Aerosol optical properties derived from POLDER-3/PARASOL (2005-2013) over the western Mediterranean Sea: I. Quality assessment with AERONET and in situ airborne observations

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Abstract

The western Mediterranean atmosphere is impacted by a variety of aerosol sources, producing a complex and variable mixture of natural and anthropogenic particles, with different chemical and physical properties. Satellite sensors provide a useful global coverage of aerosol parameters but through indirect measurements that request careful validation. Here we present the results of a long-term regional scale analysis of the full dataset (March 2005 and October 2013) of POLDER-3/PARASOL ocean operational retrievals of the total, fine and coarse aerosol optical depth (AOD, AOD_F and AOD_C), Angstrom exponent (AE), and the spherical/non-spherical partition of coarse-mode AOD (AOD_{CS} and AOD_{CNS}), respectively. The evaluation is performed using data from seventeen coastal and insular ground-based AERONET sites on one side, and airborne vertical profiles of aerosol extinction and number size distribution obtained by the SAFIRE ATR-42 aircraft operated in the area during summer 2012 and 2013 on the other side. This study provides the first regional evaluation of uncertainties of the POLDER-3 products, and highlights their quality. The POLDER-3 Ångström exponent, representing AOD spectral dependence in link with the aerosol particle size distribution, is biased towards small values. This bias, however, does not prevent using AE for classifying the regional aerosol laden air masses. AOD_F corresponds to particles smaller than 0.6-0.8 µm in diameter and appears suitable to monitor the aerosol submicron fraction from space. We also provide an original validation of POLDER-3 AOD_C and its spherical/non-spherical partition, which shows agreement within 25% with AERONET shape retrievals when the aerosol coarse fraction dominates.

1. Introduction

Aerosols include a large variety of particles (mineral dust, sea salt, soot carbon and organic species, sulphates, nitrates…) emitted by natural and anthropic sources and different mechanisms (combustion, wind erosion, gas-to-particle conversion, etc.). Aerosols have a short lifetime in the troposphere (Boucher, 2015) but they are key to many atmospheric processes, as the redistribution of solar and thermal radiation by scattering and absorption, cloud formation and precipitation, and air quality degradation, which, in turn are relevant in shaping the Earth climate and liveability (Pope et al., 2002; Akimoto, 2003; Pope and Dockery, 2006; Monks et al., 2009; Boucher et al., 2013).

Despite its importance, the global aerosol radiative effect is far from being certain, as both aerosol spatial distribution and optical properties are affected by large unknowns (Boucher et al., 2013; Myhre et al., 2013). Furthermore, the apportionment of aerosols to anthropogenic and natural sources is critical to evaluate the perturbative forcing of human activities on the Earth radiative budget and ultimately climate (Myhre et al., 2013; Shindell et al., 2013; Kim et al., 2014; Pan et al., 2015). In this general context, the Mediterranean basin is a region of great interest. Submitted to demographic pressure and experiencing bad air quality (Monks et al., 2009; Kovats et al., 2014), the Mediterranean is a high emission and transport region of all kinds of anthropogenic and natural aerosols (e.g. Moulin et al., 1998; Lelieveld et al., 2002; Pace et al., 2005 and 2006; Querol et al., 2009; Pey et al., 2013; Becagli et al., 2017), as well as one of the most vulnerable areas to climate change (Giorgi, 2006), with severe future warming leading to a reduction in precipitation and soil moisture, and henceforth a significant water stress towards the end of the century (Giorgi and Lionello, 2008; García-Ruiz et al., 2011; Christensen et al., 2013) and likely positive feedbacks on the aerosol load.

Through the years, the Mediterranean aerosols have been investigated through a number of dedicated local and regional scale experiments (e.g. Söderman and Dulac, 1998; Formenti et al., 2002; Lelieveld et al., 2002; Zerefos et al., 2002; Dulac and Chazette, 2003; Cros et al., 2004; Putaud et al., 2004, Mallet et al., 2016), surface monitoring stations and networks (e.g. Bergametti et al., 1989; Migon et al., 1993; Mihalopoulos et al., 1997; Meloni et al., 2007; di Sarra et al., 2008; Pérez et al., 2008; Querol et al., 2009; Kalivitis et al., 2011; Mallet et al., 2013; Pappalardo et al., 2014; Lyamani et al., 2015) and satellite observations (e.g. Dulac et al., 1992; Moulin et al., 1998; Barnaba and Gobbi, 2004; Antoine and Nobileau, 2006; Papadimas et al., 2008; Gkikas et al., 2009 and 2016). More recently, the regional-scale Chemistry-Aerosol Mediterranean Experiment (ChArMEx, http://charmex.lsce.ipsl.fr/) within the international Mediterranean Integrated STudies at Regional And Local Scales (MISTRALS, http://www.mistrals-home.org) program has significantly added to the existing body of knowledge by providing new ground-based, airborne and balloon-borne observations over the western part of the basin (Mallet et al., 2016; see also this special issue).

ChArMEx has also provided a new momentum in the analysis of regional ground-based and satellite aerosol observations on long and short periods (e.g. Mallet et al., 2013; Nabat et al., 2013; Lyamani et al., 2015; Gkikas et al., 2016; Granados-Muñoz et al., 2016; Sicard et al., 2016). Satellite data are highly valuable to provide information on the regional and global aerosol spatial and temporal distribution and optical properties which are input to climate models. Most multi-spectral imagery instruments (e.g., MODIS, SEAWIFS, AVHHR, SEVIRI…) retrieve the aerosol optical depth (*AOD*), representing the column-integrated optically-active content of atmospheric aerosols, and also proportional to the net change in the clear sky outgoing radiative flux at the top of the atmosphere (Boucher, 2015). *AOD* is an essential parameter to establish the climatology of the distribution and effects of atmospheric aerosols and it is often used for model evaluation (e.g., Chin et al., 2002; Huneeus et al., 2011; Nabat et al., 2013). With this respect, advanced spaceborne retrievals deriving the AOD as a function of particle size and shape, and possibly of wavelength, are most useful in evaluating the origin and the radiative effect of aerosols of different nature.

In this paper, we present a first comprehensive quality-assessment study of the advanced dataset provided by the operational ocean retrieval algorithm of the third multi-spectral, multi-directional and polarized POLDER-3 (POLarization and Directionality of the Earth’s Reflectances) radiometer on PARASOL (Polarization & Anisotropy of Reflectances for Atmospheric Sciences coupled with Observations from a Lidar) satellite (Herman et al., 2005; Tanré et al., 2011) over the western Mediterranean basin. POLDER-3 operated from March 2005 to October 2013 and provided the total, fine and coarse mode aerosol optical depth (AOD, AOD_F and AOD_C) at the wavelength of 865 nm, the spectral dependence of the AOD between 670 and 865 nm (Angström exponent, *AE*), and the partition of spherical and non-spherical AOD_C (AOD_{CS} and AOD_{CNS}, respectively). This paper extends previous evaluations of AOD and AOD_F (Goloub et al., 1999; Fan et al., 2008; Bréon et al., 2011) with a focus on the western Mediterranean basin, and provides the first estimate of the significance of the coarse mode spherical and non-spherical components (AOD_C, AOD_{CS} and AOD_{CNS}).

This study is based on comparisons with co-localised observations from the sun/sky photometers of coastal and insular stations of the Aerosol Robotic Network (AERONET; Holben et al., 1998), and with the in situ measurements of vertical profiles of aerosol extinction and size distribution which were performed by the French ATR-42 environmental research aircraft of the Service des Avions Français Instrumentés pour la Recherche en Environnement (SAFIRE, www.safire.fr) during two ChArMEx intensive campaigns (Di Biagio et al., 2016, Denjean et al., 2016, Mallet et al., 2016). In particular, the use of the size distribution vertical profiles measured in situ allows us to calculate the aerosol optical depth over different size ranges, and the evaluation of AOD_F and AOD_C.

The analysis presented in this paper is essential to geophysical analyses of observations by POLDER-3 of the spatial and temporal variability of the aerosol load over the western Mediterranean basin. The investigation of temporal trends over the 8-year operating period, will be presented in a follow-up dedicated paper (part II of the present manuscript).

2. Measurements

2.1. POLDER-3/PARASOL

The third radiometer POLDER-3 on PARASOL, operational from March 2005 to October 2013, was part of the A-Train constellation operated on a sun-synchronous orbit at 705 km crossing the Equator at 13:30 (Equator local time) (Tanré et al., 2011). In December 2009, it left the A-Train, and continued the observations at 3.9 km below, and at 9.5 km below in 2011. This changed its hour of passage, which was 16:00 Equator local time at the end of the operational period.

POLDER-3/PARASOL used a 274 x 242-pixels CCD detector array, each pixel covering 5.3 x 6.2 km² at nadir. The size of the POLDER-3 images was 2100 x 1600 km², allowing to achieve a global coverage within two days. The western Mediterranean area could be covered in less than 5 minutes along its north-to-south axis. The spatial resolution of POLDER-derived (Level 2) aerosol parameters is about 18.5 x 18.5 km² (corresponding to 3 x 3 pixels of the Level-1 grid; http://www.icare.univ-lille1.fr/parasol/products).

The instrument measured Earth radiance at 9 wavelengths from 443 to 1020 nm, three of which with polarisation (490, 670, 865 nm), and at up to 16 different angles (±51° along, ±43° across track). Cloud screening according to Bréon and Colzy (1999) was applied to minimize possible cloud contamination of aerosol products.

In this paper, we used the latest algorithm update (collection 3) performed in 2014 of the operational clear-sky ocean retrieval algorithm (Deuzé et al., 1999, 2000; Herman et al., 2005). This latest version includes calibration improvements and uses the total and polarized radiances at 670 and 865 nm. For each clear sky pixel, the algorithm recalculates the observed polarized radiances at several observational angles from a Look-Up Table (LUT) built on aerosol micro-physical models. These are constructed as follows: (i) aerosol are not-absorbing, that is, the imaginary part m_i of their complex refractive index ($m = m_r$ -i m_i) is zero. Only the real part m_r is attributed, and considered as invariant with wavelength between 670 and 865 nm; (ii) the aerosol number size distribution is bimodal and lognormal with a fine mode with effective diameter (D_{eff}) smaller than 1.0 µm and a coarse mode with D_{eff} larger than 1.0 µm. The coarse mode includes a non-spherical fraction based on the spheroidal model from Dubovik et al. (2006). Collection 3 increases the number of modes with respect to the previous versions reported by Herman et al. (2005) and Tanré et al. (2011), and allows spheroidal D_{eff} to take two values (2.96 or 4.92 µm). The summary of LUT parameters are presented in the supplementary material (**Table S1**).

The calculations of the multi-spectral, multi-angle polarized radiances are done using a Mie model for homogeneous spherical particles or the spheroidal optical model developed by Dubovik et al. (2006). A quality flag index (0 indicating the lowest and 1 the highest quality) is attributed to each pixel depending on the quality of radiance simulation.

In this paper, we target the following POLDER-3 oceanic (i.e. over ocean surfaces) aerosol products, in which AODs are at 865 nm:

- The total aerosol optical depth (*AOD*),and the Ångström Exponent (*AE*) representing the spectral dependence of AOD, and calculated as

$$AE = -\frac{\ln(AOD_{865}/AOD_{670})}{\ln(865/670)} \quad (1)$$

- The aerosol optical depth due to the fine particle mode (AOD_F)

all obtained for clear-sky pixels.

In addition, for favourable viewing geometries (scattering angles between 90° and 160°), we have enough information to consider that the coarse mode is a mixing of spherical and non-spherical particles. We assume that the fraction of non-spherical particles (f_{CNS}) of the coarse mode (AOD_c) can be equal to 5 discrete values (0, 0.25, 0.50, 0.75, 1.0). Then, the AOD_{CNS} (respectively AOD_{CS}) is derived from

$$AOD_{CNS} = f_{CNS} \times AOD_C \quad (2\text{-a})$$

$$AOD_{CS} = (1-f_{CNS}) \times AOD_C \quad (2\text{-b})$$

AOD_C can also be calculated as $AOD–AOD_F$. A maximum difference of ±0.002 due to rounding errors was found for days when both methods are applicable.

Only the POLDER-3 aerosol products from pixels with a quality flag index ≥0.5 have been considered in the following discussion.

2.2. AERONET

AERONET is a global network of ground-based multi-spectral sun/sky photometers (Holben et al., 1998; 2001) dedicated to real time monitoring of aerosol properties and widely used as ground-based reference for validation of aerosol satellite retrievals (e.g., Goloub et al., 1999; Bréon et al., 2011). It uses standardized sun/sky photometers (CIMEL CE-318, Cimel Electronique, Paris) measuring solar extinction and sky radiances (at times with polarization) in the almucantar plane at wavelengths between 340 and 1020 nm (most commonly 440, 675, 870, and 1020 nm), that allow deriving a number of aerosol optical and microphysical parameters (Dubovik and King, 2000; Dubovik et al., 2006).

