerosol chemical, physical and optical properties as well as their possible mixing and their vertical structure at local scale (Table 1). The main characteristics of these two surface stations are presented here. The first station was located in Ersa on Cape Corsica (42°58′10″ N, 09°22′49″ E), near the North tip of Corsica Island. This station was primarily instrumented for investigating polluted air masses transported over the Mediterranean basin from the highly industrialized regions of the Po Valley (Royer et al., 2010) and/or the Marseille-Fos-Berre (Cachier et al., 2005) zone and Rhone Valley. This ground-based remote station is located at an altitude of about 530 m above mean sea level (a.m.s.l.) on a ridge equipped with wind mills and benefit from a direct view to the sea over a North sector of ~ 270° extending from the SW to SE. The Cape Corsica peninsula is a remote site excluding important local anthropogenic sources that could affect the in-situ measurements.

The Lampedusa super-site (35°31′5″ N, 12°37′51″ E) was established at the “Roberto Sarao” station permanently operated by ENEA in the small island of Lampedusa (~ 20 km²), and it was augmented during the field campaign by the observations of the Portable Gas and Aerosol Sampling UnitS (PEGASUS) mobile station operated by LISA. This surface station was mainly used for documenting very aged air masses

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in south westerly flow from Europe, southern air masses from northern Africa (Tunisia, Algeria and Libya) possibly laden with mineral dust, as well as marine aerosols. It is situated on a cliff at about 45 m a.s.l. on the NE tip of the island.

The complete instrumentation deployed during the SOP-1a experiment for both super-sites is detailed in Table 1. Briefly, it served to determine the complete aerosol physical, chemical and optical properties as well as vertical profiles, and to measure radiative fluxes (broadband SW and LW, and spectral SW).

2.1.1 In situ measurements at super-sites

Both super-sites measured the mass concentration online using Tapered Element Oscillating Microbalance (TEOM) analysers. The number size distribution of particles are also measured, including fine and coarse fractions (radius ranges and corresponding instruments are reported Table 1). The aerosol composition was derived from chemical analyses of filters and cascade impactors (DEKATI and MOUDI) with time resolution varying from 12 to 48 h (depending on the aerosol load), but also from high-time resolution online measurements by an ACSM (Aerosol Chemical Speciation Monitor) at Ersa, a C-TOF-AMS (Time of Flight Aerosol Mass Spectrometer) at Lampedusa, and two PILS (Particle Into Liquid Sampler) systems at both sites (Table 1). Concerning aerosol optical properties, scattering and absorption coefficients (at wavelengths listed in Table 1) have been estimated for both super-sites using a 3 λ nephelometer and a 7 λ aethalometer, respectively. At Ersa station, the extinction coefficient (at 870 nm) was also estimated using a Photoacoustic Extinctionmeter (PAX) instrument, while it has been estimated at 2 λ (450 and 630 nm) at Lampedusa using 2 Cavity Attenuated Phase Shift Spectroscopy (CAPS) systems.

Additional in-situ measurements were performed at the Ersa station. The mixing state of fine particles (at the two selected diameters of 50 and 110 nm in dry conditions) has also been estimated from their hygroscopic behaviour using a VHTDMA (volatilization and humidification tandem differential mobility analyser) system (Johnson et al., 2004). In parallel, a TSI (model 3800) aerosol time of flight mass Spectrom-

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eter (ATOFMS) (Gard et al., 1997) was used to measure the size-resolved chemical composition of single particles in the vacuum aerodynamic diameter (d_{va}) size range 100–3000 nm.

2.1.2 Remote sensing and radiation measurements at super-sites

A Leosphere Raman lidar model RMAN510 was setup at low altitude (~ 11 m a.s.l.) in the small village of Macinaggio (42°57'44" N, 9°26'35" E) located on the eastern coast of Cape Corsica. The lidar was operated at about 6 km East from the Ersa station and less than 700 m from the shoreline. The RMAN510 uses a laser emitting at 355 nm. It measures the total and polarized backscatter at 355 nm and the Raman nitrogen signal at 387 nm at night-time. A second ALS300 510 lidar system has been deployed in Lampedusa (Formenti et al., 2015) as well as a more powerful University of Rome-ENEA homemade lidar measuring backscatter at 532 and 1064 nm (Di Iorio et al., 2015). The main characteristics of lidar systems are provided and detailed in Table 1.

At each station, a multi-wavelength sun-photometer from the AERONET/PHOTONS network was operated, allowing the operational retrieval of column integrated AOD at 340, 380, 440, 500, 675, 870, 1020 nm (and also at 1650 nm at Ersa) and aerosols optical and microphysical properties such as the single scattering albedo, refractive index and particle size volume distribution (Dubovik and King, 2000; Dubovik et al., 2000, 2002, 2006). The Ersa sun-photometer is positioned since June 2008 near the navy semaphore on the northwestern tip of Cape Corsica (43°00'13" N, 09°21'33" E, alt. ~ 75 m a.m.s.l.) at about 4.2 km NNW of the Ersa surface station.

Both super-sites were complemented by a pyrgeometer and a pyranometer for monitoring longwave and shortwave downward fluxes measurements, respectively. Additional radiation measurements were performed at Lampedusa (Table 1). Spectral measurements of global, diffuse, and direct radiation were carried out with other instruments deployed by ENEA and the Physikalisch-Meteorologisches Observatorium Davos, World Radiation Center, (PMOD/WRC, Switzerland). Multi-filter rotating shadowband radiometer observations were carried out jointly with AERONET sun-

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photometer (di Sarra et al., 2015) and allowed the derivation of the AOD at several wavelengths. By combining these two measurements, a long-term series of AOD, started in 2001, was obtained. Measurements of the spectral actinic flux, allowing the determination of the photolysis rates (Mailler et al., 2015), were carried out with a diode array spectrometer. Measurements of broadband irradiance included a CG3 pyrgeometer sensitive to radiation in the atmospheric infrared window. Finally, the total ozone and spectral UV irradiance were obtained with a Brewer spectrophotometer. Several radiosondes were also launched from Lampedusa during the SOP-1a, and vertical profiles of temperature and humidity were continuously measured by a microwave radiometer.

2.2 The secondary sites

2.2.1 Montesoro station

The Cape Corsica station was complemented by an additional remote-sensing setup at the peri-urban air quality station of Montesoro, southward of Bastia at about 45 m a.s.l. (Leon et al., 2015), including a Leosphere model EZ lidar operating at 355 nm (42°40′17″ N, 09°26′05″ E) and a Cimel AERONET/PHOTONS sun-photometer (42°40′19″ N, 09°26′06″ E). In addition, some air-quality parameters were monitored by Qualitair Corse, including PM_{2.5} and PM₁₀. This station is less than 1 km far from the shore on the northeastern coast of Corsica, about 32 km South of Macinaggio.

2.2.2 Barcelona station

The Barcelona station (41.39° N, 2.11° E, 115 m a.s.l.) was equipped with the following fixed instruments including an AERONET sun-photometer, an automated Sigma Space-NASA Micro Pulse Lidar (MPL) and a Universitat Politècnica de Catalunya (UPC) home-made multi-wavelength lidar (Kumar et al., 2011). The MPL lidar works

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at 532 nm and has a depolarization channel, while the UPC lidar works at 355, 532 and 1064 nm, and also includes two N₂- (at 387 and 607 nm) and one H₂O-Raman (at 407 nm) channels. The MPL system worked continuously. The UPC system was operated on alert in coordination with the two research aircraft plans involved in the SOP-1a campaign. The UPC system is part of the EARLINET network.

2.2.3 Minorca station

An additional station was setup during the campaign, located at Cap d'en Font, on the southeastern coast of the Balearic island of Minorca (Spain, 39°53'12" N and 4°15'31" E, ~ 10 ma.m.s.l.), which is relatively central in the western Mediterranean basin. The Mobile Aerosol Station (MAS) of the LSCE (Laboratoire des Sciences du Climat et de l'Environnement) laboratory was equipped with the new Raman lidar WALI (Chazette et al., 2014a, b), an AERONET/PHOTONS sun-photometer, and a set of in-situ instruments. A 5-wavelength Solar Light Microtops-II manual sun-photometer was also used. The WALI instrument, its calibration and the associated errors are documented in Chazette et al. (2014a). During all the experiment, the acquisition was performed continuously with a vertical resolution of 15 m. AOD at the lidar wavelength of 355 nm has been extrapolated from that measured by sun-photometer at 380 and 440 nm using the Angström exponent (Chazette et al., 2015).

The in-situ instruments installed on-board the MAS included a 3-wavelength TSI nephelometer, a Magee Scientific Model AE31 7-wavelength aethalometer, a TEOM microbalance, and a Vaisala meteorological probe type PTU300. The nephelometer was sampling through a PM₁₀ inlet to measure the aerosol scattering coefficient at 3 wavelengths (450, 550 and 700 nm) with an integrating time step of 5 min. The aethalometer was sampling through a PM_{2.5} inlet to measure aerosol absorption (at 7 wavelengths) and derive a 5 min average black carbon concentration. The TEOM measured dry PM₁₀ concentration every 30 min. In addition two optical particle counters (OPCs) were installed outdoors next to the sun-photometer on a mobile platform. A MetOne HHPC-6 and a LOAC (Renard et al., 2015a, b) respectively measured

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aerosol particle number concentration in 6 channels above 0.3 μm in diameter and in 19 channels above 0.2 μm . The LOAC instrument accuracy is discussed in detail by Renard et al. (2015a, b).

2.2.4 Granada station

5 The station of the Atmospheric Physics Group (GFAT) is located in the Andalusian Institute for Earth System Research (IISTA-CEAMA), in Granada, Spain (37.16° N, 3.61° W, 680 m a.s.l.). The station is at a relatively short distance, about 200 km away, from the African continent and approximately 50 km away from the western Mediterranean Sea. During the SOP-1a campaign, lidar measurements were performed simultaneously with a multiwavelength Raman lidar and a scanning Raman lidar both from Raymetrics S.A. The multi-wavelength Raman system is part of the EARLINET network. In addition, a ceilometer was operated. Column integrated characterization of the atmospheric aerosol was performed following AERONET protocols with two Cimel sun-photometers deployed at two different heights: Granada (680 m a.s.l.) and Cerro Poyos (37°6'32" N, 03°29'14" W, 1790 m a.s.l.) stations. In addition, in-situ instrumentation was continuously operated providing measurements of aerosol light-absorption coefficient at multiple wavelengths (multi-angle absorption photometer (MAAP) from Thermo ESM Andersen Instruments and Aethalometer model AE31), size distribution and particle number concentration for diameters larger than 0.5 μm (TSI aerodynamic particle sizer APS model 3321) and light-scattering and backscattering coefficient at dry and at relative humidity of 85 % by means of a TSI tandem nephelometer humi-
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dograph system. Furthermore, the chemical composition in the PM₁ and PM₁₀ size fractions was determined during 16 and 17 June by collecting aerosol samples using two high-volume samplers.

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2.2.5 Capo Granitola station

Several instruments were also deployed at Capo Granitola (37°34' N, 12°40' E), a site along the Southern coast of Sicily. The site, within a combined effort of ENEA, Univ. of Florence, and Univ. of Valencia, was equipped with a PM₁₀ sampler, a MultiFilter Rotating Shadowband Radiometer (MFRSR) to derive spectral AOD, and radiometers and spectrometers for the measurement of global, direct, and diffuse radiation throughout the SW and LW spectral ranges.

2.3 Surface remote-sensing network

Two surface remote-sensing networks were operated during the ChArMEx SOP-1a experiment, namely the AERONET/PHOTONS and EARLINET/ACTRIS (Pappalardo et al., 2014) networks. These networks were highly useful as they allow estimating the column-integrated aerosol loading as well as the vertical structure of particles.

2.3.1 The AERONET/PHOTONS Sun-Photometer Network

AERONET (Aerosol Robotic Network; <http://aeronet.gsfc.nasa.gov/>) is a federated network of ground-based sun-photometers and the associated data inversion and archive system, that routinely performs direct sun observations about every 15 min during daytime, and both almucantar and principal plane sky radiance measurements, at selected solar angles (Holben et al., 1998). Along with AOD observations, the AERONET aerosol retrieval algorithm (Dubovik and King, 2000) delivers the complete set of column-effective aerosol microphysical parameters, including volume size distribution, refractive index at several wavelengths and fraction of spherical particles (Dubovik et al., 2006, see also description in Dubovik et al., 2011). In addition, using these microphysical parameters, the algorithm provides other column-effective aerosol optical properties such as wavelength dependent SSA, phase function, and asymmetry parameter, as well as integral parameters of bi-modal particle size distributions (concen-

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tration, mode radii and variances) (Dubovik et al., 2002). The accuracy of AERONET retrievals is evaluated and discussed by Dubovik et al. (2000, 2002). In addition to microphysical and optical aerosol properties, we also have used direct radiative forcing calculations operationally provided at any AERONET location as an operational product of the network. The method of derivation is described in detail by Garcia et al. (2012). Briefly, the broadband fluxes were calculated using the radiative transfer model GAME (Dubuisson et al., 2004; Roger et al., 2006) that has been integrated into operational AERONET inversion code. Sun-photometer stations deployed during the SOP-1a campaign over the Western basin are listed in the Table 2.