AOD and *AE* are obtained about every 15 minutes from the measurement of the direct sun extinction and are reported as the average of a triplet of acquisitions lasting approximately 30 s. We consider here AERONET *AOD* at 870 nm and the *AE* value obtained between 870 and 675 nm. For freshly calibrated and well maintained instruments, the accuracy in AOD is of the order of 0.01-0.02 regardless of the AOD value (Holben et al., 1998). The aerosol optical depth in the fine and coarse mode (AOD_F and AOD_C, respectively) are recalculated from the column-integrated particle volume size distribution retrieved between 0.1 and 30 µm in diameter by the inversion algorithm described in Dubovik and King (2000) and Dubovik et al. (2006). The fine and coarse modes of the retrieved volume size distribution are defined as the modes below and above a threshold diameter ($D_{cut\text{-}off}$) corresponding to the minimum in the particle size distribution. The $D_{cut\text{-}off}$ value is not fixed but can vary between 0.44 and 0.99 µm. AOD_F and AOD_C values are estimated by recalculating the extinction due to the fine and coarse modes of the aerosols. The latest AERONET retrieval scheme considers an aerosol mixture of polydisperse, randomly-oriented homogeneous spheroids with a fixed distribution of aspect ratios (Mishchenko et al., 1997) and provides fraction (in percentage) of non-spherical/spherical particles, i.e. f_{NS}/f_S (Dubovik et al., 2006). For clear sky, there are about 10 measurements per day of this fraction in the early day or late afternoon (solar zenith angle ≥50°).

We used AERONET V2 level-2 quality assured aerosol products. Seventeen coastal AERONET stations, shown in **Figure 1,** were selected in this study, (see also **Table 1** for their respective geographical coordinates and covered periods). Their regional distribution covers the entire western Mediterranean basin, including south Europe (e.g., near coastal stations of Barcelona, Toulon, Villefranche-sur-Mer…), North Africa (Blida), and island locations in the northern (Ersa), central (Palma de Mallorca) and southern (Lampedusa and Alboran) basin, therefore capturing the diversity of the aerosol population, resulting from the different sources contributing to the Mediterranean aerosol (desert dust, marine, urban and industrial pollution, and biomass burning). The dataset also includes the ground-based super-sites of Ersa and Lampedusa of the ChArMEx project (Mallet et al., 2016). Considering the 17 stations altogether, more than 18000 daily observations of AOD are available in total in both POLDER-3 and AERONET datasets, among which 6421 are concurrent (see section 3.2 below) and thus available for comparison. We did not consider for tentative matching with POLDER in this study a rather limited number (<100) of daily observations obtained from manual sun-photometers on-board ships in our area (Figure 1) and period of interest, which are also available from the Maritime Aerosol Network component of AERONET (Smirnov et al., 2011).

2.3. ChArMEx airborne measurements

The airborne measurements relevant to this paper were performed on-board the French ATR-42 environmental research aircraft of SAFIRE during two of the intensive observational periods of the ChArMEx project:

- The Transport and Air Quality (TRAQA) campaign, dedicated to the study of air pollutants transport from Europe to the Mediterranean, their evolution and their impact on regional air quality (Di Biagio et al., 2015; 2016; Nabat et al., 2015a; Rea et al., 2015);
- The Aerosol Direct Radiative Forcing on the Mediterranean (ADRIMED) campaign was dedicated to the characterization of aerosol optical properties in the Mediterranean and their direct radiative effect in clear sky conditions (Denjean et al., 2016; Mallet et al., 2016).

During TRAQA, the ATR-42, based at the Francazal airport near Toulouse, France (43°36'N, 1°26'E), conducted 17 flights from 20 June to 13 July 2012 encountering weather conditions favouring the transport of pollution aerosols from continental Europe, and particularly from the Rhone valley, the Gulf of Genoa and Barcelona, giving raise to AOD values in the range of 0.2-0.6 at 550 nm over the northwestern Mediterranean. From 17 to 23 June, and then on 29 June, two episodes of desert dust transport were observed in the free troposphere, increasing the AOD up to 1.4 on June 29. (Di Biagio et al., 2015; 2016). During ADRIMED, the ATR-42, based in Cagliari, Italy (39°15'N, 9°03'E), flew 16 scientific flights between 14 June and 4 July 2013 (Denjean et al., 2016; Mallet et al., 2016). Several episodes of desert dust transport from southern Algeria and Morocco and northern Algeria and Tunisia were observed over the western and central Mediterranean, particularly off the Balearic Islands and above the Lampedusa island offshore Tunisia (Denjean et al., 2016).The total optical depth at 550 nm remained moderate, in the order of 0.2-0.4 even during dust events (Mallet et al., 2016).

2.3.1. Airborne instrumentation measuring aerosol optical properties

2.3.1.1. PLASMA photometer

PLASMA (Photomètre Léger Aéroporté pour la Surveillance des Masses d'Air), developed by LOA (Laboratoire d'Optique Atmospherique, Lille), is a multi-spectral sun-photometer which measures the direct sun radiance and retrieves the AOD at 15 wavelengths between 343 and 2250 nm, including 869 nm (Karol et al., 2013). The estimated uncertainty ranges between 0.005 and 0.01 (Karol et al., 2013). PLASMA was operated during the ADRIMED campaign only, when it was mounted on the roof of the ATR-42, allowing the retrieval of a vertical profile of both the spectral AOD and the aerosol particle size distribution (Torres et al., 2017).

2.3.1.2. CAPS-PMex

The Cavity Attenuated Phase Shift in situ instrument (CAPS-PMex, Aerodyne Research Inc.) measures the extinction coefficient σ_{ext} at 532 nm with an estimated relative uncertainty of ±3.2% (Kebabian et al., 2007; Massoli et al., 2010; Petzold et al., 2013). The operating principle is based on the modulation of the frequency and the phase changes of the light emitted by a LED source due to aerosols, after correction of the Rayleigh scattering by the molecules present in the air mass. As described in Denjean et al. (2016), the instrument was available during the ADRIMED campaign only, when it was located inside the cabin behind the Communautary Aerosol Inlet (CAI), and operated at 0.85 L min^{-1} and with a temporal resolution of 1 second. In this paper, the extinction coefficient σ_{ext} is expressed in Mm^{-1} (1 Mm^{-1} = 10^{-6} m^{-1}).

2.3.1.3. Nephelometer

The scattering coefficient σ_{scat} at 450, 550 and 700 nm was measured by a spectral integrating nephelometer (model 3563, TSI Inc.) described extensively by Anderson et al. (1996) and Anderson and Ogren (1998). During both TRAQA and ADRIMED, the instrument was operated at 30 L min^{-1} with a temporal resolution of 1-2 seconds downstream the AVIRAD inlet also onboard the ATR-42 (Di Biagio et al., 2015; 2016; Denjean et al., 2016). The AVIRAD inlet estimated size cut-off, corresponding to the diameter at which particles are collected with a 50% efficiency, is 12 µm in optical diameter.

The instrument uses a halogen lamp as light source and three photomultipliers preceded by spectral filters. Due to the geometry of its sensing volume, the nephelometer measures the scattering coefficient (σ_{scat}) between 7° and 170° and the backscattering coefficient (σ_{bscat}) between 90° and 170°. The scattering Angström exponent (AE_{scat}) representing the scattering spectral dependence can be calculated as

$$\mathbf{AE_{scat}} = -\frac{\ln(\sigma_{scat,450}/\sigma_{scat,700})}{\ln(450/700)} \quad (3)$$

The relative uncertainty in σ_{scat} due to calibration, counting statistics and non-idealities of detector surfaces, is estimated to be ±1-2% for submicron aerosols and ±8-15% for supermicron aerosols (Müller et al., 2009). To these values usually adds the error related to the geometric truncation of the measured angular range of the scattering phase function due to the sensing volume (Anderson and Ogren, 1998). This truncation induces an underestimation of σ_{scat} and σ_{bscat}, which depends on the angular distribution of the scattered light, and thus on particle size. Anderson and Ogren (1998) have shown that the uncertainty induced by the underestimation of σ_{scat} can be parameterized by the scattering spectral dependence for submicron aerosols. This parameterization is not possible for aerosols of larger particle size (diameter greater than 1 µm), because the Ångström exponent tends to zero whereas the underestimation is important (50-60%) because of the increase of the forward scattering. In this case, the correction is performed by optical calculation if the particle size distribution and refractive index are known (Müller et al., 2009; Formenti et al., 2011). As for σ_{ext}, in this paper σ_{scat} is expressed in Mm^{-1}.

2.3.2. Aerosol particle size distribution

Because of its extent, the aerosol particle size distribution is measured in situ by the combination of several instruments, often based on different physical principles (Wendisch and Brenguier, 2013). In our work, we used a combination of different optical counters operating on the fine and coarse modes of the aerosols, that is:

- a Passive Cavity Aerosol Spectrometer Probe (PCASP, Droplet Measurement Technologies, Boulder, Colorado), operated at 632.8 nm with a temporal resolution of 1 second. The PCASP measures light scattering between 35 and 135° to derive the particle number size distribution over 31 channels between 0.1 and 3.0 µm in diameter (Liu et al., 1992; Reid et al., 1999). The PCASP was operated on a wing pod of the ATR-42 during the TRAQA campaign only.
- an Ultra High Sensitivity Aerosol Spectrometer (UHSAS, Droplet Measurement Technologies, Boulder, Colorado), operated at 1054 nm with a temporal resolution of 1 second. The UHSAS measures light scattering between 22 and 158° to derive the particle number size distribution over 99 size channels between 0.04 and 1.0 µm in diameter (Cai et al., 2008). The UHSAS replaced the PCASP under the aircraft wing during the ADRIMED campaign.
- a Sky-Grimm counter (1.129 model, Grimm Aerosol Technik; Grimm and Eatough, 2009), operated at 632.8 nm with a temporal resolution of 6 seconds. The instrument integrates light scattering between 30° and 150° to derive the particle number size distribution over 32 channels between 0.25 and 30 µm in diameter (Grimm and Eatough, 2009). The instrument was available during both TRAQA and ADRIMED, operated inside the aircraft cabin and behind the AVIRAD inlet. Due to a flow problem, measurements during TRAQA are restricted to the portions of the flights when the ATR-42 remained below 350 m above sea level.

3. Validation strategy

3.1. Matching POLDER-3 and in situ aircraft measurements

In situ aircraft measurements provided direct and indirect observations for validation. Direct observations of the total *AOD* were obtained by the reading of the PLASMA sun-photometer for those portions of the flights when the ATR-42 flew at its lowest altitude and by integrating the vertical profile of the extinction coefficient σ_{ext} measured by the CAPS-PMex instrument between the minimum and the maximum heights (z_{min} and z_{max}) of the ATR-42 during profile ascents or descents.

Indirect validation of the size-dependent optical depth (AOD, AOD_F and AOD_C) was performed by optical calculation from the number size distribution d$N(D,z)$/dlog$D$ measured by the combination of the PCASP, UHSAS and Grimm optical counters as

$$\mathrm{AOD_x}\ (865\ \mathrm{nm}) = \int_{\mathrm{z_{min}}}^{\mathrm{z_{max}}} \mathrm{dz}\ \sigma_{\mathrm{ext}}(\mathrm{z}) = \int_{\mathrm{z_{min}}}^{\mathrm{z_{max}}} \mathrm{dz} \int_{\mathrm{D_{x'}}}^{\mathrm{D_x}} \pi \mathrm{D}^2 \mathrm{Q_{ext}}(\mathrm{z, D, m}) \frac{\mathrm{dN(D,z)}}{\mathrm{dlogD}} \mathrm{dlogD} \quad (4)$$

The suffix x in Equation 4 indicates the size domain of the aerosol optical depth (total, fine or coarse) considered in the calculations.

Equation 4 allows one to estimate the aerosol optical depth over a variable size domain, whose boundaries ($D_{min'}$ and D_{max}) can be adjusted to represent the fine and the coarse modes, as well as the total particle size distribution.

The iterative procedure used for the calculation is presented in **Figure S1**. All calculations used the optical Mie theory for homogeneous spherical particles (Mie, 1908). The initial step of the procedure consisted in estimating the aerosol number size distribution, input of Equation 4, from the measurements of the PCASP, UHSAS and Grimm optical counters operated on board the ATR-42 during TRAQA and ADRIMED. This required two actions, described in details in the Supplementary material**.**

1. The conversion of the nominal “optical equivalent spherical diameter" (D_{EO}) characteristic of each particle counter to a "geometric equivalent spherical diameter" (D_{EG}). The operating principle of the particle optical counters is based on the angular dependence of the light scattering intensity to the particle size (Wendisch and Brenguier, 2013). The proportionality factor between angular light scattering and particle size depends on the particle complex refractive index. At calibration, the optical particle counters provide an “optical equivalent spherical diameter" (D_{EO}), corresponding to the diameter of standard material, generally spherical latex beeds, which refractive index (m_{latex} = 1.59-0i) is usually different from the real aerosol refractive index measured in the atmosphere. It is therefore necessary to convert the

measured D_{EO} value into a so-called "geometric equivalent spherical diameter" (D_{EG}) value taking into account the actual refractive index of ambient particles.

2. The combination of measurements over different size ranges. Since no optical counter completely covers the full size range of atmospheric aerosol particles, measurements of the PCASP, UHSAS and Grimm were combined by examining their agreement on their size overlap domains. When successful, the particle number size distribution obtained by the combination was normalised to the total particle number and fitted using a multi-mode lognormal distribution to eliminate discontinuities and extend the representation beyond the lower and upper operating size ranges of the optical counters.

The capability of the derived number size distributions to represent the aerosol extinction coefficient, henceforth to estimate aerosol optical depth, was assessed by comparing the calculated extinction and scattering coefficients σ_{ext} and σ_{scat} to the measurements of the CAPS-PMex and the nephelometer at 450, 532, 550 and 700 nm. The scattering coefficient σ_{scat} was calculated by integrating the scattering phase function between 7° and 170°, corresponding to the aperture of the sensing volume of the nephelometer.

All optical calculations performed in this paper assumed the spectral complex refractive index m, representing the aerosol composition, as independent of size. An initial dataset per aerosol type was chosen (Table S2 in the Supplementary material). The calculations were iterated by varying the initial values of the complex refractive indices until both 1/ the adjusted value for the calculation of the extended size distributions and 2/ the comparison between calculations and measurements of the extinction and scattering coefficients agreed within errors. Results of these comparisons are presented in the Supplementary material.

3.2. Constitution of the data set

This section describes the choices of temporal and spatial coincidences adopted for the comparisons between POLDER-3, AERONET and in situ data.

3.2.1. Coincidence with AERONET

As described in previous evaluation studies of aerosol products derived from satellites (e.g., Bréon et al., 2011), two approaches can be considered in order to compare coincident ground-based photometer and satellite aerosol data. One option is to select only the closest (in time) photometer measurement and the closest (in distance) satellite pixel from the photometer site. Another method consist in performing averaging within a certain time window for photometer data, and a spatial average of the satellite data within a given distance from the photometer site. Bréon et al. (2011) have shown that these two approaches give very comparable results for POLDER-3 aerosol products over oceans. In this study we adopted the second approach, considering the POLDER-3 aerosol products from pixels within ±0.5° around the AERONET sites. For AERONET *AOD* and *AE*, the averaging temporal window was set to ±1 h around the time of the POLDER-3 passage. For AERONET AOD_F, AOD_C, and shape retrieval, this temporal window produces an insufficient number of data, in particular for springs and summers in the period 2005-2011 due to the temporal time shift of the POLDER-3 passage towards the afternoon. For these two variables, the averaging temporal window was extended to the whole afternoon (that is, all data points later than 12:00 UTC) in order to allow for a significant dataset for comparison.

Table 1 reports the number of available observational days for POLDER-3 and AERONET aerosol parameters at each station in the period March 2005-October 2013, as well as the number of coincident days obtained between POLDER-3 and AERONET. The stations are ranged regarding the number of coincident days obtained for *AOD* and *AE*, this number representing the upper limit of the number of common POLDER-3/AERONET observations days available. Including all 17 stations, 18634 occurrences of comparable POLDER-3 and AERONET observations are available for *AOD*, *AE*, AOD_F and AOD_C, and 7923 occurrences for AOD_{CS} and AOD_{CNS}, due to specific constraints on geometric conditions in the POLDER-3 algorithm necessary to derive shape-related parameters (non sphericity). Per site, the number of clear sky observational days for POLDER-3-derived *AOD*, *AE*, AOD_F and AOD_C varies from 668 to 1392. Part of this variability also depends on the percent of sea pixels in the 1° x 1° area around the sites, which is lower for coastal (e.g., Burjassot or Roma) than insular stations (e.g., Alboran, Lampedusa or Gozo). Between 1 pixel in the case of inland stations of

Roma and Burjassot, and up to 29 pixels in the case of the small remote island of Alboran were considered. Overall, the number of available AERONET observation days is important both for *AOD* and *AE* (18223), and AOD_F and AOD_C (11228). The number of days with AERONET-derived f_{NS} was less significant (4976 data points), due to additional constrains in the inversion necessary to derive this parameter.

The number of available AERONET observations per site varied from 158 to 2059 for *AOD* and *AE*, and from 43 to 1333 for AOD_F and AOD_C, mainly due to partial functioning of the instruments or maintenance of the sites. At some stations, measurements started years after the beginning of POLDER-3 mission (e.g., 2011 for Alboran, 2013 for Gozo). Finally, the number of POLDER-3/AERONET coincident days available for analysis is 6421 for *AOD* and *AE*, 3855 for AOD_F and AOD_C, and 730 for the percentage of spherical coarse particles (f_{NS}).

3.2.2. Coincidence with airborne observations

The comparison between POLDER-3 and airborne measurements was conducted for profile ascents or descents of the ATR-42 close in time with POLDER-3 overpasses. Flight tracks and profiles locations are shown in **Figure 2**, whereas additional details (dates, geographical coordinates, altitude span and duration) are given in **Table 2**. Data from the PLASMA sun-photometer, operated only during ADRIMED, were available only on 8 profiles (also indicated in Table 2) for which the minimum flight altitude was as close as possible to the surface. The data set was limited to ATR-42 profiles extending as much as possible over the column. To evaluate whether the aircraft profile sampled entirely or only partially the aerosol layers, we compared the *AOD* measured by PLASMA to that obtained by integrating the extinction profile of the CAPS-PMex instrument (not shown). By examining the AERONET time series, we also excluded episodes when the *AOD* had significantly varied in time between the POLDER-3 overpass and the aircraft profile. This mostly happened for cases when the aerosol optical depth exceeded 0.2 due to the transport of mineral dust (flights T-V22 and T-V23 during TRAQA and V31-S3 and V42-S2 during ADRIMED). The profiles discarded for comparison with POLDER-3 were used for the validation of the optical calculations presented in section 4 (not shown in Table 2 nor Figure 2).

Prior to analysis, all in situ airborne data were synchronised and then averaged to 30 seconds to reduce the noise due to the native resolution of the measurements (1 to 6 seconds). POLDER-3 data were averaged over pixels within ±0.5° around the lowest altitude of each profile. In order to analyse the aerosol vertical stratification, we examined the magnitude of the scattering coefficient σ_{scat} at 550 nm as a function of altitude and its spectral behaviour, represented by the scattering Angström Exponent (AE_{scat}) measured by the airborne nephelometer. As in previous similar studies (Pace et al., 2006; Formenti et al., 2011; Di Biagio et al., 2015; 2016; Denjean et al., 2016), the aerosol layers were classified in four categories (clear/background maritime, desert dust, pollution, and mixture), following the criteria reported in **Table 3**. The mixture category, indicating mixing between desert dust and pollution, as observed by Denjean et al. (2016), was further detailed to distinguish dust-dominated layers (AE_{scat} between 0.5 and 0.75) and pollution-dominated layers (AE_{scat} between 0.75 and 1).

3.3. Statistical indicators

The agreement between the POLDER-3, AERONET and airborne datasets was quantified by several evaluation metrics, including the number of matchups (*N*), the linear correlation coefficient (*R*), the slope (*S*) and intercept (*I*) of the linear regression, the root mean square error (*RMS*), and the bias (*B*), representing their mean difference.

$$RMS = \sqrt{\frac{1}{\mathrm{n}}\sum_{\mathrm{i}=1}^{\mathrm{n}}(\mathrm{y_i} - \mathrm{x_i})^2} \quad (5)$$

$$B = \frac{1}{\mathrm{n}}\sum_{\mathrm{i}=1}^{\mathrm{n}}(\mathrm{y_i} - \mathrm{x_i}) \quad (6)$$

where x and y are generic datasets, and n the number of pairs of compared values.

Additional metrics is provided by the “fraction of accurate retrievals” (G_{frac}) defined by Bréon et al. (2011). This quantity is defined as

$$G_{frac} = \frac{\#\mathrm{obs}(\Delta < EE)}{\#\mathrm{obs}} \quad (7)$$

and quantifies the fraction of POLDER-3 data points for which the absolute difference (Δ) between reference and evaluated data is lower than the estimated error (*EE*).

In accordance to Bréon et al. (2011), EE was calculated as

$$EE = \pm\,(0.03 + 0.05 \times AOD) \qquad (8)$$

and applied to all the *AOD* advanced products. Because G_{frac} is only appropriate for large datasets whose number of data points exceeds 100 (Bréon et al., 2011), it was calculated only for comparisons with AERONET data.

4. Results

4.1. Evaluation of the total aerosol optical depth

Figure 3 shows the results of comparison of the *AOD* retrieved by POLDER-3 between 2005 and 2013 with respect to the 6421 observations at the seventeen AERONET stations and those on the vertical profiles of the ChArMEx campaigns (PLASMA sun-photometer and calculations from the in situ size distributions).

The comparison with AERONET shows a good correlation (regression coefficient R =0.88, G_{frac} =73%), with a statistically low dispersion and bias (*RMS* =0.04, *B* =0.003). Twenty-seven percent of the observations do not meet the criteria of the G_{frac} parameter. Cases outside the G_{frac} boundary were characterized by large standard deviations, either because the spatial distribution of *AOD* was heterogeneous in the 1° x 1° area of the pixels surrounding the AERONET sites, or because it varied significantly on the time window of ±1 hour around the POLDER-3 overpass. In our dataset, the highest value of *AOD* retrieved by POLDER-3 was 1.4 (±0.1) during a desert dust transport event over Lampedusa observed on April 25, 2011. This is the only event coincident with an AERONET measurement (1.50 ±0.06) with POLDER-3 *AOD* >1.

Figure 3 also shows the comparison with the PLASMA observations and with the calculations initiated by the measured airborne number size distributions.

On those, the *AOD* did not exceed 0.2, whereas *AE* ranged from 0.31 ±0.07 to 1.09 ±0.08, indicating that these cases are representative of aerosols of different origins. The comparison was also very satisfactory and confirmed the more extensive results from the comparison with AERONET-derived *AODs*. POLDER-3 provides higher values of *AOD* for mineral dust (lowest *AE* values) compared to those calculated from in situ aerosol measurements, which could reflect an underestimate of the coarse mode distribution from the in situ aircraft measurements. On the other hand, POLDER-3 tends to underestimate *AOD* with respect to PLASMA at low AE values, resulting in a negative bias of the correlation (*B* =-0.02). In both cases, *RMS* remained low and below 0.05.

4.2. Evaluation of fine and coarse aerosol optical depth

4.2.1. Comparison with AERONET observations

Figure 4 shows the comparison between POLDER-3 and AERONET for AOD_F and AOD_C. AOD_F remained below 0.25, smaller than AOD_C, which reached 0.8. The correlation coefficient for AOD_C (*R* =0.81) is closer to the correlation coefficient for *AOD* (0.88) than that for AOD_F (0.63). The agreement between POLDER-3 and AERONET is confirmed by the G_{frac} values of 74% for AOD_C and 88% for AOD_F, the low statistical bias (-0.007 for AOD_F and 0.01 for AOD_C), and the moderate dispersion (RMS values between 0.02 for AOD_F and 0.04 for AOD_C). The weaker correlation and the dispersion observed for AOD_F can be attributed to the difficulty in retrieving low values of optical depth. Additionally, Tanré et al. (2011) pointed out that differences could arise by the definitions of the cut-off diameter ($D_{cut\text{-}off}$) used in the POLDER-3 and AERONET retrievals to estimate AOD_F. In the AERONET retrievals, AOD_F is calculated from the fine mode of the particle size distribution defined for a value of $D_{cut\text{-}off}$ forced between 0.44 and 0.99 µm. In the POLDER-3 algorithm, AOD_F is calculated from the full particle size distribution of the retrieved fine mode, without cuf-off. However, because of its use of polarisation, POLDER-3 is the most sensitive to particles smaller than 0.6-0.8 µm in diameter (Tanré et al., 2011 and references therein).