2.3.2 The EARLINET/ACTRIS network

Between 22 and 24 June, four ACTRIS/EARLINET lidar stations located in Italy, in addition to the EARLINET stations of Barcelona and Granada, were operated in support of aircraft operations (Sicard et al., 2015):

- Naples (40.84° N, 14.18° E, 118 m a.m.s.l.); measurements of backscatter profiles at 355 and 532 nm, as well as depolarization ratio profiles at 532 nm, on 22 June 2013.
- Serra La Nave (Sicily, 37.68° N, 14.98° E, 1735 m a.m.s.l.); measurements of backscatter profiles at 355 nm, as well as depolarization ratio profiles at 355 nm, on 22 June 2013.
- Potenza (40.60° N, 15.72° E, 760 m a.m.s.l.); measurements of extinction profiles at 355 and 532 nm, backscatter profiles at 1064 nm, as well as depolarization ratio profiles at 532 nm (Madonna et al., 2011), on 22 and 23 June 2013.
- Lecce (40.30° N, 18.10° E, 30 m a.m.s.l.); measurements of extinction profiles at 355 and 532 nm, backscatter profiles at 1064 nm, water vapour profiles, as well as depolarization ratio profiles at 355 nm, on 22 and 24 June 2013.

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SEVERI sensors and averaged for the June–July 2013 period (Fig. 5), which show an average AOD ranging between 0.2 and 0.4 (at 550 nm) over the western and central Mediterranean basins. During the SOP-1a, distinct meteorological conditions have led to the transport of mineral dust over the basin as shown in the Figs. 5 and 6. Figure 7 shows the dust mass concentration together with the geopotential and wind at 700 hPa for the 16, 19, 22, 29 June and 2 July. In the following sections, we discuss the meteorological conditions (surface wind, sea level pressure, 700 hPa geopotential and wind direction) for these different days in order to understand the transport of both mineral dust and anthropogenic aerosols over the Mediterranean.

Wind direction vertical profiles as simulated by the ALADIN regional model (outputs every 3h) as a function of time, for the 11 June to 6 July period and for the whole SOP-1a period at three different sites: Ersa, Minorca and Lampedusa islands are shown in Fig. 8. At the beginning of the SOP-1a, the northwestern Mediterranean area was under the influence of a large pressure ridge at 700 hPa, generating a westerly to south-westerly flow over Spain and southern France. Over Minorca, the near surface (1000–850 hPa) winds were generally from the easterly to north-easterly direction (indicated by the blue color in the Fig. 8) while the wind direction estimated between 700 and 500 hPa was clearly from the south, southwest direction (brown color), which is a favourable condition for the transport of mineral dust above South-Spain and then Balearic islands (Fig. 6). This point is well observed in Fig. 7, showing the geopotential at 700 hPa for the 16 June. The general circulation at 700 hPa during this dust event indicates a reinforcement of the southwesterly winds in southern Spain advecting air masses with large concentrations of dust aerosols as shown by SEVERI AOD for that day (Fig. 6). A low pressure system moved from the British Isles towards the Gulf of Biscay and then the Iberian Peninsula between the 17 and 20 June, leading to veering winds that became southerly over the northwestern Mediterranean. Thus in Minorca, the direction of the wind changed from easterly to southerly direction between 1000 and 850 hPa. A more pronounced southerly-southwesterly flow was also observed at 700 hPa in Minorca (19–21 June) as shown by the geopotential at 700 hPa. This cir-

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5 culation characterized by the presence of the low geopotential over the Gulf of Biscay induced a strong southerly flow at 700 hPa between the Balearic and Corsica islands associated with large dust optical depth concentrated in this zone as shown by SEVERI AOD for 19 June (Fig. 6). This period of the SOP-1a corresponds to the two ATR-42
10 flights 33 and 34 (Fig. 3). After 20 June, this low pressure system moved eastward, generating a trough located between France and Italy, and inducing a waving westerly flow over the north-western Mediterranean. As a result, the aerosol loading over the western basin decreased between 21 and 24 June, but the westerly (resp. northerly) winds observed at 700 hPa in Minorca (resp. Ers) (Fig. 8) reinforced the transport of
15 dust aerosols over the central basin and the Lampedusa station (where winds were from the north westerly direction at 3 km height). These meteorological conditions lead to an increase of the dust optical depth over the central Mediterranean as shown by the SEVERI instrument and AERONET/PHOTONS data. Between 25 and 29 June, a northwesterly flow set up between the Gulf of Lions and Sicily. The vertical profiles of the wind direction reveal a remarkable transition on 29 June with significant changes in direction from westerlies to north, north-westerlies, notably over the Minorca and Ers stations above 850 hPa. The 700 hPa geopotential field on 29 June at 12:00 UTC from the ALADIN atmospheric model analysis shows a maximum over the Atlantic Ocean whereas a deep low pressure system was located over southern Algeria. This strong
20 geopotential gradient lead to intense northerly to north-westerly winds at 700 hPa over the western basin leading to significant AOD over Libya and the Alboran sea as shown in Fig. 6. These meteorological conditions lead to low dust optical thickness over the central Mediterranean as observed by AERONET/PHOTONS data. Finally, during the last period of the SOP-1a experiment, (30 June–5 July), weather conditions became more anticyclonic over the region while low systems were confined to northern Europe.
25 Figure 8 shows north-westerly winds in the whole troposphere in Lampedusa and Minorca, limiting the presence of dust aerosols to the southern part of the north-western Mediterranean.

4.2 Surface temperature, cloud cover and precipitation

In terms of surface temperature, which is one of the most important meteorological variables that control biogenic or biomass burning aerosol emissions over the Euro-Mediterranean region, the summer 2013 was mostly characterized by moderate values as shown in Fig. 9. Indeed, during the SOP-1a period, surface temperatures (in °C and at 12:00 UTC) derived from NCEP reanalysis for different days reveal moderate values especially over the western Mediterranean region (South-West France and Spain). One can observe temperatures of about 15–20 °C (at 12:00 UTC) over Spain and Portugal, which are one of the main regions of the Mediterranean where large fire events occur. In addition, part of France was also characterized by moderate surface temperature but slightly higher than over Spain especially over northeastern regions. A strong west to east gradient is observed over Europe with strongest values over the eastern regions (around 30 °C over Greece and the Balkans) compared to the western basin. A similar conclusion is obtained over the Mediterranean Sea with differences of about 5 °C between the eastern (around 25 °C for the SOP-1a period) and the western (around 20 °C) basin. Among other factors (such as cloud fraction and shortwave radiations), such moderate surface temperatures do not create favourable meteorological conditions to produce intense Mediterranean biomass burning events and/or significant production of secondary organic and inorganic aerosols. Concerning smoke aerosols, GAFS-V1 emission data, analysed for the SOP-1a period, do not reveal important primary BC and OC fluxes emissions (not shown). This is consistent with the APIFLAME biomass burning emission estimates (Turquety et al., 2014) data as reported by Menut et al. (2015).

During the SOP-1a, the cloud cover retrieved over the Euro-Mediterranean region (excluding the Mediterranean Sea) from CRU (Climate Research Unit) data (Fig. 10) indicates the largest values (between 75 and 95 %) over France, Benelux and Eastern Europe regions. In parallel, southern France, as well as western Spain and the Balkans are characterized by moderate cloud cover with values around 50–60 % for

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June 2013. Over the Mediterranean coast, the cloud cover strongly decreases for most of countries, with values lower than 40 %. Such spatial cloud cover (observed during the SOP-1a) over the Euro-Mediterranean could limit the photochemical processes over the main anthropogenic sources (such as the Benelux and Po Valley) and the associated production of secondary aerosols. This could explain for a part the low to moderate contribution of fine anthropogenic particles to the total atmospheric loading during the SOP-1a. In parallel, the mean precipitation (averaged for June 2013), obtained from the TRMM (Tropical Rainfall Measuring Mission) instrument over land and sea (CRU observations are only available over land, see Fig. 10), are found to be very heterogeneous over the Euro-Mediterranean continental region, with some important values over the Balkans, Alps and eastern Europe (from 100 to 250 mm for the month of June 2013) and moderate values over Italy, Croatia, western France and Benelux (80 to 100 mm, as shown in the Fig. 11). Over the Mediterranean Sea, southern Spain and northern Africa, the precipitation was smaller, with most of values lower than 20 mm during the SOP-1a.

To summarize, this global view of the synoptic situation, cloud cover and regional precipitation patterns indicate that the meteorological conditions during the experimental campaign were favourable to moderate mineral dust emissions, associated with a weak contribution of anthropogenic aerosols over the western basin. This important characteristic of the SOP-1a is well observed in Fig. 12, which indicates the AOD anomalies of summer 2013 compared to all AOD summer derived from MODIS and MISR data. Indeed, negative AOD anomalies of about -0.05 are found over the western Mediterranean basin for the summer 2013, both from MODIS and MISR observations. To conclude, it appears that the period of observations during the SOP-1a was characterized by aerosol concentration slightly lower than usually observed during summer over the western Mediterranean. The level of aerosol concentration was found to be moderate but allows investigating several dust and sea-salt events as well as an unexpected and interesting intense biomass burning plume advected from North America.

4.3 An aged smoke plume advected over Europe

During the SOP-1a, several large forest fires occurred in North America (Colorado, Alaska, Canada) from 17 to 24 June 2013, as identified by the MODIS instrument. Absorbing aerosol index produced from GOME-2 by KNMI (<http://www.temis.nl/aviation/aai-pmd-gome2b.php?year=2013>) shows that a large smoke plume crossed the north Atlantic and reached Western Europe coasts on 25 June. Main fire areas, with fire radiative power higher than 50 MW (Shroeder et al., 2010), have been detected over Canada (Ancellet et al., 2015). Average MODIS AOD during the same period (23 to 28 June 2013) indicate values as high as 1 over the Atlantic Ocean, suggesting that a significant fraction of the aerosol produced by the fires was transported to Western Europe during the ChArMEx/ADRIMED field campaign. To investigate if the western Mediterranean has been impacted by these fires, a forward simulation of the Lagrangian plume dispersion model FLEXPART (Ancellet et al., 2015) has been conducted to quantify the spatial extent of the fire plume transport for 11 days. Fires emissions areas were identified by MODIS observations over several locations in Canada and Colorado. The aerosol mass is emitted in the transport model from 17 June to 28 in a 3 km layer as suggested by the CALIOP lidar observations over Canada. The biomass burning plume reaches much lower latitudes over Europe, down to the Western Mediterranean 4–10 days after the emission in Canada. During the SOP-1a, the plume was mainly present in the altitude range of 2.5–4.5 km and has been sampled by many remote sensing and in-situ instruments on 27 and 28 June; at Minorca and Cagliari surface stations, and between Sardinia and Lampedusa onboard the ATR-42 aircraft.

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5 Overview of aerosol physical-chemical-optical properties, vertical profiles and local direct radiative forcing

5.1 Aerosol physical and chemical properties

5.1.1 Aerosol mass and number concentration at the two super-sites

5 First, PM concentrations between the two different stations are reported in the Fig. 13, which reports the daily time-series of PM₁ and PM₁₀ at Ersa, as well as PM₁₀ and PM₄₀ at Lampedusa. The results indicate a significantly higher mass concentration at Lampedusa compared to Ersa. Indeed, the mass concentration observed at Lampe-
10 dusa is comprised between 10 and 75 $\mu\text{g m}^{-3}$, with a mean of 37 $\mu\text{g m}^{-3}$, which is 4 times higher than the averaged PM₁₀ ($\sim 9 \mu\text{g m}^{-3}$) measured at Ersa. One can note the significant peak (maxima of 75 $\mu\text{g m}^{-3}$) at Lampedusa during the 24 to 26 June period that corresponds to a significant production of primary marine aerosols. Finally, the PM₁ concentration at Ersa is found to be almost constant during the period of the
15 campaign, with a mean value of 6 $\mu\text{g m}^{-3}$. In addition, the background aerosol number concentrations (for $D_p > 0.01 \mu\text{m}$) observed within the boundary layer in Corsica averaged $\sim 2000 \text{ cm}^{-3}$ (not shown). The lowest concentrations ($\sim 200 \text{ cm}^{-3}$) resulted from aerosol activation to cloud droplets, and scavenging from cloud droplets and rain drops, while high concentrations as high as 10 000 cm^{-3} were observed during pollu-
20 tion events from continental European air masses. The number concentrations showed a diurnal cycle suggesting that the site was situated within the marine boundary layer during daytime and within the free troposphere during night-time. The analysis of the diurnal variation of the particle number size distribution is further indicating that nuclea-
25 tion events also increased the particle number concentration during daytime, about one third of the time (Sellegrì et al., 2015). The periods of high aerosol number concentrations detected between the 12 and 25 June were also dominated by a single mode with diameters between 30 and 150 nm. The small Aitken mode ($d_g < 50 \text{ nm}$) as-

sociated with pollution events suggests a relatively fresh aerosol that has been formed during transport from the European continent. The largest mode ($d_g \sim 150$ nm) occurred during the dust event on 18 June.

5.1.2 Columnar particle volume size distribution

We have used the column-integrated particle size volume distributions derived from AERONET/PHOTONS sky radiance measurements (Dubovik et al., 2000). These size distributions allow investigating the changes in aerosol size distribution between different stations during the SOP-1a and over the western basin. Four different stations have been studied, which include the two super-sites of Lampedusa and Ersa, as well as the aircraft and balloon base stations; Cagliari and Cap d'En Font, respectively. Daily volume size distributions for both sites are represented in the Fig. 14, as well as the averaged (red curve) size distribution for the whole period (1 June to 5 July) and the number of observations. In addition, the mean values of the volume radius, concentration of fine and coarse mode and the standard deviations of the volume size distribution are reported in the Table 6. It should be noted that the scales of the y axis are different for each figure. One can note the bimodal size distribution for both stations with large spread of radius values, especially for the coarse mode. The most important concentrations are obviously observed in Lampedusa, near the mineral dust sources, with maxima of $\sim 0.12 \mu\text{m}^3 \mu\text{m}^{-2}$ for the coarse mode. In parallel, the lowest concentrations are observed at the Ersa station, near the anthropogenic sources of the southern France and Italy. This is well consistent with the absence of intense polluted-photochemical or smoke aerosol events during the SOP-1a. In that sense, the mean contribution (red curve) of the coarse mode to the aerosol volume size distribution appears to be predominant at most sites, except at the Ersa station. This point is well noted for the Cap d'En Font station, where the concentration of each modes appear as equivalent, due to the absence of pollution from the Iberian Peninsula during the period of observations. For this site, it is interesting to note the intense peak for the 27 June, with concentration near $0.08 \mu\text{m}^3 \mu\text{m}^{-2}$, which is due to the transport of an

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important smoke plume over the Mediterranean (see Ancellet et al., 2015 and Pelon et al., 2015). Finally, the contribution of the coarse mode clearly increases for the two other, more southern Italian sites of Cagliari and Lampedusa, which are more affected by the mineral dust compared to Ersa and Cap d'En Font. The variability of AERONET products collected over a period of four years at Ersa and Palma de Mallorca, near Cap d'En Font, is reported in Sicard et al. (2015). It is interesting to note the important variability in the derived size of the coarse mode at Lampedusa (see Table 6), which will be analysed in regards to dust sources in a future study. The derived volume concentrations over these two stations highlight the moderate dust activity occurring during the SOP-1a experiment, when compared to stations under high dust conditions. As an example of comparisons, Dubovik et al. (2002) reported a large range of concentration for the coarse mode for dusty sites (such as Cape Verde or Solar Village), which are characterized by larger concentrations, close to $0.30 \mu\text{m}^3 \mu\text{m}^{-2}$. In parallel, the Bahrain (Persian Gulf) AERONET station is characterized by a concentration of $0.14\text{--}0.15 \mu\text{m}^3 \mu\text{m}^{-2}$.

5.1.3 Particle size distribution during transport

Figure 15 presents an example of the evolution of the aerosol particle number concentrations in the 19 particle size classes of the LOAC instrument as measured along the northward trajectory of the BPCL balloon B74 from Minorca Island to the French coast (see Fig. 4). The balloon was launched at 09:46 UTC on 16 June 2013 during a moderate desert dust event shown on top of Fig. 6 (AERONET-derived AOD at 500 nm of 0.15). It drifted at a constant altitude of ~ 2.1 km at the bottom of the African dust layer observed with the WALI lidar at Minorca (not shown; see Chazette et al., 2015), and was automatically forced to land on the sea before reaching the coast South of Marseille, after a 12 h flight of 368 km. The dominant mineral dust nature of the particles was confirmed by the LOAC particle typology measurements (Renard et al., 2015). The figure illustrates that LOAC has detected large particles of up to $50 \mu\text{m}$ in diameter, although the plume originated from North-Africa a few days before (Renard et al.,

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2015b). The concentrations of particles remained relatively constant during the flight, suggesting either no significant sedimentation of the largest particles during the flight or compensation by particles coming from above. The BPCL balloon B70 launched a few minutes later drifted at an upper altitude of ~ 3.1 km and followed a different trajectory towards East (Fig. 4) but showed a quite similar extended particle size range with larger concentrations in almost all channels except the extremes (not shown). The 4 other drifting balloons launched in the dust layer during this event on 17 and 19 June (Table 5) did confirm the presence of very large particles ($> 20 \mu\text{m}$), which cannot be reported by AERONET particle size distribution retrieval algorithm (Hashimoto et al., 2012). In addition, observations of large particles ($> 15 \mu\text{m}$) was systematically found during all other LOAC balloon flights drifting in African dust layers, which will need further analysis to better understand the process that can maintain such large particles in suspension during several days.

5.1.4 Aerosol chemical composition

15 In terms of aerosol chemical properties, an example of averaged mass-size distributions for carbonaceous (Elemental and Organic Carbon, EC and OC) species (mass size distribution of inorganic and mineral dust aerosols are not shown) obtained at Ersa from a 12-stage cascade impactor (DEKATI system, see Table 1) is reported in Fig. 16. As mentioned in Table 1, the measurements were obtained by using a 2 day collection period in order to obtain a sufficient aerosol mass on filters for chemical analyses. 20 This system provides the speciation of the mass size distribution, including fine and coarse fractions. Such information is very useful to derive optical properties using Mie calculations (Mallet et al., 2011) for the main particle types (sulfates, ammonium, nitrates, sea-salt, dust, black and organic carbon). This provides crucial information's on key radiative properties which are classically used in regional climate models (mass 25 extinction efficiencies, SSA and asymmetry parameter). Furthermore, it allows one to assess the spectral dependence of radiative properties, which cannot always be estimated from in-situ instrumentation.

5.2 Aerosol optical properties

5.2.1 In-situ optical properties at the surface

Figure 17 reports the (daily mean) time-series of nephelometer observations obtained at the surface for the Ersa and Lampedusa stations during the SOP-1a. Daily scattering coefficients (at the three nephelometer wavelengths of 450, 550 and 700 nm) are reported, as well as the scattering Angström exponent (AE) calculated between 450 and 700 nm. At 550 nm and at Ersa, the scattering coefficient presents a significant variability during the SOP-1a with peaks of about $35\text{--}40 \text{ Mm}^{-1}$ during the dust event (19–20 June) transported over the Corsica island, associated to low values (15 Mm^{-1}) for certain periods of time, as for 21–22 June. The mean scattering coefficient (at 550 nm) is 24 Mm^{-1} . Such scattering coefficient values are comparable to observations reported by Vaishya et al. (2012) at the Mace Head station for Atlantic marine air, with scattering coefficient (at 550 nm) comprised between 10 and 25 Mm^{-1} during the summer period. In terms of scattering spectral dependence, the calculated scattering AE is found to be almost constant, with $\text{AE} \sim 1.5\text{--}2$ and a mean value of 1.71 (indicating that scattering is mostly dominated by fine aerosols) during the SOP-1a, except for the 23–24 June. The lowest values ($\text{AE} \sim 0.3\text{--}0.5$) observed during this period are the result of a large contribution of coarse sea-salt aerosols (Claeys et al., 2015), which is also observed from the filter chemical size-resolved analyses and detected on the A-TOFMS and VHTDMA data. In parallel, we observe that the dust event occurring in Ersa on 18–20 June is not correlated to low scattering AE, revealing a possible contribution of fine dust particles only to scattering, result of a possible deposition of the coarse dust fraction during transport. The AERONET-derived AE between 440 and 870 nm shows values < 1 in the afternoon of 19 June and early morning of 20 June suggesting that coarse dust is present in the column. At Lampedusa, where the nephelometer was sampling from a PM_{40} inlet, the daily scattering coefficient (at 550 nm) is between 20 to 90 Mm^{-1} (mean value of 50 Mm^{-1}), which is twice higher than at Ersa (Fig. 17). The scattering

well as their spectral dependence are presented and discussed in details by Denjean et al. (2015) and Nicolas et al. (2015). The airborne SW and LW radiation measurements and the comparison with radiative transfer model simulations at Lampedusa are presented by M. Meloni et al. (2015).

5.3 Aerosol vertical structure

5.3.1 Lidar surface observations

Although deeply analysed in other dedicated papers, some examples of the aerosol vertical profiles are presented here. First and over the Minorca station, surface lidar observations in Fig. 21a were obtained during 16 and 17 June, that corresponds to the first event of transported mineral dust over the western basin. They show a dust aerosol layer located between 1.5 and 5 km, with a maximum of aerosol extinction (at 355 nm) around 0.10 km^{-1} on 16 June between 12:00 and 14:00 LT. Comparisons of retrieved AOD with the lidar system is shown to be very consistent with sun-photometer observations for these two days (Fig. 21a, top), with moderate AOD (at 355 nm) comprised between 0.2 and 0.4 at maximum. During 17 June, the dust layer is less intense and the aerosol extinction above 1.5 km decreases. After 14:00 LT, Fig. 21a clearly shows that most of the contribution to AOD is due to the MBL over the Minorca station. At Ersa (Fig. 21b), the dust event reached the northern tip of Corsica on 19 June. A deep depolarizing aerosol layer was observed at altitudes between 3 and 6 km. In the night of the 20, the particulate depolarization ratio is close to 18 % and the lidar ratio within the dust layer was estimated at 46 sr. The extinction coefficient remains moderate within the dust layer $\sim 0.05 \text{ km}^{-1}$ (Fig. 21b) between 4 and 6 km. It should be noted that a complete analysis of lidar observations series obtained over the cape Corsica site is reported in Leon et al. (2015). The dust event vertical distribution is further analysed by means of the EARLINET lidar stations in Sicard et al. (2015) and by means of the EARLINET and ChArMEx lidar stations in Barragan et al. (2015).

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6 Overview of modeling activities

Several models are used to analyze the SOP-1a period: the meso-scale meteorological COSMO-MUSCAT model, the chemistry transport model (CTM) CHIMERE model, and two regional climate (RegCM and CNRM-RCSM) models. These models differ in terms of horizontal and vertical resolutions, physical parameterizations, aerosol-chemical schemes and are able to deliver complementary information to address key scientific questions of the ChArMEx/ADRIMED experiment. Their main characteristics are summarized in the Table 8.

6.1 COSMO-MUSCAT model

The parallelized multi-scale regional model system COSMO-MUSCAT (Wolke et al., 2012) consists of the non-hydrostatic atmosphere model COSMO (Consortium for Small-scale Modelling) that is on-line coupled to the 3-D chemistry tracer transport model MUSCAT (MULTiScale Chemistry Aerosol Transport Model). The atmospheric dust cycle consisting of the emission, transport and deposition of dust particles is simulated within MUSCAT using meteorological and hydrological fields from COSMO. Dust emission is calculated using the emission scheme by Tegen et al. (2002) and depends on local surface wind friction velocities, surface roughness length, soil texture and soil moisture. Calculated dust emission fluxes depend on particle diameter for individual size classes that are assumed to be log-normally distributed. Following Marticorena and Bergametti (1995), dust emission is considered as threshold function of local friction velocities and thus initial dust emission is computed as a function of soil particle size distribution. Dust emission is limited to regions where active dust sources have been identified during 2006–2009 from MSG SEVIRI observations (Schepanski et al., 2007). The advection of dust particles is described by a third order upstream scheme; dust particles are transported as passive tracer in five independent size classes with limiting radius at 0.1, 0.3, 0.9, 2.6, 8, and 24 μm . The removal of dust particles from the atmosphere is described by dry and wet deposition taking particle size, particle density,

using the new high resolution fire model presented in Turquety et al. (2014). Finally, the photolysis rates are explicitly calculated using the FastJ radiation module (Mailler et al., 2015).

6.3 The RegCM regional climate model

5 The RegCM system is a community model designed for use by a varied community composed of scientists in industrialized countries as well as developing nations. It is supported through the Regional Climate Network, or RegCNET, a widespread network of scientists coordinated by the Earth System Physics section of the Abdus Salam International Centre for the Theoretical Physics (ICTP, Giorgi et al., 2012). RegCM
10 is a hydrostatic, compressible, sigma-p vertical coordinate model. As a limited area model, RegCM requires initial and boundary conditions that can be provided both by NCEP or ECMWF analyses. The horizontal resolution used need to be higher than 10 km, due to the hydrostatic dynamic core of the model, associated with 23 vertical levels. A simplified aerosol scheme specifically designed for application to long-term climate simulations has been incrementally developed within the RegCM system. Solomon et al. (2006, 2008) first implemented a first-generation aerosol model including sulfates, organic carbon, and black carbon. Zakey et al. (2006) then added a 4-bin desert dust module, and Zakey et al. (2008) implemented a 2-bin sea-salt scheme. In RegCM, the dust emission scheme accounts for sub-grid emissions by different types of soil. The
20 dust emission size distribution can now also be treated according to Kok (2011). When all aerosols are simulated, 12 additional prognostic equations are solved in RegCM, including transport by resolvable scale winds, turbulence and deep convection, sources, and wet and dry removal processes. In RegCM, the natural/anthropogenic aerosols are radiatively interactive both in the solar and infrared regions and so are able to feedback
25 on the meteorological fields.

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6.4 The CNRM-RCSM Regional Climate model

The fully coupled RCSM (Regional Climate System Model), which is developed at CNRM has been also used within the ChArMEx/ADRMED project. This model includes the regional climate atmospheric model ALADIN-Climate (Déqué and Somot, 2008), the regional ocean model NEMOMED8 (Beuquier et al., 2010) and the land-surface model ISBA (Noilhan and Mahfouf, 1996). We used here the version described in Nabat et al. (2015b) with a 50 km horizontal resolution. ALADIN-Climate includes the Fouquart and Morcrette radiation scheme based on the ECMWF model incorporating effects of greenhouse gases as well as direct effects of aerosols. The ocean model NEMOMED8 is the regional eddy-permitting version of the NEMOV2.3 ocean model that covers the Mediterranean Sea. Concerning the aerosol phase, the model ALADIN-Climate incorporates a radiative scheme to take into account the direct and semi-direct effects of five aerosol types (sea-salt, desert dust, sulfates, black and organic carbon aerosols) through either AOD climatologies or a prognostic aerosol scheme (Nabat et al., 2013, 2015b). On the one hand, Nabat et al. (2013) have proposed a new AOD monthly climatology over the period 2003–2009, based on a combination of satellite-derived and model-simulated products. The objective is having the best estimation of the atmospheric aerosol content for these five most relevant aerosol species. On the other hand, a prognostic aerosol scheme has been recently implemented in ALADIN-Climate, and has shown its ability to reproduce the main patterns of the aerosol variability over the Mediterranean (Nabat et al., 2015b).