In **Figure 5**, we explore the relevance of this difference in the comparison of AOD_F and AOD_C by further separating days when AERONET $D_{cut-off}$ <1.0 µm and days when $D_{cut-off}$ ≥1.0 µm. The threshold value of 1.0 µm corresponds to the D_{eff} of all the fine modes in the POLDER-3 LUT. Cases with $D_{cut-off}$ <1.0 µm were more numerous (2413 days), and showed a better agreement (B = -0.003, G_{frac} = 91%, RMS =0.02, R =0.60). Data corresponding to $D_{cut-off}$ ≥1.0 µm were less numerous (1442 days). Whereas the correlation improved slightly (R =0.69 versus R =0.60), the dispersion increased (B =-0.01, RMS =0.03) due to the appearance of points for which AERONET AOD_F almost doubled that of POLDER-3. Colouring the data points by AE showed that the data points with $D_{cut-off}$ below 1.0 µm mostly corresponded to aerosols with a weak-to-moderate spectral dependence (low AE), whereas cases with $D_{cut-off}$ above 1.0 µm mostly (but not exclusively) corresponded to aerosols with a moderate-to-strong spectral dependence (high AE).

The size cut-off definition also affects the comparison for AOD_C. For $D_{cut-off}$ <1.0 µm, AOD_C values were high and the correlation was significant. Conversely, AOD_C remained low (≲0.2) when $D_{cut-off}$ ≥1.0 µm. This is consistent with the fact that the contribution of AOD_C to AOD decreases as the $D_{cut-off}$ increases (**Figure S2** in the supplementary material). **Figure 5** shows that discriminating data on the basis of $D_{cut-off}$ results in attributing AOD_F and AOD_C to different aerosol types.

4.2.2. Comparison with airborne measurements

To understand further the previous comparisons, POLDER-3 AOD_F and AOD_C were recalculated from the measured number size distributions (Equation 4) by varying the lower limit of the size integration between 0.4 and 1.0 µm in diameter with a step of 0.2. Results are shown in **Figure 6**. As expected, the comparison for AOD_F is very sensitive to the size range. The best agreement between the retrieved and the calculated AOD_F is obtained for $D_{cut-off}$ between 0.6 and 0.8 µm, both showing high correlation coefficient R and low RMS. Conversely, the AOD_C comparison is almost independent of the value of $D_{cut-off}$ but more affected by the upper limit of the size range in Equation 4.

4.3. Evaluation of the Ångström Exponent

Figure 7 shows the comparison of AE retrieved by POLDER-3 with values obtained by AERONET, PLASMA and the optical calculations. The comparison with AERONET was restricted to days when the POLDER-3 *AOD* exceed 0.1 (2031 out of the 6421 data points) to take into account only those values with relative uncertainties within 50%. The comparison showed a significant spread and a moderate correlation coefficient (R =0.70). However, POLDER-3 tends to underestimate values of *AE* larger than 1 with respect to AERONET, and overestimate values smaller than 0.5, yielding a significant bias (–0.11). The values obtained by POLDER-3 compare well with the airborne observations of PLASMA (R =0.84), but less well to the optical calculations (R =0.42). In both cases, the bias is positive (0.1 with PLASMA and 0.2 with in situ AE). This fact, observed previously by Goloub et al. (1999) and Tanré et al. (2011), can be explained by considering that the values of AE are calculated from the retrieved *AOD* at 865 and 670 nm (Equation 1), which, in the ocean retrieval algorithm of POLDER, is obtained by the fit of measured radiances. The current aerosol models in the LUT (modal diameters and real part of the refractive index) provide *AE* values in the range –0.18 to 3.3. However, the extreme values are obtained only if the size distribution allowing to match the observed radiances consists of a single mode of non-spherical coarse particle (modal diameter of 0.9 µm for *AE* =–0.18) or a single mode of fine spherical particles (modal diameter of 0.08 µm for *AE* =3.3). **Figure 8** compares the scatterplots of *AE* and *AOD* obtained for the coincident POLDER-3 and AERONET datasets. The tendency of POLDER-3 to underestimate *AE* shows up clearly by the absence of values of *AE* larger than 2.5, which, conversely, are retrieved by AERONET. On the other end of the spectrum, values down to –0.5 are found in the AERONET data set when POLDER-3 hardly retrieves negative values. Both POLDER-3 and AERONET show a trend with the largest AOD values at lower *AE* values. However, high *AOD* values (>0.9) are found with POLDER but not AERONET, and are all except one associated to relatively low *AE* (<1). Because the cloud screening of AERONET is relatively robust thanks to triplet measurements (Smirnov et al., 2000), these outliers may result from undetected cloud contamination in the POLDER algorithm.

4.4. Evaluation of aerosol sphericity

When the geometrical conditions of observations are favourable, the coarse mode optical depth (*AOD_C*) retrieved by POLDER-3 is quantified and apportioned into a spherical and a non-spherical fraction (AOD_{CS} and AOD_{CNS}, respectively). These products are potentially very useful in discriminating the mineral dust contribution, dominated by non-spherical coarse particles (e.g., Dubovik et al., 2002; Chou et al., 2008), when marine aerosols can be considered as spherical at relative humidities characteristics of coastal and open-sea sites (Sayer et al., 2012a; 2012b).

As a prerequisite, we investigated the comparison between POLDER-derived f_{CNS} (percent fraction of non-sphericity in the coarse mode AOD_C, that is, $f_{CNS} = AOD_{CNS}/(AOD_{CNS}+AOD_{CS})$ retrieved by POLDER-3 and f_{NS} (percent of non-sphericity of the total *AOD*) estimated by AERONET. In the operational ocean algorithm, f_{CNS} can only take discrete values equal to 0, 0.25, 0.50, 0.75, and 1. The averaging process produces intermediate values when there is local variability between the pixels around a given AERONET station

In general, the POLDER-3 f_{CNS} and the AERONET f_{NS} are poorly correlated. The correlation coefficient *R* is 0.29 for the coincident data points of all the 17 stations (*N* =730, **Table 1**). At individual coastal and insular stations (Lampedusa and Malaga), notably impacted by mineral dust , the correlation between POLDER-3 f_{CNS} and AERONET f_{NS} is more significant (*R* =0.73 for *N* =54 and *R* =0.59 for *N* =53, respectively). This is also seen when restricting the data set of Ersa and Lampedusa to the summers of 2012 and 2013 (*R* =0.55 at Ersa, *N* =11; *R* =0.70 at Lampedusa, *N* =10).

The robustness of the comparison can be increased by further constraining the dataset to POLDER-3 and AERONET *AOD* values larger than 0.10 and limiting the comparison to AERONET data for which AOD_C is at least 30% of the total *AOD*. By applying these thresholds (**Figure 9a**), the correlation between f_{CNS} and f_{NS} is R = 0.56 (N = 274 for the 17 stations)**.** Overall, 80% of the POLDER-3 f_{CNS} agrees within 25% with the AERONET values. The largest differences occur when AERONET retrieves f_{NS} values lower than 50%. In this case, only 40% of the POLDER-3 f_{CNS} are in the ±25% agreement interval with AERONET. Conversely, for AERONET f_{NS} >50%, 88% of the POLDER-3 f_{CNS} agree within ±25% with the AERONET estimate of f_{NS}.**Figure 10** shows that a relatively good agreement is obtained by comparing broad classes 25% wide, providing confidence to the classification of non-sphericity by POLDER-3.

Finally, Figure 10 shows the implication on those results on the evaluation of the POLDER-3 AOD_{CNS} and AERONET AOD_{NS}. With the previous thresholds (POLDER 3 and AERONET AOD values larger than 0.10 and AERONET AOD_{C}/AOD larger than 30%), the correlation obtained between coincident POLDER-3 AOD_{CNS} and AERONET AOD_{NS} at 865 nm is significant (R = 0.87).

The two datasets are very consistent. However, the POLDER-3 AOD_{CNS} is almost systematically lower than the AERONET AOD_{NS}, regardless of the percent that it represents with respect to the AOD (not shown). The physical reasons behind this evident discrepancy are beyond the scope of this paper and we recommend addressing them in future research.

5. Discussion

5.1. Evaluation of uncertainties on the advanced POLDER-3 oceanic aerosol products

In this paper we provide a first comprehensive evaluation of the advanced POLDER-3 aerosol products over ocean by the latest operational algorithm, based on ground-based remote sensing (AERONET) but also airborne remote sensing and in situ observations (TRAQA and ADRIMED campaigns) over the western Mediterranean sea. **Table 4** summarizes it by presenting the absolute errors (Δ) derived from the RMS (representing the precision) and the bias (B) as a measure of accuracy. For consistency with previous similar analyses and as an acknowledgment of the large size of the dataset, only the *RMS* and bias of the linear regressions with the AERONET data have been reported. The uncertainties in AOD_{CS} and AOD_{CNS} were calculated as the square-root of the quadratic sum of the errors in AOD_{C} and f_{CNS}.

5.2. Our estimate of ΔAOD indicates that, for the western Mediterranean basin, the accuracy and the precision of the POLDER-3 are better than those derived by the error analysis of Tanré et al. (2011), also reported in Table 4, based on a global comparison with AERONET of the POLDER-1 instrument. It is noteworthy that the POLDER-1 retrieval algorithm was using a single mode spherical particle size distribution (Goloub et al., 1999) instead of the current two

modes allowing, in addition, an aspherical component. Furthermore, from our regional evaluation of the whole latest collection 3 of the POLDER-3 data set, G_{frac} value for *AOD* (73%) is much better than that reported by Bréon et al. (2011) (G_{frac}= 45%), based on previous collection of POLDER-3 retrievals at a global scale. Evaluation of the fine and coarse aerosol optical depth

Table 4 reports the uncertainties in AOD_F and AOD_C based on estimates *RMS* and bias. It is interesting to notice that the precision in AOD_C is apparently lower than in AOD_F (higher *RMS*), despite the correlation being far better for the former than for the latter. We have shown that the direct comparison between POLDER-3 and AERONET should take into account the differences in the definition of the fine size fraction in the respective retrieval algorithms. The AERONET AOD_F is recalculated from the fine mode of the volume size distribution retrieved from the measured total radiance, and defined as the mode below an upper limit diameter ($D_{cut\text{-}off}$) varying between 0.88 and 1.98 µm. Conversely, our comparison with airborne measurements indicates that AOD_F retrieved by POLDER-3 corresponds to a fine mode extending to values of $D_{cut\text{-}off}$ between 0.6 and 0.8 µm. This is expected as POLDER-3 uses polarised radiances, highly sensitive to fine particles, in agreement with previous regional validations of POLDER AOD_F over land (Kacenelenbogen et al., 2006; Fan et al., 2008; Wang et al., 2015). Our comparison with in situ data shows that the POLDER-3 AOD_C is less sensitive to the $D_{cut\text{-}off}$ value (**Figure 6**), but mostly to the extent of the coarse mode towards the largest particles.

It should also be noted that the values of AOD_F might be biased low at 865 nm, which will result in a lower *RMS* and correlation as compared to the coarse mode (see Figures 4 and 5). This mightminimise the effects of the lack of aerosol absorption in the POLDER algorithm, which could affect the retrieval of pollution and dust aerosols at shorter wavelengths where absorption is more significant.

5.3. Regional aerosol distribution

The ability of POLDER-3 in representing the spatial distribution of aerosols in the Mediterranean region is demonstrated in Figure 11 showing the retrieved products averaged over the operating period. These regional maps highlights a north-south gradient for *AOD* and AOD_{CNS}, with, on average, the highest values in the southernmost part of the western Mediterranean region, especially over south Ionian Sea off Libya, as previously reported by former satellites *AOD* products (e.g., Moulin et al., 1998; Antoine and Nobileau, 2006). The distribution of POLDER-3 *AE* indicates high values along the European coasts (especially over the Adriatic Sea), and low along the North African coasts indicative of the dominance of desert dust in the South and anthropogenic aerosol in the North of the basin. AOD_F and AOD_{CS} maps show moderate spatial variability over the basin, associated to averaged values (AOD_F of 0.033, AOD_{CS} of 0.021) 2 to 3 times lower than those retrieved by POLDER-3 for AOD_{CNS} (0.065). Despite these low spatial patterns, it is noticeable that AOD_F values tend to increase in the Eastern part of our region of study, suggesting the complexity of various aerosol types influences over the Mediterranean Sea.

The detailed investigation of the aerosol climatology and regional distribution of the aerosol optical depth of the fine and coarse mode aerosol, including spherical and non-spherical components, retrieved by POLDER-3 over the western Mediterranean Sea, will be presented in the second part of this paper. This analysis, including the investigation of temporal trends over the 8-year operating period, will provide an important support to the ongoing aerosol research in the region.

6. Concluding remarks

The western Mediterranean aerosol is a complex mixture with a significant temporal and spatial variability at small scales (Pace et al., 2005; 2006; Di Iorio et al., 2009; Mallet et al., 2016 and references therein), and significant impact on present and future regional climate (Nabat et al., 2014; 2015a; 2015b; 2016). High-resolved long-time series of spaceborne observations of aerosol optical depth on different size classes and for differing particle shapes, such as provided by POLDER-3, are essential in exploring those evolutions, directly, but also indirectly, as a term of comparison for climate and transport models (Nabat et al., 2014). In the past, quantitative remote sensing of the aerosol optical depth has proven most useful in establishing decadal climatology of the transport of mineral dust over the basin, highlighting its seasonal variability, geographic distribution and sources, link to large-scale atmospheric dynamics (Dulac et al., 1992; Moulin et al., 1997a; 1997b; 1998; Antoine and Nobileau, 2006; Papadimas et al., 2008).