6.5 SOP-1a multi-model aerosol simulations

6.5.1 Aerosol Optical Depth

Figure 27 reports the AOD (in the visible range) simulated for the SOP-1a period and for the COSMO-M, RegCM, CNRM-RCSM and CHIMERE models. Except the CTM-CHIMERE model which includes all the secondary species (SOA and inorganic), the

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5 already mentioned, one reason of lowest time-correlation for these models is related to the fact that they are only forced at the boundaries and the synoptic conditions inside the domain can derive during the simulation. This effect is limited for CNRM-RCSM that used the spectral nudging technique and for CHIMERE forced by WRF meteorological field (Menut et al., 2015). For each models, biases are shown to be low, both positive (for CNRM-RCSM and CHIMERE) and negative (for COSMO-M and RegCM).

10 For the Ersa station, less influenced by long-range transport of mineral dust during this period, temporal correlations are lowest and found to be moderate (0.40) for CHIMERE and COSMO-M and low for RegCM and CNRM-RCSM. In terms of bias, values are positive and low (0.02 to 0.04) for all models, except for COSMO-M (−0.07) that does not include anthropogenic aerosols nor sea-salt in the present simulation (Table 8). For each model, calculated standard deviations are in the same range of magnitude but slightly higher than observations, especially for RegCM (bias of 0.08) that simulated a large AOD for 19–20 June period. By comparison with the values
15 obtained in Lampedusa, these low correlations at Ersa reveal the limitations of these models in terms of horizontal resolution with respect to the representativeness of the site. Lampedusa being isolated in the middle of the Mediterranean and under the main pathways of African mineral dust, AOD is mostly related to long-ranged transport. On the other hand, the site of Ersa in Corsica may be under several types of aerosols contributions (anthropogenic, biogenic, vegetation fires) more intense and more spatially
20 variables than in Lampedusa. Ersa being closer to large industrial areas, the models with a horizontal resolution of tens of kilometers are probably not highly enough resolved to catch small scales aerosols plumes from the continent.

6.5.2 Regional SW 3-D direct radiative forcing

25 The SW (clear-sky) DRF, averaged for the SOP-1a period, has been estimated from the RegCM and CNRM-RCSM models, both at the surface and TOA, as shown in the Fig. 29. For this discussion, we only consider these two models as they estimate the clear-sky SW DRF by taking into account natural and anthropogenic aerosols, con-

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no. 262 254) and ACTRIS-2 (grant agreement no. 654 109). Measurements at Lampedusa by ENEA were partly supported by the Italian Ministry for University and Research through the NextData and Ritmare Projects. This study, especially the balloon campaign and part of the aircraft operations has also been supported by the French space agency (CNES). The technical staff of SAFIRE, INSU Technical Division and the CNES Balloon sub-directorate (with special mention to Aurélien Bourdon and Gilles Dupouy) are warmly acknowledged for their contribution to the success of the experimental work. Contributions by Didier Bruneau (Latmos), Silvia Becagli (Univ. of Florence, Italy), Marco Cacciani (Univ. of Rome, Italy), Julian Groebner and Natalia Kouremeti (Physikalisch-Meteorologisches Observatorium Davos, World Radiation Center, Switzerland), and José Antonio Martínez Lozano (University of Valencia, Spain) are gratefully acknowledged. Granada station was partially supported by the Andalusian Regional Government through project P12-RNM-2409 and by the Spanish Ministry of Science and Technology through project CGL2013-45410-R. Sahar Hassazadeh, Constantino Munoz Porcar, and Santi Bertolin and Diego Lange are also acknowledged for their kind assistance in operating the Menorca surface station, as well as François Gheusi, Brice Barret, Flore Tocquer, and Yves Meyerfeld for their contribution to the balloon campaign preparation and/or deployment. Claude Basdevant, Alexis Doerenbecher, and Fabien Bernard are acknowledged for their help and very useful tools in support of our drifting balloon experiment.

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Table 1. List of the Instrumentations deployed over the two super-sites (Ersa and Lampedusa) during the SOP-1a experiment for the characterization of physical, chemical and optical properties of aerosols, vertical profiles, columnar-averaged properties and radiation measurements. Meteorological parameters and gases concentrations are not included in this Table.

	Ersa		Lampedusa	
	Instruments	Frequency	Instruments	Frequency
Number concentration	1 CPC (0.01–3 μm)	continuous (1')	1W-CPC (0.01–3 μm)	continuous (2')
CCN concentration	1 CCN counter	continuous	1 CCN counter	continuous
Mass concentration	1 PM _{2.5}	continuous	1 PM ₄₀ (TEOM)	continuous
	1 PM ₁₀	continuous		
Number size distribution	1 OPC (0.3–5 μm)	continuous	2 GRIMM (0.25–32 μm)	continuous
	1 APS (TSI)	continuous	1 APS (TSI) (0.5–20 μm)	continuous
	1 SMPS (3–300 nm)	continuous	2 (dry/ambient) SMPS	continuous
Mass size distribution	2 Impactor DEKATI (13 stages)	48 h	2 Impactor DEKATI (13 stages)	48 h
			1 Impactor Nano-MOUDI	24 h
PM ₁ composition	1 PILS	continuous	AMS (Aerodyne)	continuous
			1 PILS	continuous
PM ₁₀ composition			1 FAI Hydra Sampler	12 h
Mass BC concentration	1 (7λ) aethalometer	continuous	1 PSAP continuous	(1 h)
			1 MAAP	continuous
Vertical Profiles	1 (1λ 350 nm) Leosphere	continuous	1 (1λ) Leosphere ALS 300	continuous (20')
			2 (3-λ) ENEA/Univ. of Rome lidar	continuous (1')
			microwave radiometer (<i>p</i> , <i>T</i> , RH)	continuous (15')
			radiosondes	on event
Scattering coefficient	1 (3λ) TSI nephelometer (450–550-700 nm)	continuous (1')	1 (3λ) TSI nephelometer (450–550-700 nm)	continuous (1')
Absorbing coefficient	1 (7λ) aethalometer (370–420-490–520-660–880-950 nm)	continuous	1 (7λ) aethalometer (370–420-490–520-660–880-95 nm)	continuous
Extinction coefficient	1 (1λ) (860 nm) PAX	continuous (1')		
Column optical properties	1 (9λ) AERONET/PHOTONS	continuous (15' for AOD)	1 (9-λ) AERONET/PHOTONS	continuous (15' for AOD)
			2 (12-λ) MFRSRs	continuous (15 s)
Mineral Aerosol Deposition	1 CARAGA	continuous (7 days)	1 CARAGA	continuous (7 days)
Downward shortwave irradiance	1 pyranometer	continuous (30 s)	1 (CMP 21) pyranometer	continuous (30 s)
Downward longwave irradiance	1 pyrgeometer	continuous (30 s)	1 (CGR4) pyrgeometer	continuous (30 s)
Downward window (8–14 μm) irradiance			1 modified CG3 pyrgeometer	continuous (60 s)
Direct Solar radiance			1 CHP1 Pyrheliometer	continuous (30 s)
Direct spectral solar radiation			1 PMOD Precision SpectroRad.	continuous (30 s)
Spectral downward global solar irradiance			1 HyperOCR spectrometer	continuous (30 s)
Spectral downward diffuse solar irradiance			1 HyperOCR spectrometer	continuous (30 s)
Spectral direct solar irradiance			1 spectroradiometer	continuous (60 s)
Downward spectral actinic flux			1 Diode array spectrometer	continuous (60 s)

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Table 3. In-situ instrumentation deployed onboard the ATR-42 during the SOP-1a experiment.

Parameter measured	Instrument	Abreviation	Location in the aircraft	Wavelength (nm)	Nominal size range (μm)
Size distribution	Forward Scattering Spectrometer Probe, Model 300, Particle Measuring Systems	FSSP-300	wing-mounted	632.8	0.28–20
	Ultra High Sensitivity Aerosol Spectrometer, Droplet Measurement Technologies	UHSAS	wing-mounted	1054	0.04–1
	Sky-Optical Particle Counter, Model 1.129, Grimm Technik	GRIMM1	AVIRAD inlet	655	0.25–32
	Optical Particle Counter, Model 1.109, Grimm Technik aerosol inlet	GRIMM2	Communautory	655	0.25–32
	Optical Particle Counter, Model 1.109, Grimm Technik	GRIMM3	Communautory aerosol inlet	655	0.25–32
	Scanning mobility particle sizer, custom-built (Villani et al., 2007)	SMPS	Communautory aerosol inlet	NA	0.03–0.4
Integrated number concentration	Condensation Particle Counters, Model 3075, TSI	CPC	AVIRAD inlet	NA	> 0.005
Scattering coefficient	3 λ Integrated Nephelometer, Model 3563, TSI	Nephelometer	AVIRAD inlet	450, 550, 700	NA
Absorption coefficient	3 λ Particle Soot Absorption Photometer, Radiance Research	PSAP	Communautory aerosol inlet	467, 530, 660	NA
Extinction coefficient	Cavity Attenuated Phase Shift, Aerodyne Research Inc.	CAPS	Communautory Aerosol inlet	530	NA
	Photomètre Léger Aéroporté pour la Surveillance des Masses d'Air	PLASMA	roof-mounted	340–2250	NA
Chemical composition	Filter sampling	NA	AVIRAD inlet	NA	NA
	Single particle soot photometer, Droplet Measurement Technologies	SP2	Communautory aerosol inlet	1064	0.08–0.5

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Table 4. Characteristics of the 15 sounding balloons flights from Sant Lluís, Minorca Island, during the ChArMEx SOP1a/ADRI-MED campaign.

No.	Date (2013)	Start time (UTC)	Ceiling altitude (m)	Latitude at ceiling	Longitude at ceiling	Sensors
BLD1	12 Jun	21:13	21 178	39.5156° N	04.3010° E	T, U
BLD2	15 Jun	21:40	32 119	39.9903° N	04.1801° E	T, U, LOAC, O ₃
BLD3	16 Jun	10:29	31 880	40.0527° N	04.1524° E	T, U, LOAC, O ₃
BLD4	16 Jun	21:13	33 390	40.0999° N	04.0118° E	T, U, LOAC, O ₃
BLD5	17 Jun	10:01	32 744	40.2109° N	03.9672° E	T, U, LOAC, O ₃
BLD6	17 Jun	18:25	33 411	40.2502° N	03.9402° E	T, U, LOAC, O ₃
BLD7	18 Jun	16:34	35 635	40.5832° N	04.0515° E	T, U, LOAC
BLD8	18 Jun	21:17	21 507	40.6372° N	04.4889° E	T, U, LOAC, O ₃
BLD9	19 Jun	10:12	30 902	40.6794° N	04.3691° E	T, U, LOAC, O ₃
BLD10	19 Jun	13:48	36 129	40.6553° N	04.1970° E	T, U, LOAC
BLD11	27 Jun	09:43	35 832	39.7546° N	04.4746° E	T, U, LOAC
BLD12	28 Jun	05:36	36 293	39.4505° N	04.1709° E	T, U, LOAC
BLD13	29/30 Jun	23:31	36 310	39.6168° N	03.7383° E	T, U, LOAC
BLD14	30 Jun	14:03	36 319	39.8937° N	03.9568° E	T, U, LOAC
BLD15	02 Jul	10:27	32 833	39.9942° N	04.2996° E	T, U, LOAC, O ₃

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Table 5. Characteristics of the 14 BPCL drifting balloon flights.

Date and time of launch (UT)	Balloon Nbr and type of sensor	Last data time (UT)	Last data location	Trajectory length (km)	Flight duration (h)	Approximate float altitude (m)
16 Jun, 09:46	B74, LOAC	16 Jun, 21:51	43.0265° N 05.2285° E	368	11:57	2100
16 Jun, 09:53	B53, O3	17 Jun, 00:26	40.6541° N 06.2398° E	203	14:28	3000–3050
16 Jun, 09:58	B70, LOA	16 Jun, 23:01	40.1825° N 06.1293° E	174	13:17	3050–3150
17 Jun, 09:27	B54, O3	17 Jun, 16:49	43.1433° N 03.5293° E	371	07:22	1850–2000
17 Jun, 09:29	B75, LOAC	17 Jun, 16:51	43.0868° N 03.6866° E	365	07:23	1950–2050
17 Jun, 11:07	B72, LOAC	17 Jun, 19:07	43.2333° N 04.7403° E	382	08:03	2750
19 Jun, 10:34	B77, LOAC	19 Jun, 17:59	43.1576° N 04.7562° E	387	07:37	2550
19 Jun, 10:35	B71, LOAC	19 Jun, 15:03	43.0560° N 05.1336° E	369	04:39	3250–3350
27 Jun, 10:00	B80, LOAC	28 Jun, 12:07	37.9165° N 12.1605° E	759	26:19	2950–3050
28 Jun, 05:20	B73, LOAC	28 Jun, 17:24	37.4095° N 09.2346° E	523	12:16	2650–2750
02 Jul, 13:03	B76, LOAC	03 Jul, 09:38	37.8897° N 12.1312° E	731	20:39	3150–3250
02 Jul, 13:11	B57, O3	03 Jul, 22:43	35.0900° N 14.1140° E	1053	33:44	3100–3200
02 Jul, 17:59	B55, O3	04 Jul, 02:20	37.3545° N 12.21980E	762	32:32	2400–2450
02 Jul, 17:50	B78, LOAC	04 Jul, 02:13	37.5639° N 12.1507° E	755	32:25	2350–2450

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Table 6. Main aerosol volume size distribution characteristics: r_{vf} (μm), σ_f , r_{vc} (μm), σ_c , C_{vf} , C_{vc} , for the four different AERONET/PHOTONS stations: Ersa, Lampedusa, Cagliari and Cap d'En Font. C_{vi} denotes the particle volume concentration, r_{vi} is the median radius, and σ_i is the standard deviation. Each average value in the table is accompanied by its standard deviation (this is not an accuracy of the retrieval).

	Ersa	Lampedusa	Cagliari	Cap d'En Font
Number of observations	33	18	20	20
r_{vf} (μm)	0.16 ± 0.02	0.14 ± 0.01	0.15 ± 0.03	0.17 ± 0.03
σ_f	0.43 ± 0.03	0.50 ± 0.06	0.46 ± 0.04	0.45 ± 0.04
r_{vc} (μm)	2.49 ± 0.43	2.36 ± 0.48	2.52 ± 0.28	2.48 ± 0.30
σ_c	0.69 ± 0.03	0.68 ± 0.05	0.71 ± 0.04	0.71 ± 0.04
C_{vf} ($\mu\text{m}^3 \mu\text{m}^{-2}$)	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01	0.02 ± 0.01
C_{vc} ($\mu\text{m}^3 \mu\text{m}^{-2}$)	0.03 ± 0.01	0.08 ± 0.05	0.05 ± 0.03	0.04 ± 0.03

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Table 7. Averaged mean Single Scattering Albedo (SSA) estimated from the AERONET/PHOTONS network at four stations during the SOP-1a, at 440, 670, 870 and 1020 nm.

	Ersa	Lampedusa	Cagliari	Cap d'En Font
Number of observations	33	18	20	20
440/970/870/1020	0.977/0.970/0.965/0.964	0.916/0.922/0.925/0.929	0.927/0.915/0.914/0.916	0.957/0.951/0.948/0.948

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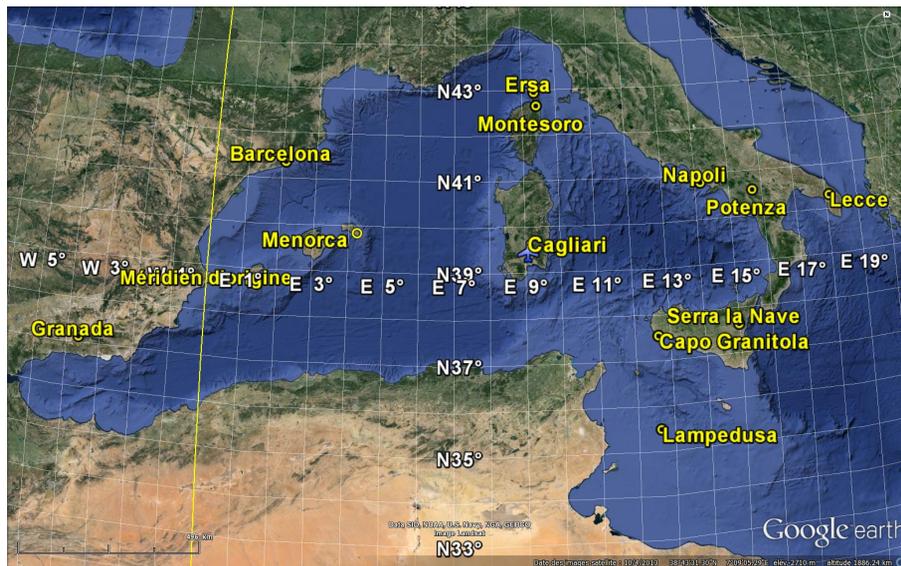
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Figure 2. The regional experimental set-up deployed in the western and central Mediterranean during the campaign ChArMEx SOP-1a. The two aircraft were based at Cagliari.

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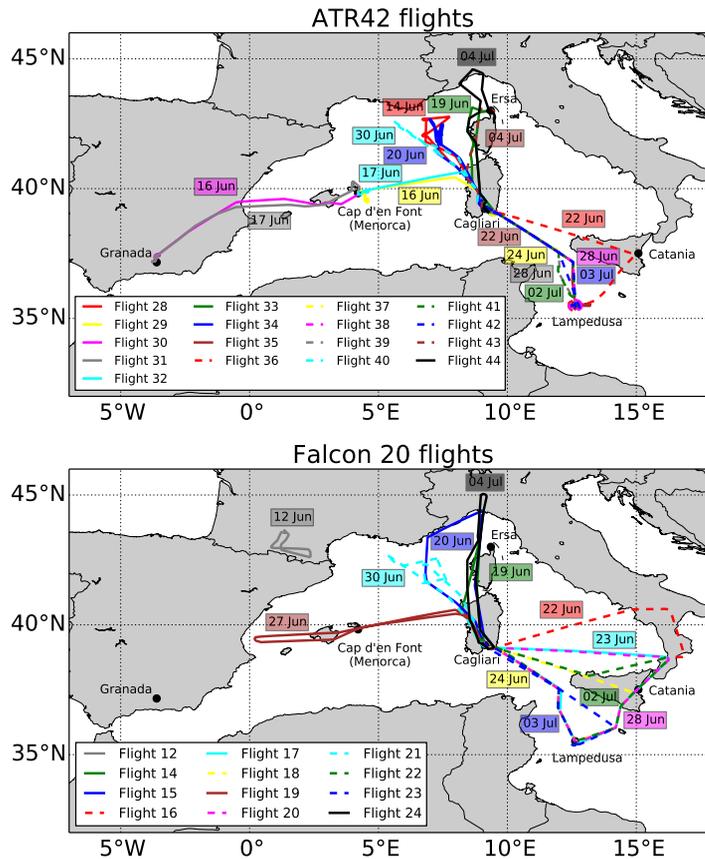


Figure 3. Overview of the different ATR-42 and F-20 flights trajectories performed during the SOP-1a experiment.

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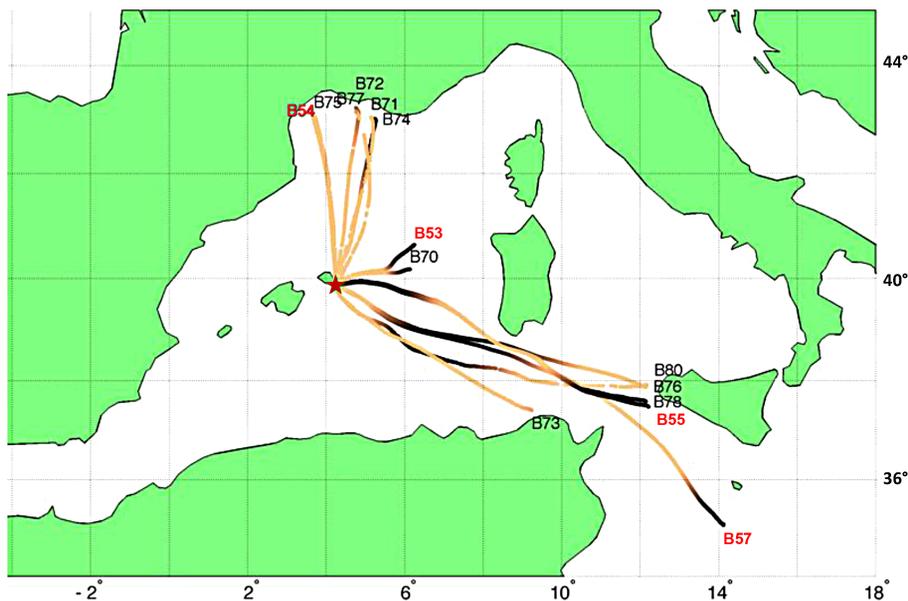


Figure 4. Trajectories of the 14 BPCL drifting balloons launched from Minorca Island during the campaign. Dark portion along trajectories correspond to night-time conditions. The four red labels from B54 to B57 indicate balloons with an ozone sonde and the 10 others carried a LOAC instrument.

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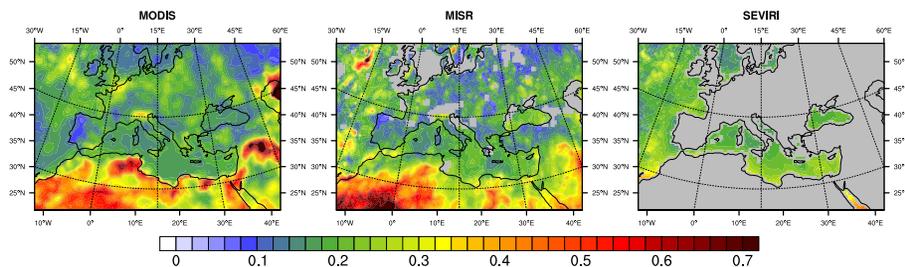


Figure 5. Total AOD (500 nm) obtained from the MODIS, MISR and SEVIRI (sea only) sensors for the June–July 2013 period.

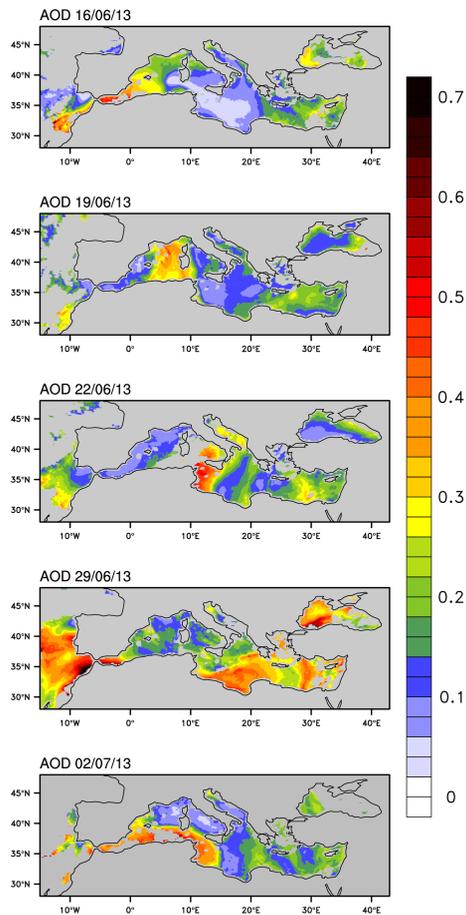


Figure 6. AOD MSG/SEVIRI observations for five different days during the SOP-1a experiment (16, 19, 22, 29 June and 3 July).

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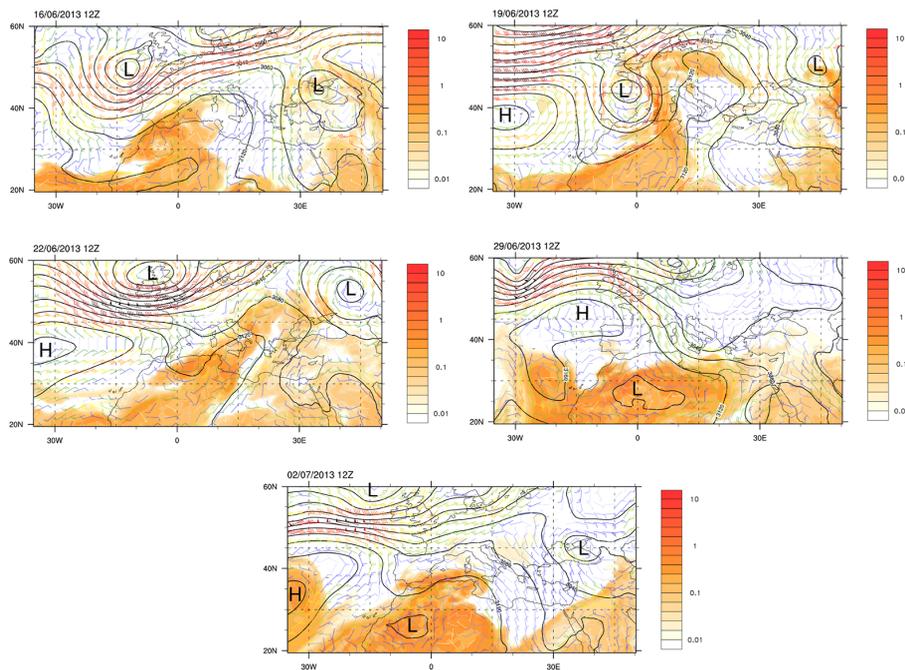


Figure 7. Geopotential at 700 hPa, mass dust concentration (in mgm^{-3}), and wind intensity at 700 hPa for the 6, 19, 22, 29 June and 2 July at 12:00 UTC, simulated from the ALADIN model.

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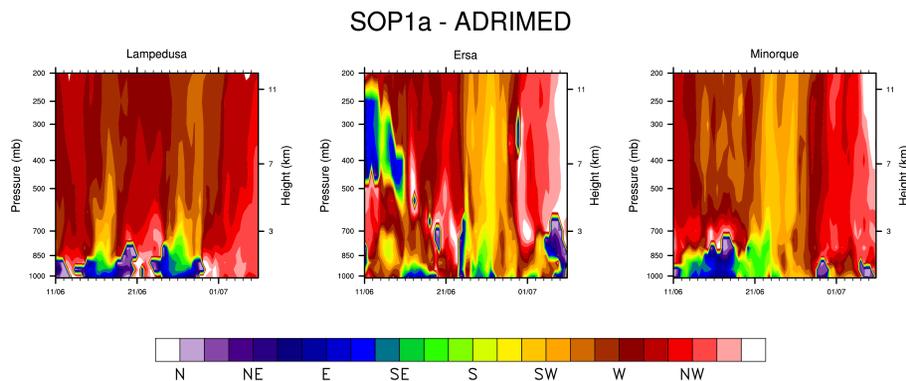


Figure 8. Wind profiles between 1000 and 200 hPa during the SOP-1a experiment for three different sites (Ersa, Lampedusa and Minorca) simulated from the ALADIN model.