The quality of the observations is surely key to those surveys, and has motivated the comparative analysis of the advanced POLDER-3 oceanic aerosol products during the whole period of operation (March 2005 to October 2013) presented in this paper, with regards to co-located and coincident ground-based measurements by AERONET, and airborne vertical profiles of aerosol optical depth and size distribution during the TRAQA and ADRIMED campaigns of the ChArMEx project.

The results presented in this paper indicate that overall the operational oceanic algorithm of POLDER-3 provides with a very good evaluation of the various components of the aerosol optical depth at the regional scale of the Western Mediterranean. Our results confirm previous validations (Goloub et al., 1999; Kacenelenbogen et al., 2006; Fan et al., 2008; Bréon et al., 2011; Tanré et al., 2011), and provide a first evaluation of the uncertainties on the fine and coarse fractions of the aerosol optical depth, and the partitioning of the coarse mode *AOD* into its spherical and non-spherical components. We highlighted some differences with respect to AERONET and the in situ data, for example in the evaluation of the Angstrom exponent and the non-spherical coarse fraction of the AOD. The physical reasons behind those differences remain unresolved. They might require re-examining the basic assumptions of the LUT or the observational constraints, which is beyond the scope of this paper, but which we recommend addressing in future research.

Our results advance the classification of Mediterranean aerosols, and in particular in the investigation of the anthropogenic fraction, which is relevant to climate change. As a matter of fact, our results indicate that the fine-fraction *AOD* at 865 nm is contributed by the aerosol accumulation mode below 0.6-0.8 µm in diameter. On the basis of this result, we recommend that any further comparison to AERONET should be restricted to values corresponding to $D_{cut\text{-}off}$ < 0.8 µm. This suggests that AOD_F measured by POLDER-3 could be used for predicting the submicron column concentrations for air quality studies, and for evaluating the radiative effect of fine aerosols.

Data availability

POLDER-3 data extraction was performed with the program PARASOLASCII (http://www-loa.univ-lille1.fr/~ducos/public/parasolascii/). This version is made available from the AERIS Data and Service Center (http://www.icare.univ-lille1.fr/parasol). Technical details are described at http://www.icare.univ-lille1.fr/projects_data/parasol/docs/Parasol_Level-2_format_latest.pdf. The definition of the flag index is detailed at page 18 (parameter: quality of the fit).

The AERONET version 2.0 aerosol products at the level 2.0 quality (cloud screened and quality assured with up-to-date calibration) were obtained from the official website at http://aeronet.gsfc.nasa.gov/.

The SAFIRE ATR-42 aircraft data are available at the Mistrals/ChArMex database maintained by the AERIS Data and Service Center (http://mistrals.sedoo.fr/ChArMEx/).

Single particle Mie scattering calculations were performed with the Mie_single.pro routine under IDL available at http://eodg.atm.ox.ac.uk/MIE/mie_single.html.

Competing interests

The authors declare that they have no conflict of interest.

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Table 1. List of AERONET stations available in the western Mediterranean region retained for this study. The number of ocean POLDER pixel within 0.5° from the position of the station is indicated (N_{PIXEL}). The number of observations by POLDER-3 and AERONET between March 2005 to October 2013, and the number of coincident days (within brackets) are also reported.

AERONET station	**Latitude, Longitude**	**Altitude (m)**	**AERONET period**	**N_{PIXEL}**	**AOD and AE**	**AOD_F and AOD_C**	**f_{CNS} and f_{NS}**
					POLDER/AERONET (coincidences)		
Barcelona	41°23'N, 02°07'E	125	04/03/2005 - 10/10/2013	13	1171/2059 (827)	1171/1333 (514)	485/623 (116)
Villefranche-sur-Mer	43°41'N, 07°19'E	130	17/02/2005 - 21/08/2013	9	1097/1589 (641)	1097/999 (359)	470/452 (77)
Toulon	43°08'N, 06°00'E	50	04/03/2005 - 04/12/2010	9	1114/1503 (630)	1114/962 (343)	429/393 (67)
Ersa	43°00'N, 09°21'E	80	09/06/2008 - 11/10/2013	17	1178/1252 (541)	1178/676 (281)	504/240 (37)
Malaga	36°42'N, 04°28'W	40	23/02/2009 - 23/09/2013	10	1193/1359 (539)	1193/1036 (419)	465/377 (53)
Lampedusa	35°31'N, 12°37'E	45	06/03/2005 - 11/10/2013	28	1301/1177 (513)	1301/663 (307)	604/285 (54)
Messina	38°11'N, 15°34'E	15	01/05/2005 - 23/20/2012	9	1119/1340 (507)	1119/739 (281)	538/399 (63)
Roma Tor Vergata	41°50'N, 12°38'E	130	10/03/2005 - 11/10/2013	1	725/1954 (486)	725/1199 (280)	297/683 (66)
Blida	36°30'N, 02°52'E	230	06/03/2005 - 19/02/2012	7	989/1357 (475)	989/813 (280)	427/484 (85)
Burjassot	39°30'N, 00°25'W	30	16/04/2007 - 24/04/2013	1	668/1506 (372)	668/1045 (277)	249/480 (54)
Palma de Mallorca	39°33'N, 02°37'E	10	03/08/2011 - 10/10/2013	11	1136/524 (214)	1136/395 (155)	504/162 (19)
Porquerolles	43°00'N, 06°09'E	22	10/05/2007 - 17/07/2013	11	1106/537 (195)	1106/260 (95)	431/82 (9)
Frioul	43°15'N, 05°17'E	40	07/07/2010 - 11/10/2013	8	1037/481 (162)	1037/324 (118)	373/91 (10)
Gozo	36°02'N, 14°15'E	32	25/02/2013 - 11/10/2013	24	1320/210 (102)	1320/162 (67)	633/90 (9)
Montesoro Bastia	42°40'N, 09°26'E	49	26/07/2012 - 23/07/2013	14	1161/240 (76)	1161/43 (7)	506/12 (1)
Alboran	35°56'N, 03°02'E	15	29/06/2011 - 23/01/2012	29	1392/158 (73)	1392/103 (46)	609/47 (7)
Tizi Ouzou	36°41'N, 04°03'E	133	11/04/2012 - 11/10/2013	5	927/238 (68)	927/98 (26)	399/76 (3)
TOTAL	-	-	-	-	18634/18223 (6421)	18634/11228 (3855)	7923/4976 (730)

Table 2. List of vertical profiles made by the SAFIRE ATR-42 during the TRAQA and ADRIMED campaigns in coincidence with the passage of POLDER-3. For each profile is indicated: the flight number, the name of the profile, the date, the time period of the profile, the area covered by the flight, the geographical coordinates, the minimum and maximum altitude of the flight and then, the hour of POLDER-3 overpass in UTC.

Campaign	Flight ID	Profile ID	Date	Time (UTC)	Area	Geographical span		Altitude span (m asl)	POLDER-3 overpass (UTC)	PLASMA
						Beginning	end			
TRAQA	T-V21	T-V21-S1	27/06/2012	10h31–10h52	Corse	42°59'N–7°43'E	42°59'N–7°41'E	122–3534	14h19	----
	T-V24	T-V24-S1	03/07/2012	15h39–16h08	N-East Barcelona	42°14'N–3°31'E	42°8'N–3°29'E	77–3832	15h03	----
	T-V25	T-V25-S1	04/07/2012	08h32–09h04	South of France–Lion Gulf	41°28'N–6°0'E	41°31'N–6°0'E	100–4444	14h05	----
	T-V26	T-V26-S1		16h08–16h41		42°45'N–4°13'E	42°46'N–4°13'E	128–4684		----
	T-V27	T-V27-S1	06/07/2012	09h01–09h26	South of France	42°41'N–5°19'E	42°39'N -5°14'E	115–4723	13h47	----
		T-V27-S3		09h26–11h00		42°39'N - 5°15'E	42°42'N–5°19'E	76–3782		----
	T-V28	T-V28-S2		16h20–16h42		42°19'N–7°35'E	42°44'N–6°22'E	60–3784		----
ADRIMED	A-V28	A-V28-S2	14/06/2013	10h19–10h44	East Corse–Sardinia	41°38'N–7°14'E	42°4'N–6°46'E	69–3860	14h56	Yes
	A-V29	A-V29-S1	16/06/2013	08h19–08h32	Baleares–Sardinia	39°15'N–9°3'E	39°40'N–8°59'E	6–3877	14h37	Yes
		A-V29-S4		09h46–10h15		39°34'N - 4°29'E	39°39'N -4°29'E	52–4521		Yes
	A-V30	A-V30-S1	16/06/2013	11h59-12h10	Baleares–Sardinia	39°52'N - 4°13'E	39°32'N -3°48'E	93–3240	14:37	Yes
	A-V31	A-V31-S4	17/06/2013	09h41–09h54	Baleares–Sardinia	40°11'N–3°59'E	39°52'N–4°13'E	95–2899	15h18	Yes
	A-V32	A-V32-S1		11h46–12h05		39°52'N–4°13'E	39°56'N–4°36'E	93–4519		Yes
		A-V32-S4		13h30–13h44		39°32'N–9°10'E	39°16'N–9°2'E	10–3548		Yes
	A-V33	A-V33-S2	19/06/2013	12h47–13h17	Corse–Sardinia	43°01'N–9°23'E	43°1'N–9°20'E	73–4502	15h00	Yes
		A-V33-S4		14h46–14h59		39°15'N–9°24'E	39°15'N–9°4'E	5–3224		----
	A-V38	A-V38-S2	28/06/2013	12h25–13h30	Sardinia–Lampedusa	35°30'N–12°38'E	35°30'N–12°37'E	12–5427	14h26	----
	A-V44	A-V44-S1	04/07/2013	12h22–12h33	Gulf of Genoa–Corse–Sardinia	43°02'N–9°15'E	43°2'N–9°19'E	59–3513	15h11	----
		A-V44-S2		14h35–14h51		43°35'N–9°7'E	39°15'N–9°4'E	4–3499		----

Table 3. Criteria of classification of aerosol layers encountered on the vertical profiles of TRAQA and ADRIMED, based on nephelometer measurements of the scattering coefficient (σ_{scat}) at 550 nm and on its spectral dependence (AE_{scat}) between 450 and 700 nm.

Aerosol type	AE_{scat}(450-700 nm)	σ_{scat} (550 nm)
Clean background / maritime	–	< 5 or 10 Mm^{-1}
Desert dust	< 0.5	> 10 Mm^{-1}
Pollution	> 1	
Mixed (dust-dominated)	0.5 – 0.75	> 10 Mm^{-1}
Mixed (pollution-dominated)	0.75 – 1	

Table 4. Summary of evaluated uncertainties on POLDER-3 advanced products AOD, AE, AOD_F, AOD_C, and f_{CNS}, and comparison to previous evaluations. N/A stands for not attributed.

Products	**Uncertainties**	
	This work	**Previous work**
AOD	$\Delta AOD = \pm(0.003 + 0.04 \times AOD)$	$\Delta AOD = \pm(0.05 \times AOD + 0.05)^{\$}$
AE	$\Delta AE = \pm(0.11 + 0.44 \times AE)$	$\Delta AE = 0.3–0.5^{\$}$
AOD_F	$\Delta AOD_F = \pm(0.007 + 0.02 \times AOD_F)$	N/A
AOD_F *($D_{cut\text{-}off}$ < 1 µm)*	$\Delta AOD_F = \pm(0.003 + 0.02 \times AOD_F)$	N/A
AOD_C	$\Delta AOD_C = \pm(0.01 + 0.04 \times AOD_C)$	N/A
f_{NCS}	$\Delta f_{CNS} = \pm 25\%$	N/A
AOD_{CS}	$\Delta AOD_{CS} = AOD_{CS} \times [(0.04 + 0.01/AOD_{CNS})^2 + ((1-\Delta f_{CNS})/(1-f_{CNS}))^2]^{1/2}$	N/A
AOD_{CNS}	$\Delta AOD_{CNS} = AOD_{CNS} \times [(0.04 + 0.01/AOD_{CNS})^2 + (\Delta f_{CNS}/f_{CNS})^2]^{1/2}$	N/A

$^{\$}$ Tanré et al. (2011) and references therein

Figure 1. Map of the location of the 17 AERONET ground-based stations considered in this work.