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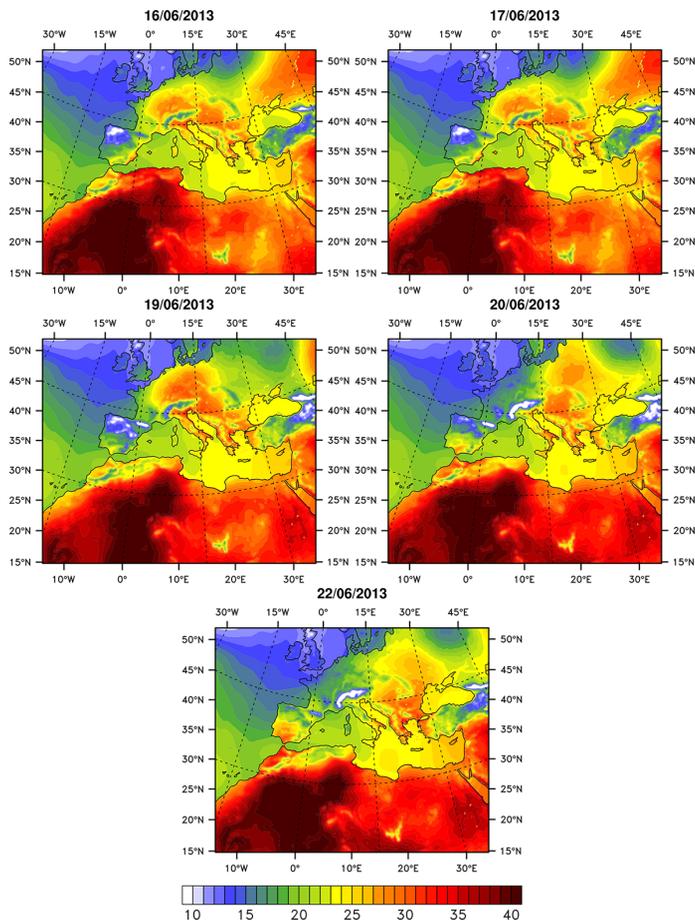


Figure 9. Surface Temperature (at 12:00 UTC) obtained from NCEP re-analysis for the 16, 17, 19, 20 and 22 June.

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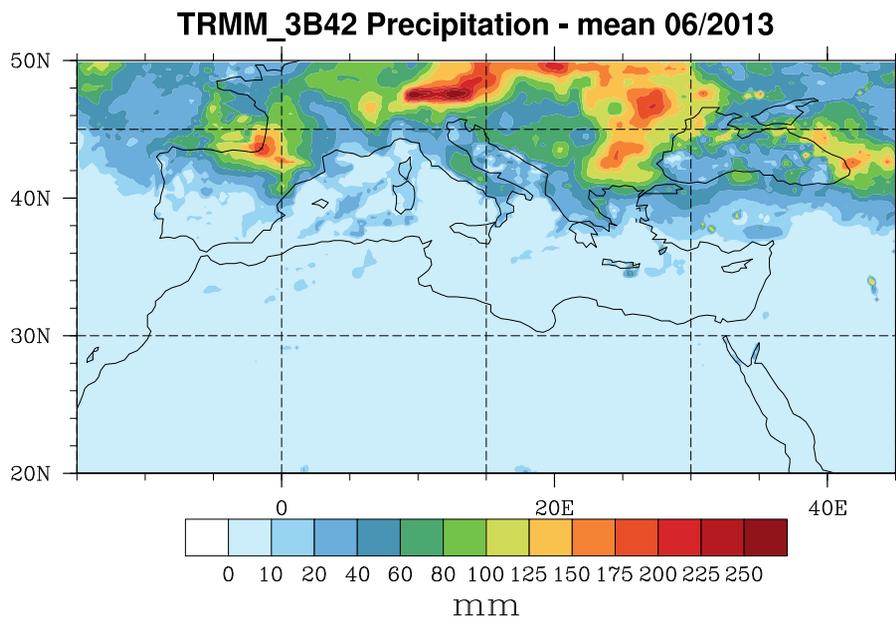
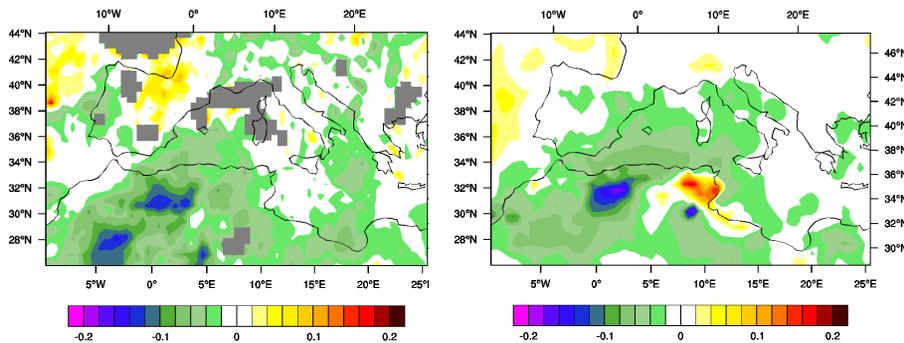


Figure 11. Same figure as 10 but for the Tropical Rainfall Measuring Mission (TRMM) precipitation observations.

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MISR observations

MODIS observations

Figure 12. AOD anomaly for summer 2013 estimated from the MODIS and MISR sensor data.

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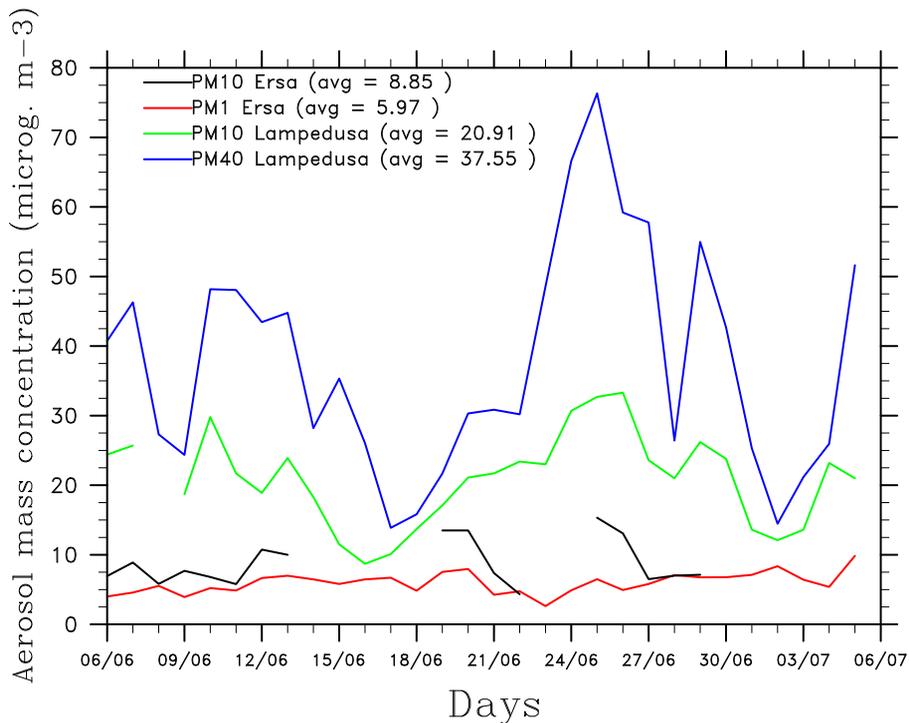


Figure 13. Time-series of daily PM mass concentrations estimated at the Lampedusa (PM₄₀ and PM₁₀) and Ersá (PM₁ and PM₁₀) super-stations.

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SOP-1a AERONET/PHOTONS Volume size distribution

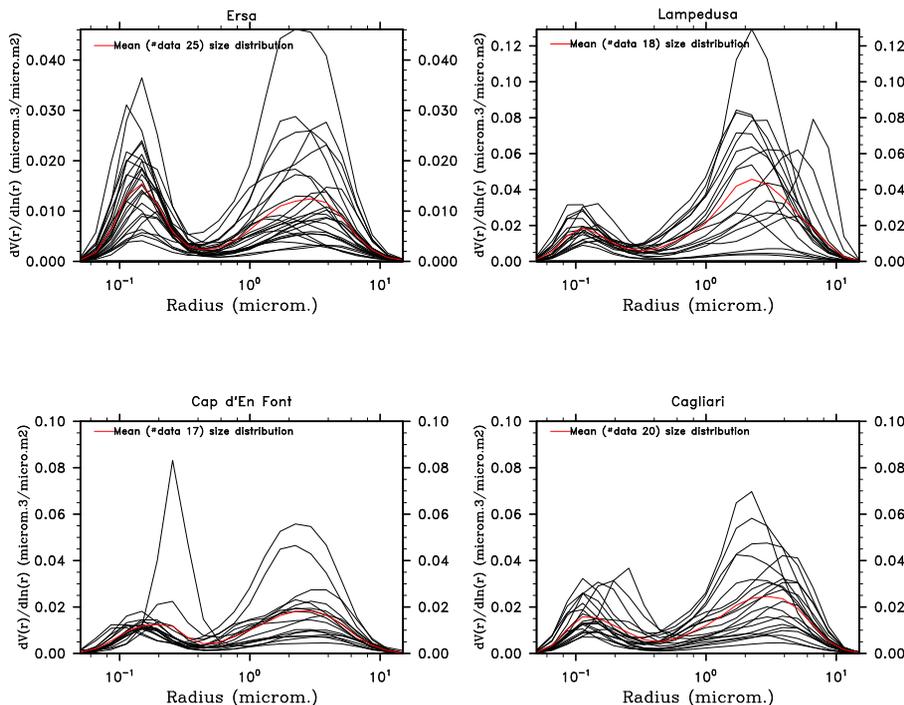


Figure 14. AERONET/PHOTONS volume size distribution derived at four different stations: Ersa, Lampedusa, Cagliari and Cap d'En Font (the red curve represents the mean of observations). The characteristics of the volume size distribution are provided in Table 6.

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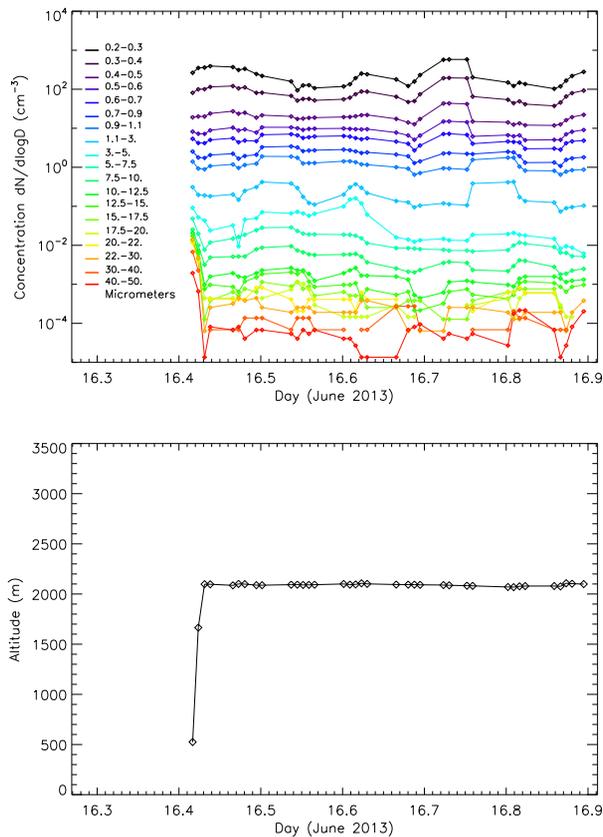


Figure 15. Particle size distribution measured with a LOAC during the ~ 12 h flight of the BPCL balloon B74 drifting from Minorca Island towards Marseille (see trajectory in Fig. 4). The first and last 20 min correspond to the ascending and descending phases of the quasi-Lagrangian flight which occurred at a constant altitude of 2091 ± 10 m.

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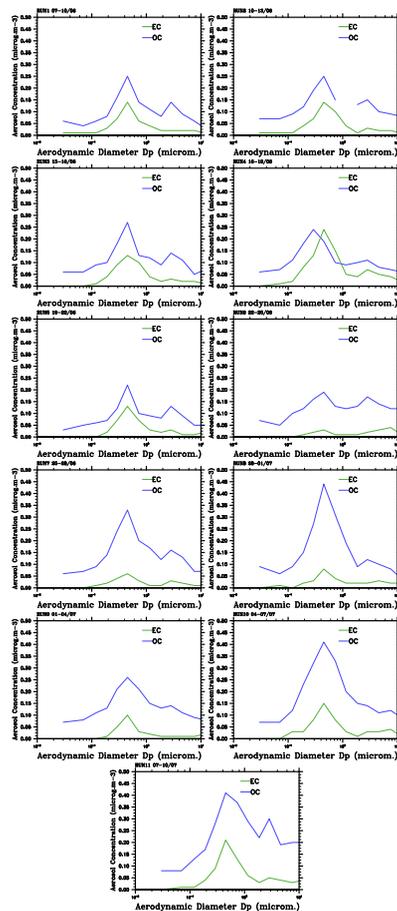


Figure 16. EC and OC (48 h-mean) aerosol mass size distributions obtained at Ersa from the impactor DEKATI instrument for all the SOP-1a period.