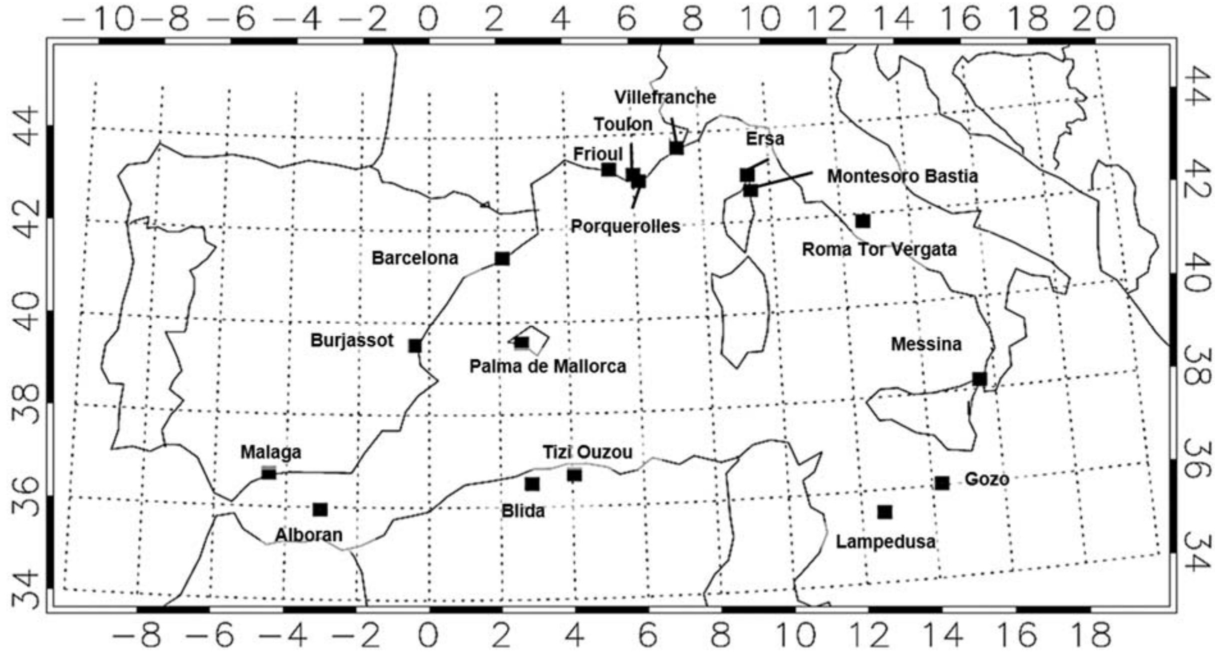
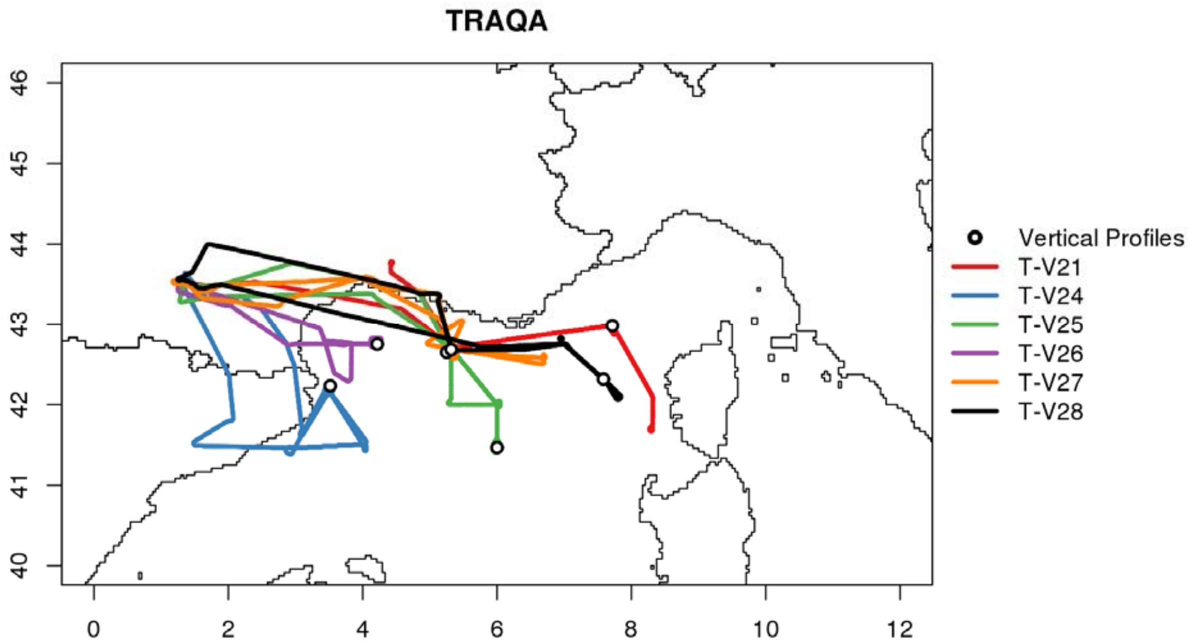


Figure 2. Flight tracks of the ATR-42 aircraft (coloured lines) during the TRAQA and ADRIMED campaigns. Only flights relevant to this study are presented. The location of the profiles coincidental, at their lowermost altitude, with a POLDER-3 overpass is shown by a circle. During the TRAQA campaigns, 7 profiles were retained for comparison on 6 flights. During the ADRIMED campaign, 12 profiles occurring during 9 flights were retained. In this second case, symbols are not always visible due to overlapping.



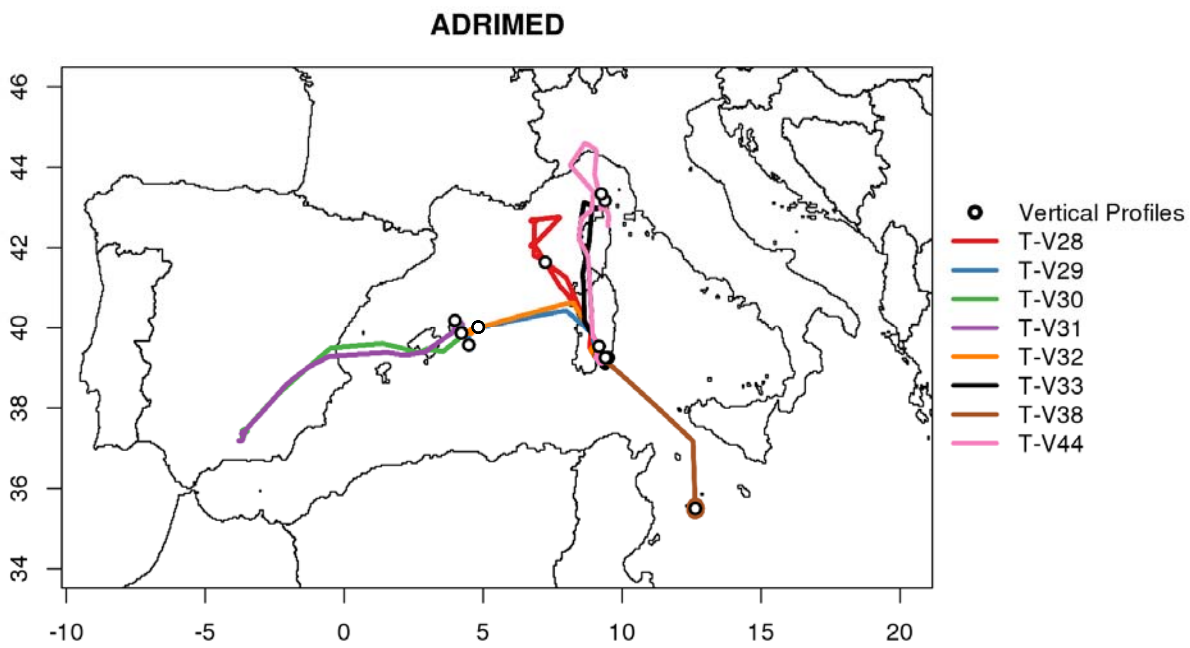


Figure 3. Scatterplots of daily *AOD* retrieved by POLDER-3 at 865 nm with respect to: (top panel) coincident and co-located values from the 17 ground-based AERONET sites at 870 nm; (middle panel) airborne PLASMA sun-photometer operated at 865 nm during ADRIMED; (bottom panel) results of the optical calculations at 865 nm according to Figure 1 from airborne measurements during TRAQA and ADRIMED. The solid line is the bisector. The dashed lines represent the limits indicated by the G_{frac} parameter. The characteristics of the linear correlation (number of points *N*, correlation coefficient *R*, G_{frac}, *RMS* and bias) are also reported.

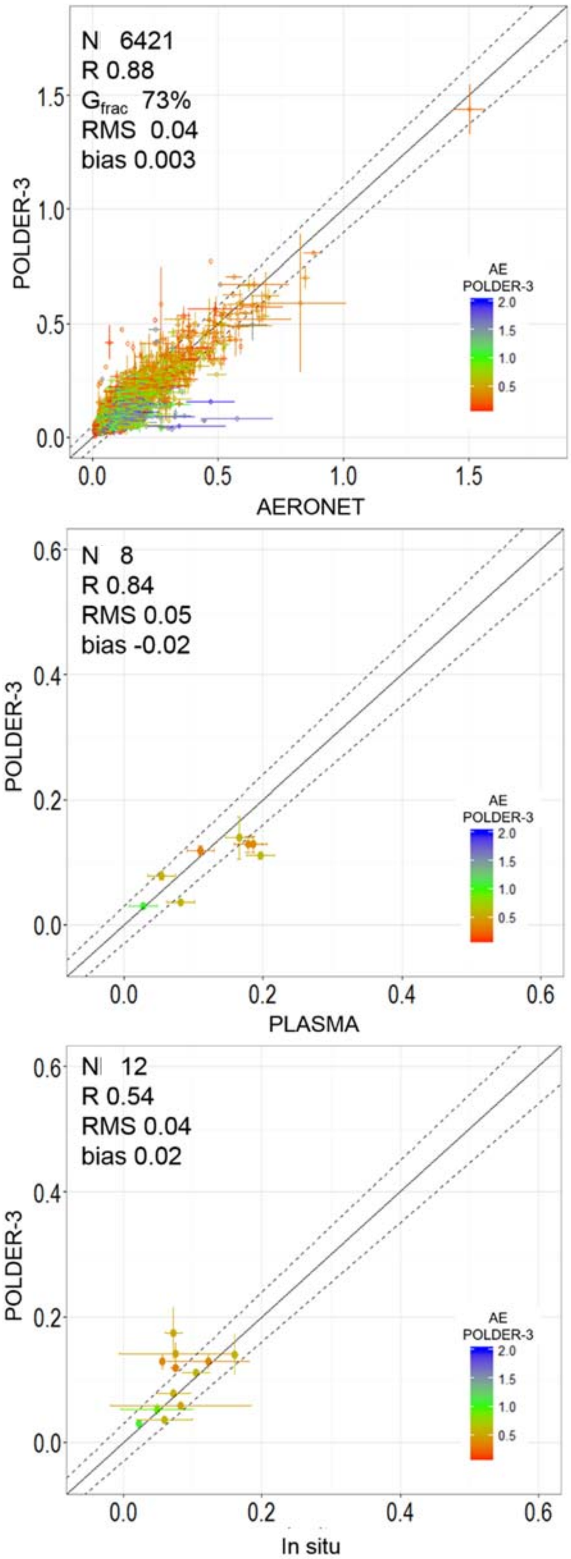


Figure 4. Scatter plots of daily AOD_F and AOD_C retrieved by POLDER-3 at 865 nm as a function of coincident AERONET values at 870 nm for the 17 sites of Western Mediterranean Sea. The solid line is the bisector. The dashed lines represent the limits indicated by the G_{frac} parameter. The characteristics of the linear correlation (number of points N, correlation coefficient R, G_{frac}, RMS and bias) are also reported.Note that, as discussed in sections 2.1 and 2.2, the definitions of AOD_F and AOD_C by POLDER-3 and AERONET are not the same.