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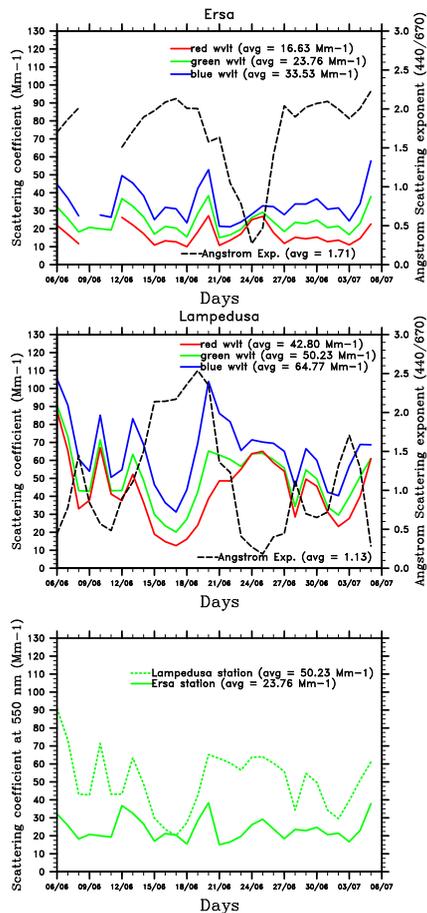


Figure 17. Time-series of daily scattering coefficient (in Mm^{-1}) estimated in the Ersa and Lampedusa stations. The daily Angström Exponent (AE), calculated between 440 and 670 nm, is also reported.

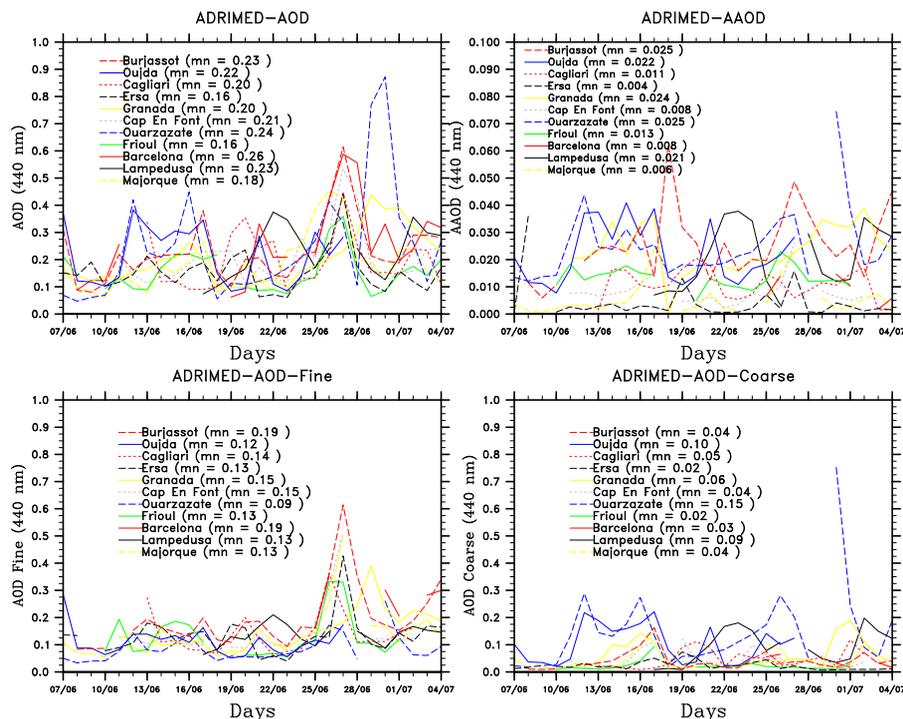


Figure 18. AERONET/PHOTONS observations of the total extinction AOD, AOD Fine (AODf), AOD Coarse (AODc) and Absorbing AOD (AAOD), at 440 nm obtained for the whole SOP-1a period.

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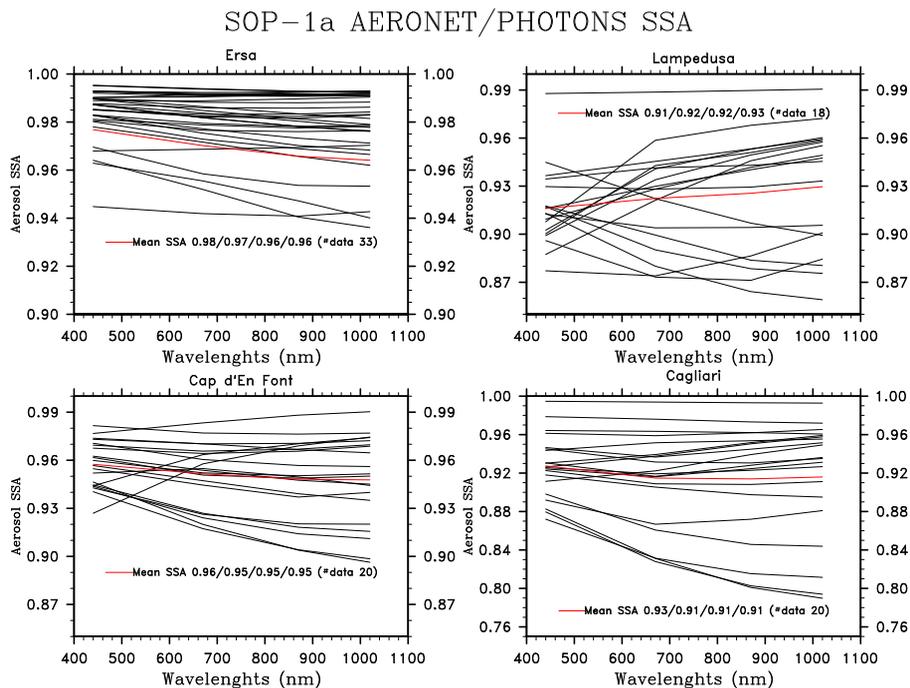



Figure 19. AERONET/PHOTONS observations of the total single scattering albedo (SSA) at 440, 670, 880 and 1020 nm obtained for the whole SOP-1a period (the red curve represents the mean of observations).

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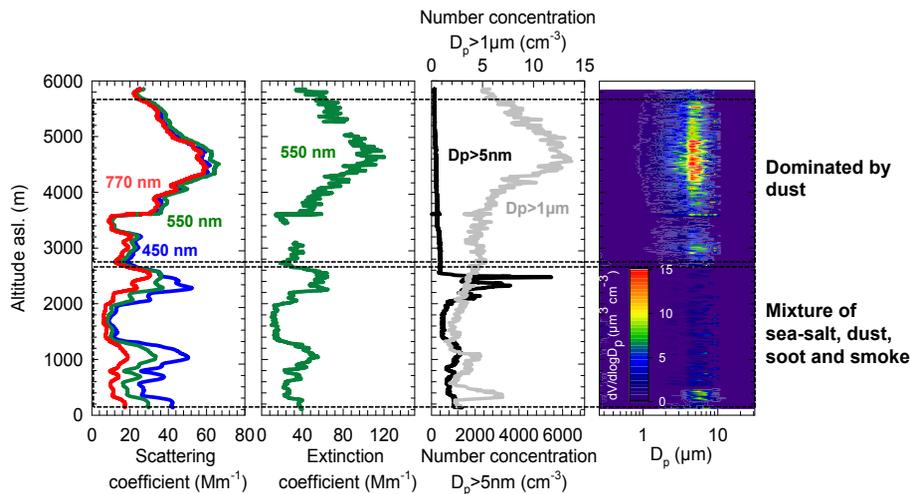


Figure 20. Optical (scattering and extinction coefficients) and physical (number concentration and volume size distribution) aerosol properties estimated along the vertical onboard the ATR-42 aircraft for the flights 35–36 on 22 June over the Lampedusa station.

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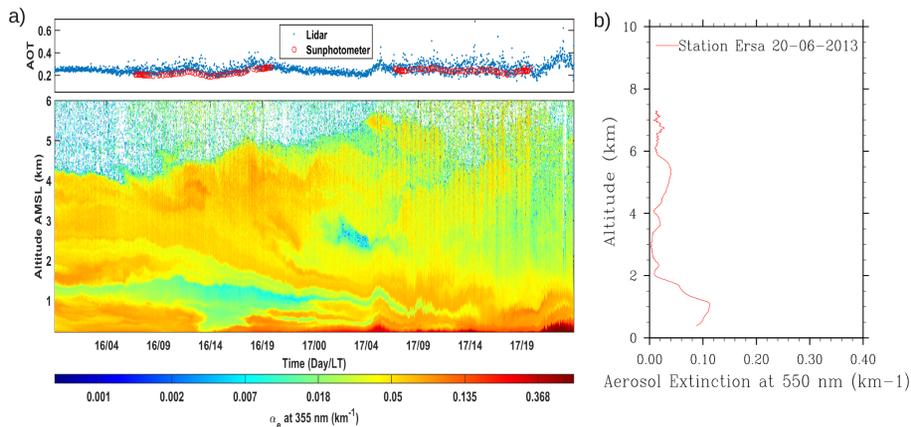


Figure 21. Minorca and Ersa lidar observations obtained during the dust plume of 16 to 17 June transported over the western Mediterranean basin.

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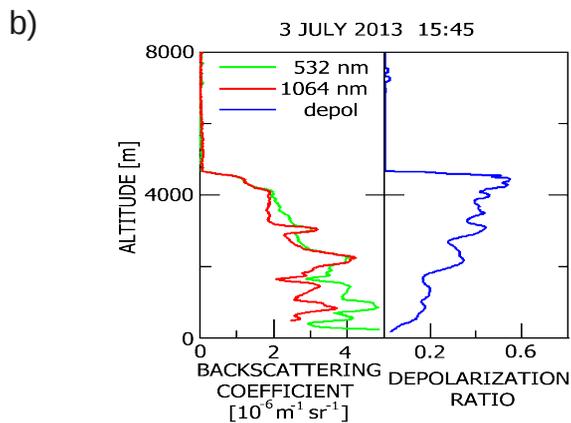
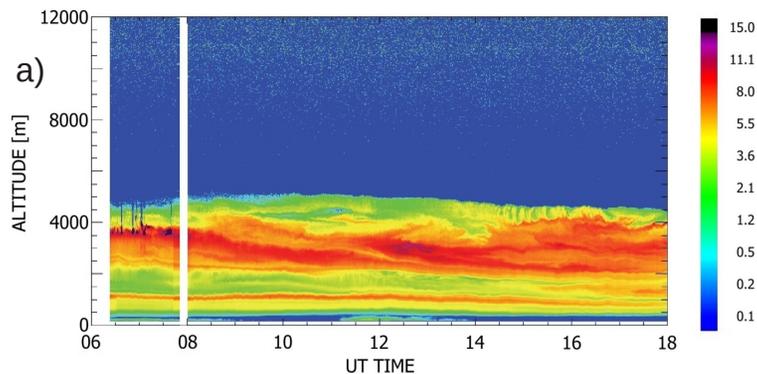


Figure 22. (a) Time evolution of the vertical profile of the aerosol backscattering coefficient at 1064 nm at Lampedusa on 3 July 2013. The color scale is in units of $10^{-7} \text{ m}^{-1} \text{ sr}^{-1}$. (b) Vertical profile of aerosol backscattering coefficient at two wavelengths and of aerosol depolarization ratio at 355 nm measured at Lampedusa on 3 July 2013 at 15:45 UT.

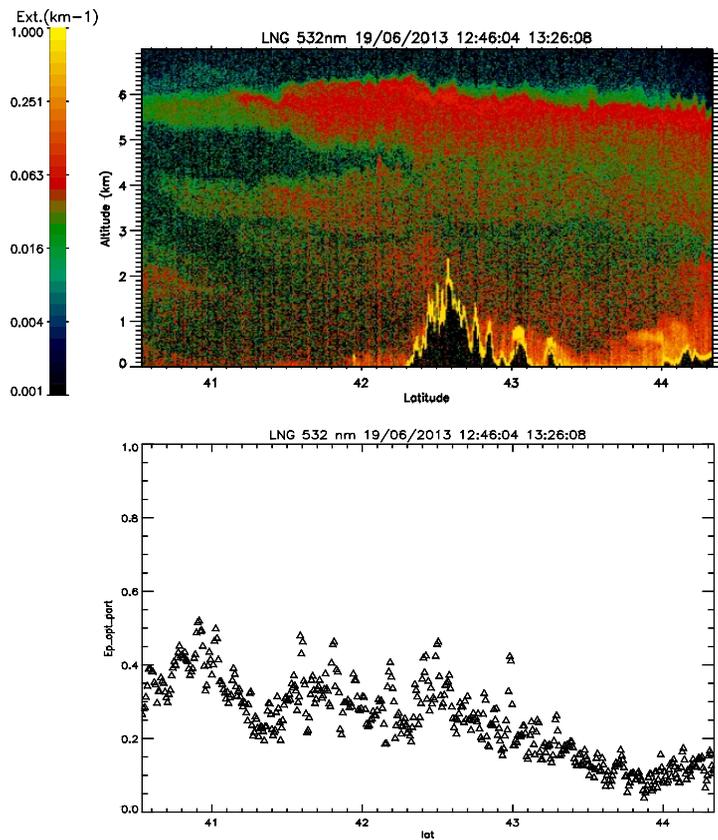


Figure 23. Observations of aerosol extinction coefficient (top, in km^{-1} at 532 nm) and aerosol optical depth (bottom) obtained from the lidar LNG system onboard the F-20 aircraft during the 19 June that corresponds to the flight (12:46 to 13:26) from Cagliari to the Gulf of Genoa.

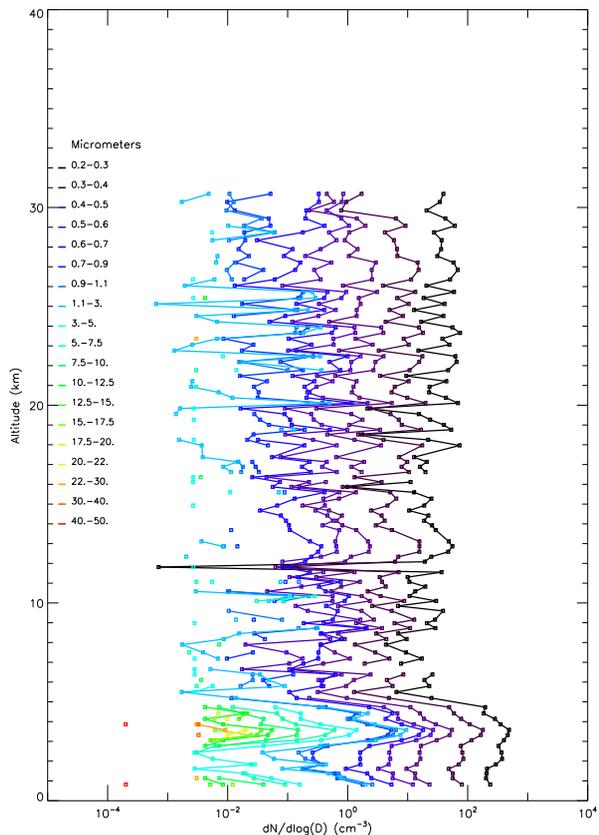


Figure 24. Particle concentrations as a function of size and altitude in the troposphere and lower stratosphere from the LOAC flight under the meteorological balloon BLD9 launched from Minorca at the end of a dust event on 19 June 2013, 10:12 UT (Table 4; see the daytime averaged aerosol optical depth over the sea in Fig. 6).

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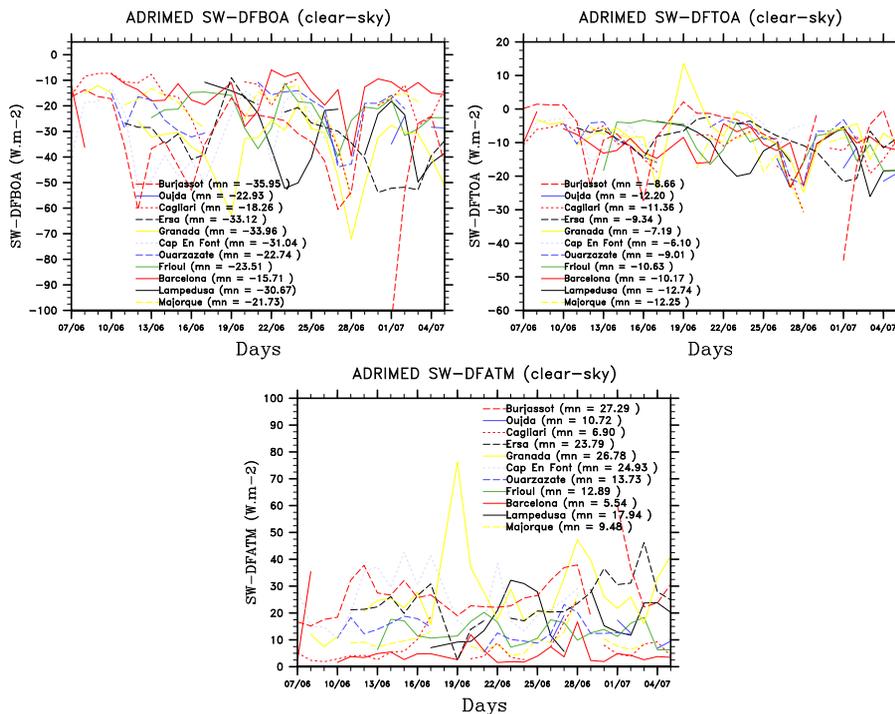


Figure 25. 1-D (clear-sky) instantaneous (shortwave only) DRF calculations (in W m^{-2}) based on AERONET/PHOTONS dataset for the different stations listed in Table 2 (BOA, TOA and ATM refer to bottom of the atmosphere, top of atmosphere and atmospheric forcings).

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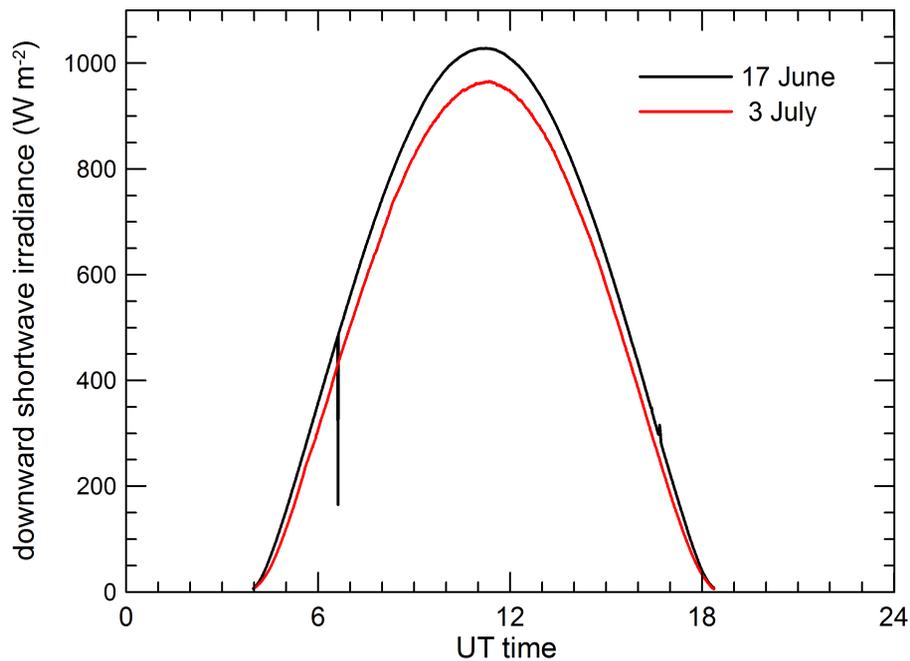


Figure 26. Time evolution of the downward solar irradiance observed at Lampedusa on 17 June and on 3 July 2013.

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Aerosol optical Depth (visible band)

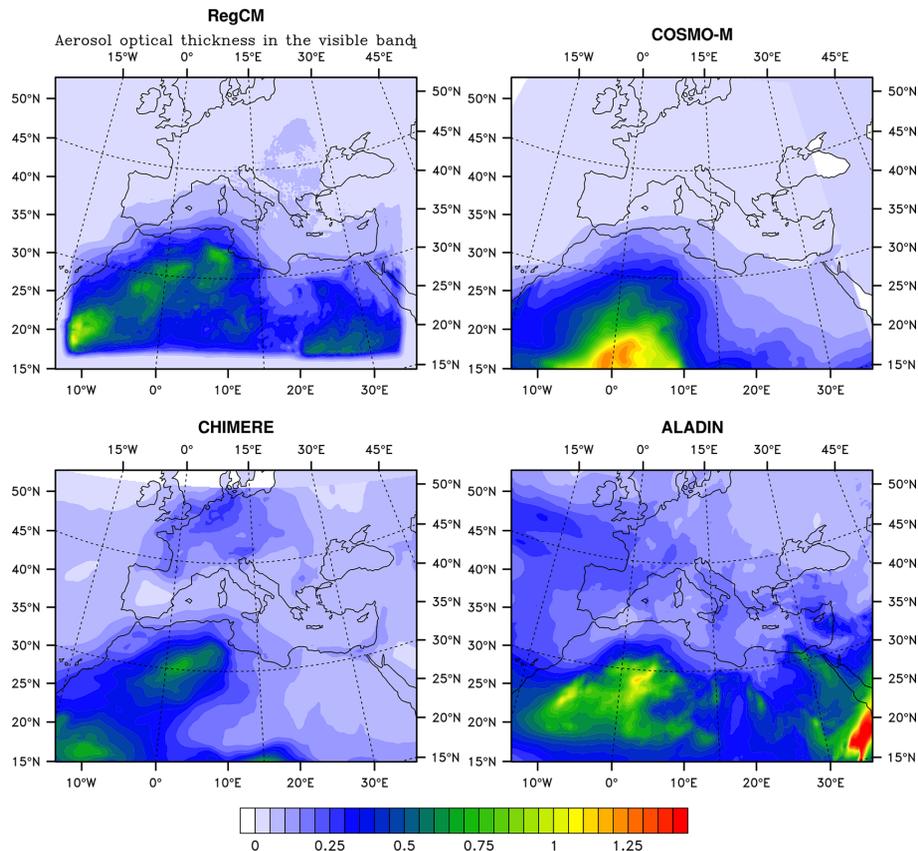


Figure 27. AOD averaged for the 15 to 25 June 2013 period from the meso-scale COSMO-MUSCAT (a), CTM-CHIMERE (b) models and the two regional climate models; CNRM-RCSM (c) and RegCM (d). Details about the model configurations are provided in Table 8.

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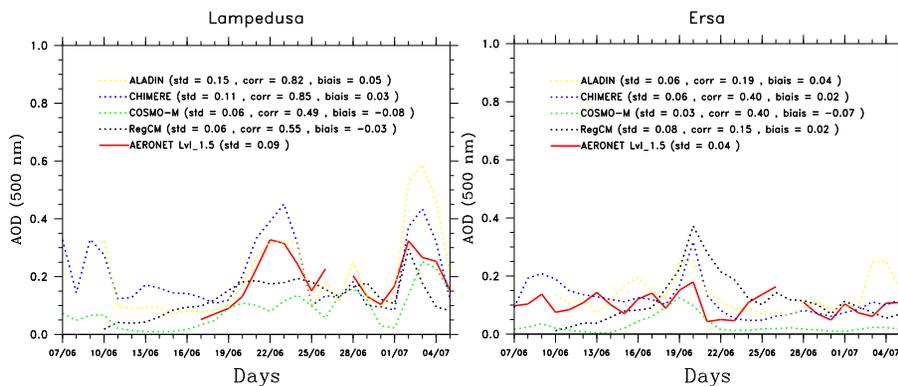


Figure 28. Times-series of AOD comparisons between AERONET/PHOTONS observations and COSMO-MUSCAT, CHIMERE, CNRM-RCSM and RegCM model outputs over the two stations of Ersa and Lampedusa.

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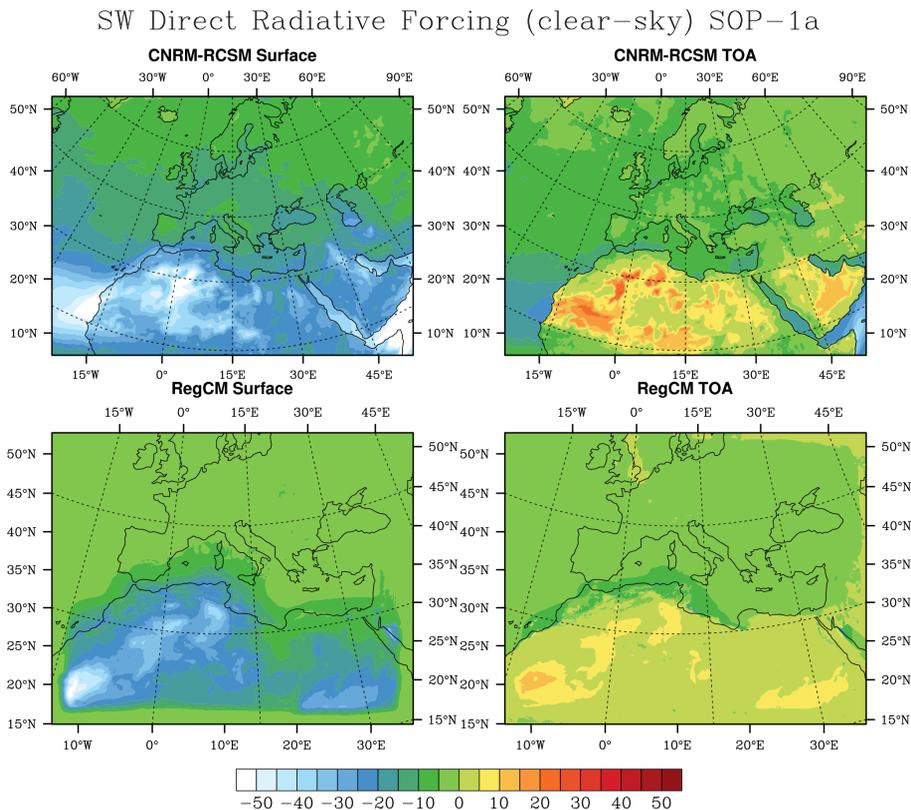


Figure 29. Averaged surface and TOA SW DRF simulated in clear-sky conditions and over the SOP-1a period by the CNRM-RCSM and RegCM models.