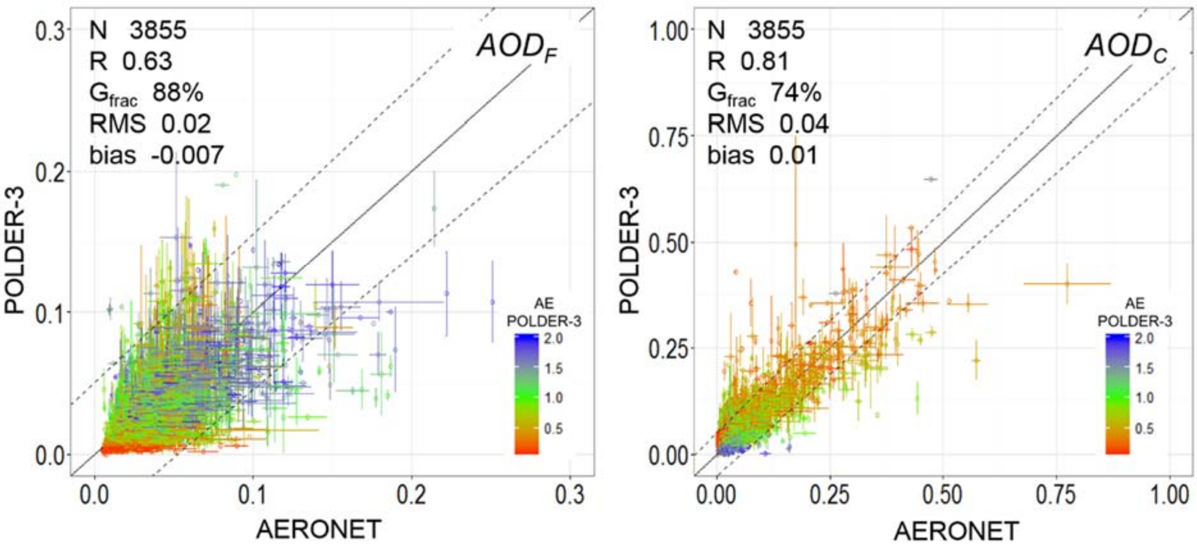
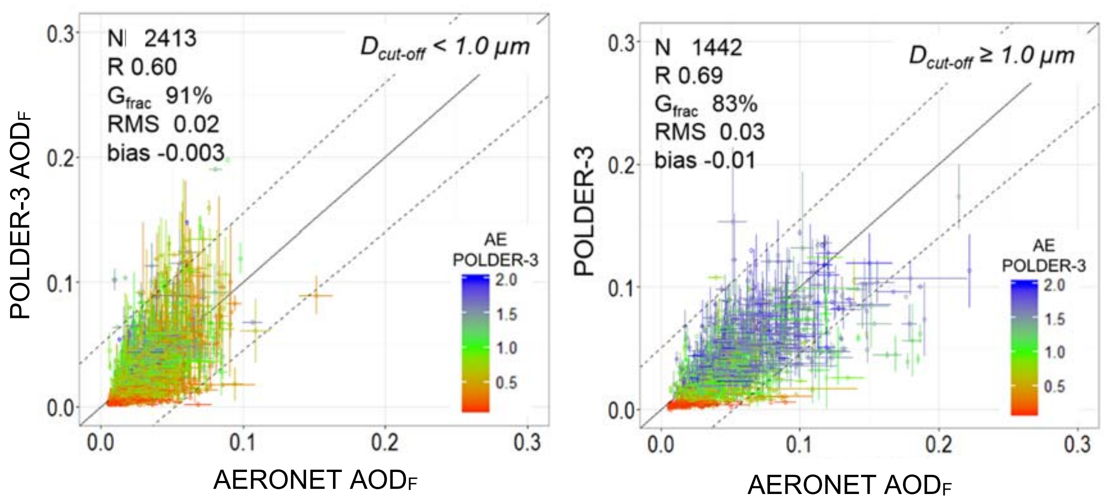


Figure 5. Scatter plots of daily AOD_F (top) and AOD_C (bottom) retrieved by POLDER-3 at 865 nm as function of coincident AERONET values at 870 nm at the 17 sites of Western Mediterranean Sea. The AERONET AOD_F and AOD_C are calculated from the fine and coarse modes of the retrieved volume size distribution defined as below and above a threshold diameter ($D_{cut\text{-}off}$) corresponding to the minimum of the size distribution. The $D_{cut\text{-}off}$ is not fixed but can vary between 0.44 and 0.99 µm. The figure presents cases corresponding to AERONET retrievals yielding a separation of the fine and coarse modes of the volume distribution at $D_{cut\text{-}off}$ <1.0 µm (left) and days with AERONET $D_{cut\text{-}off}$ ≥1.0 µm (right). The solid line is the bisector. The dashed lines represent the limits indicated by the G_{frac} parameter. The characteristics of the linear correlation (number of points N, correlation coefficient R, G_{frac}, RMS and bias) are also reported.

(a) AOD$_F$



(b) AOD$_C$

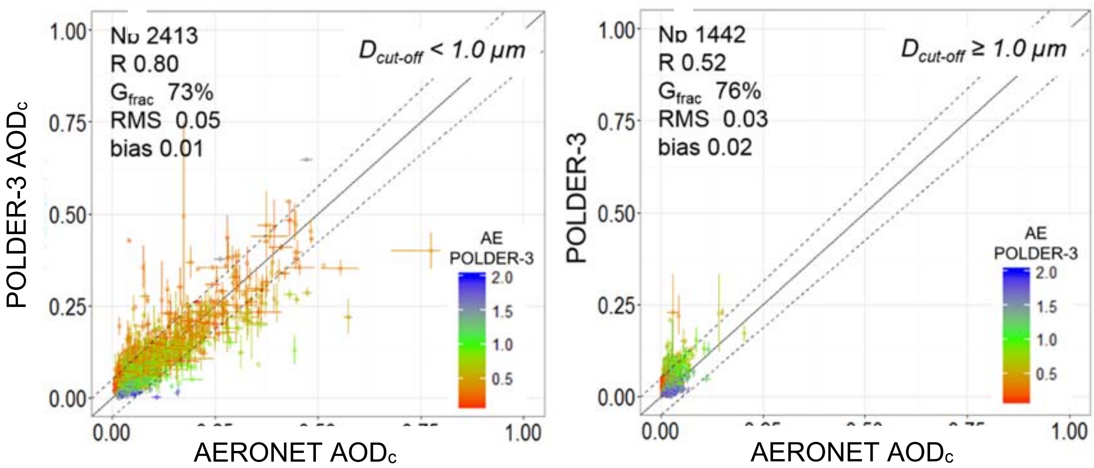


Figure 6. Scatter plots of AOD_F (left) and AOD_C (right) retrieved by POLDER-3 at 865 nm and compared to values obtained by optical calculations from airborne measurements of the number size distribution. Panels, from top to bottom, represent the results of the calculations when varying the cut-off diameter between 0.4 and 1.0 µm. Characteristics of the linear correlation are also reported (number of points *N*, correlation coefficient *R*, *RMS* and bias). Error bars of in situ measurements were calculated from the optical calculation and the instrumental uncertainties. The solid line is the bisector.

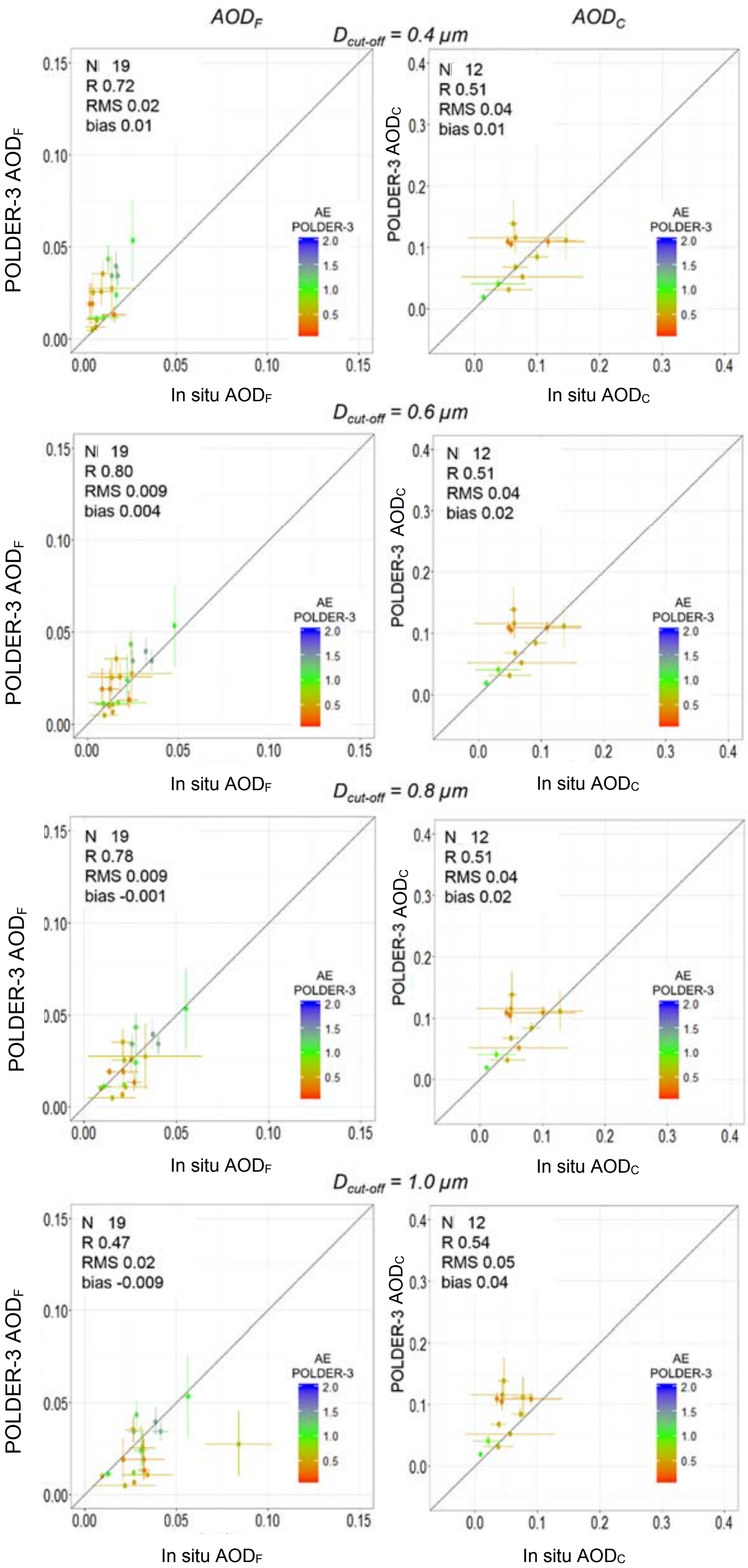


Figure 7. Scatter plots of the Angström Exponent (*AE*) retrieved by POLDER-3 between 865 and 670 nm with respect to coincident and collocated values from (top) the 17 ground-based AERONET sites between 870 and 675 nm; (middle) airborne PLASMA sun-photometer operated at 870 and 675 nm during ADRIMED; (bottom) optical calculations at 865 and 670 nm from number size distributions measured in situ during TRAQA and ADRIMED. Only AERONET values corresponding to POLDER-3 *AOD* larger than 0.1 are considered. To facilitate the reading, the standard deviations of the AERONET values are not represented. Characteristics of the linear correlations are also reported (number of points *N*, correlation coefficient *R*, *RMS* and bias).

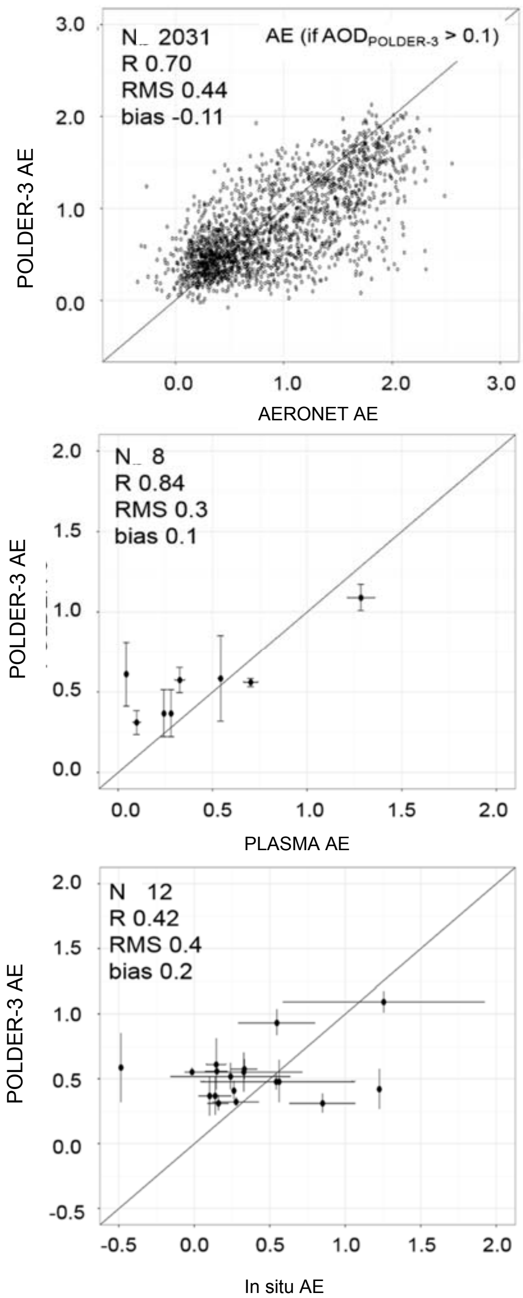


Figure 8. Scatter plot of *AE* versus *AOD* retrieved by POLDER-3 (left) and AERONET (right) on coincidental days (*N* =6421) for the 17 stations of Western Mediterranean Sea. Mean and standard deviations (in brackets) of *AOD* obtained by classifying the air masses into pollution (blue, *AE* ≥1.5), mixed (green, 0.5< *AE* <1.5) and desert dust (orange, *AE* ≤0.5) according to Pace et al. (2006) are shown.

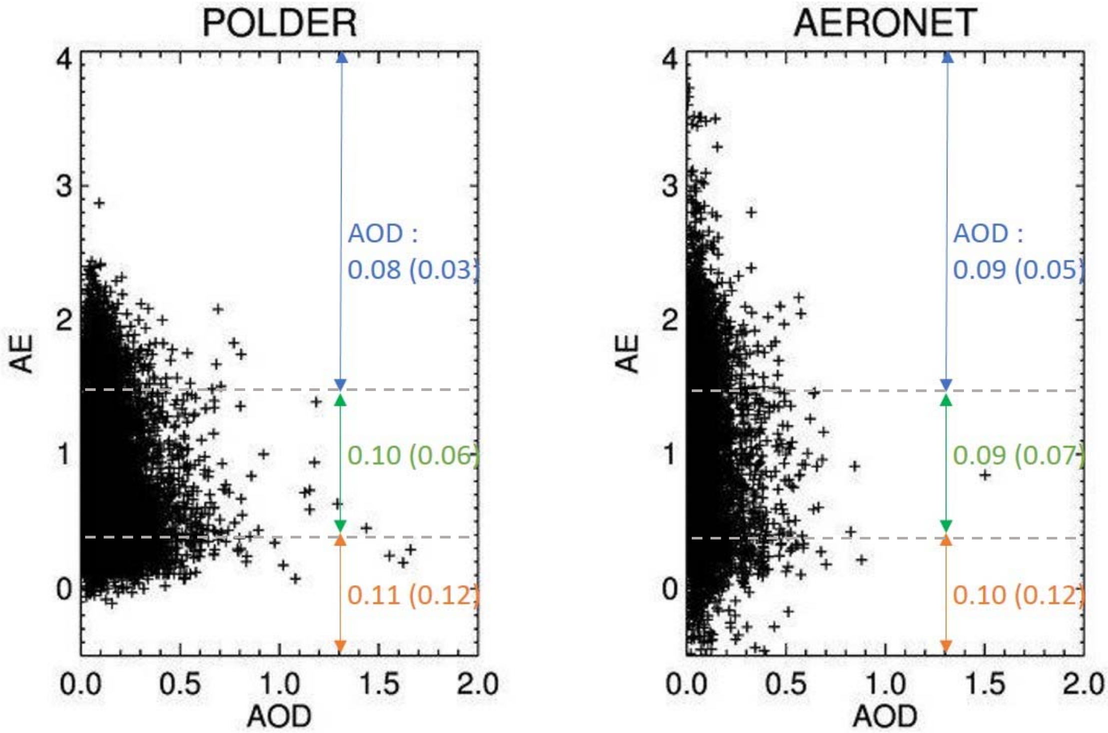
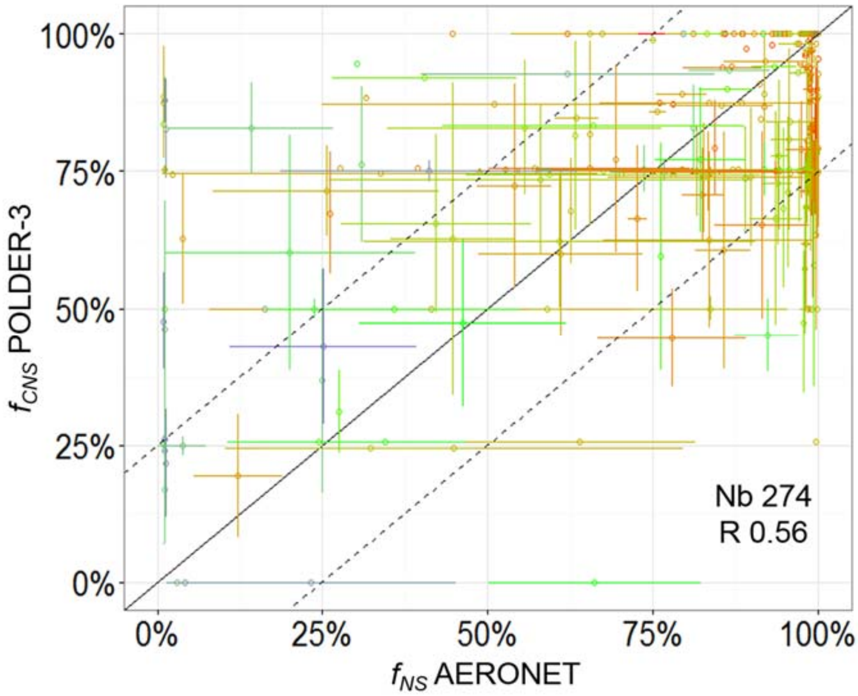
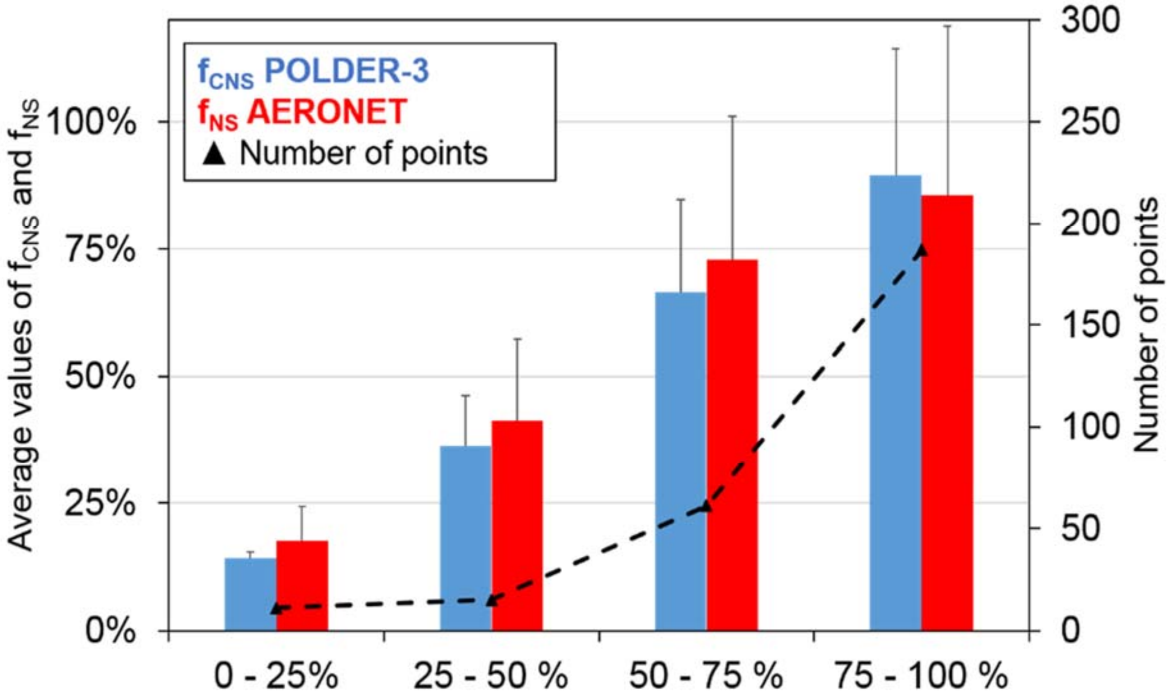


Figure 9. a) Scatterplot of the fraction of coarse mode optical depth due to non-spherical particles (f_{CNS}) retrieved by POLDER-3 and that of total optical depth (f_{NS}) estimated by AERONET. Values are expressed in percent. As the AERONET f_{NS} depends on the total aerosol optical depth, only data points for which the measured *AOD* exceeded 0.10 and the AOD_C represented more than 30% of the total *AOD* are represented. The solid line is the bisector. Dashed lines represent the interval of ±25% of agreement between POLDER-3 f_{CNS} and AERONET f_{NS}. B) Mean and standard deviations of coarse mode optical depth due to non-spherical particles measured by POLDER-3 (f_{CNS}, blue) and that of total optical depth estimated by AERONET (f_{NS}, red) classified into four classes: spherical (f_{CNS} ≤25%); predominant spherical (25%< f_{CNS} ≤50%), predominant non-spherical (50%< f_{CNS} ≤75%); non-spherical (75%< f_{CNS} ≤100%). Values are expressed in percent. Only AERONET data points for which the *AOD* >0.10 and AOD_C/*AOD* >0.30 are represented. The black triangles represent the number of points in each class (the dashed curve is represented for increased readability).





f_CNS POLDER-3
f_NS AERONET
▲ Number of points
Average values of f_CNS and f_NS
Number of points
0%
25%
50%
75%
100%
0
50
100
150
200
250
300
0 - 25%
25 - 50 %
50 - 75 %
75 - 100 %

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Figure 10. Scatterplot of the POLDER-3 coarse non-spherical AOD (AOD_{CNS}) as a function of AERONET non-spherical AOD (AOD_{NS}) at 865 nm for the same data set. The number of points and the regression coefficient R are shown. The solid think line represents the 1:1 line.

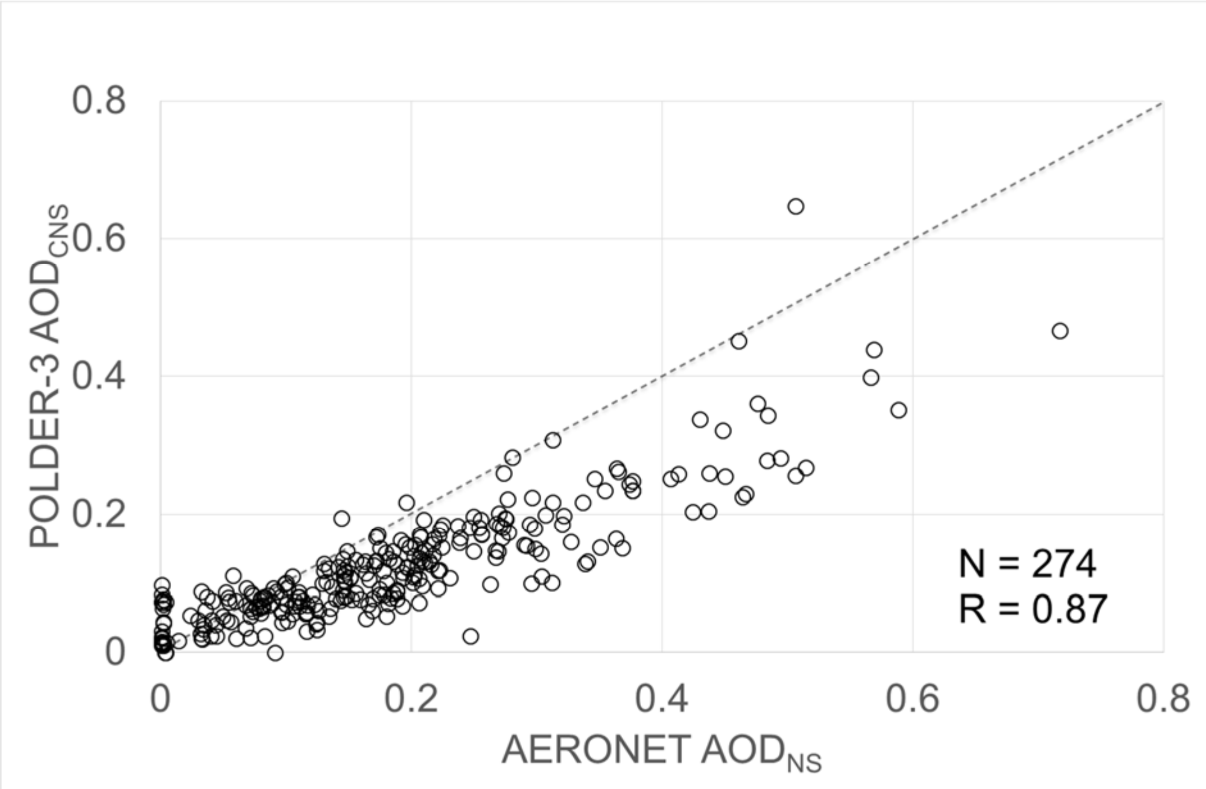

Figure 11. Regional maps for *AOD*, *AE*, AOD_F (top panel from left to right), AOD_C, AOD_{CNS} and AOD_{CS} (bottom panel from left to right) retrieved by POLDER-3 for the period March 2005-October 2013. Mean and standard deviations over the whole marine area of the window are also shown.