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Marine aerosol distribution and variability over the pristine Southern Indian Ocean

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ABSTRACT

This paper presents an 8-year (2005–2012 inclusive) study of the marine aerosol distribution and variability over the Southern Indian Ocean, precisely in the area {10° S– 40°S ; 50° E– 110°E} which has been identified as one of the most pristine regions of the globe. A large dataset consisting of satellite data (POLDER, CALIOP), AERONET measurements at Saint-Denis (French Réunion Island) and model reanalysis (MACC), has been used. In spite of a positive bias of about 0.05 between the *AOD* (aerosol optical depth) given by POLDER and MACC on one hand and the *AOD* measured by AERONET on the other, consistent results for aerosol distribution and variability over the area considered have been obtained. First, aerosols are mainly confined below 2 km asl (above sea level) and are dominated by sea salt, especially in the center of the area of interest, with $AOD \leq 0.1$. This zone is the most pristine and is associated with the position of the Mascarene anticyclone. There, the direct radiative effect is assessed around -9 W m^{-2} at the top of the atmosphere and probability density functions of the *AOD*s are leptokurtic lognormal functions without any significant seasonal variation. It is also suggested that the Madden-Jullian oscillation impacts sea salt emissions in the northern part of the area considered by modifying the state of the ocean surface. Finally, this area is surrounded in the northeast and the southwest by seasonal Australian and South African intrusions ($AOD > 0.1$); throughout the year, the ITCZ seems to limit continental contaminations from Asia. Due to the long period of time considered (almost a decade), this paper completes and strengthens results of studies based on observations performed during previous specific field campaigns.

1. Introduction

Oceans cover about 70 % of the Earth surface and thus sea-air interactions play a key role in the atmospheric system, from local to global scales. In particular, oceans are a reservoir of sea spray aerosols, i.e. sea salt aerosols (SSA) and primary organic matter (e.g. chlorophyll-*a*, see Gantt and Meskhidze (2013)). These are thus the most widely distributed natural aerosols (average total flux sea spray is 4 100 Tg yr^{-1} according to IPCC (2013, p. 596)). SSA consists of seawater drops and dry sea salt particles (ionic species such as sodium, potassium, etc.) whose sizes can extend from 0.05 μm up to 1 mm (e.g. O'Dowd et al., 1997). For detailed information about physical and chemical properties of SSA, as well as the mechanisms of production, see Lewis and Schwartz (2004, and the exhaustive reference list therein). These natural aerosols are of fundamental importance for climate and interrelated topics (IPCC, 2013): direct and indirect radiative forcing, cloud formation and lifetime, atmospheric electrical charge (e.g. Blanchard, 1985), possibly hurricane development (Emanuel, 2003), chemical cycles and health.

Marine environments are also influenced by continental emissions and contain other kinds of both natural and anthropogenic aerosols (Prospero et al., 2002) like dusts and minerals, biological matter, sulfate, nitrate, and organic aerosols (e.g. soot, carbonaceous material). Considered together, all of the aerosols in marine environments are referred as marine aerosols, (see O'Dowd and de Leeuw, 2007; Fitzgerald, 1991). In pristine regions, where land and human activities have little impact, SSA are dominant (50 % – 70 % of the aerosol mass, IPCC (2013, p. 596)). Such regions are interesting for at least two reasons: (1) they can be considered good indicators of the meteorological conditions during the preindustrial epoch, which is a crucial reference point to quantify the contribution of natural emissions to the changing climate. (2) Aerosol concentrations are relatively low and so changes in the aerosol concentration can give rise to unexpected results. For example, Koren et al. (2014) suggested that small changes in SSA

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concentration may have large effects on clouds and on the climate by favoring warm convective cloud invigoration. Pristine regions are located mainly in the Southern Hemisphere (e.g. Hamilton et al., 2014), i.e. the Southern Indian Ocean, South Pacific, and South Atlantic between Brazil and South Africa.

The focus of this paper is on the vast region of the Southern Indian Ocean located between 10°S - 40°S latitude and 50°E - 110°E longitude. To the authors' knowledge, this area has not been examined as comprehensively as others in the Pacific, Atlantic, or other seas, although it is often included in global analyses of SSA characteristics and properties (e.g. Jaeglé et al., 2011).

Various studies have already been devoted to aerosols over the Indian Ocean, especially the northest part, around India. These studies have addressed the impact of anthropogenic and land contamination from Asia on the marine background. In particular these studies have mainly exploited data collected from January to March, in 1998 and 1999, during INDOEX[1] (see http://www-indoex.ucsd.edu/ and references therein for details) which was a field study aimed at understanding the transport of continental aerosols to pristine ocean and their radiative impact. Both observations and models have revealed an abrupt transition between the northest and the southest air masses, close to the ITCZ[2], which has been called the "Chemical ITCZ" (Lawrence et al., 2003) or "Chemical equator" (Hamilton et al., 2008, for the Western Pacific). For example, from shipborne measurments, Moorthy et al. (1999) noted a sharp negative southward *AOD* gradient, across the ITCZ, at shorwaves with no longwave *AOD* variation, suggesting large aerosols of oceanic origin south of the ITCZ. In addition, Norman et al. (2003) have shown strong decreases (up to a factor of 40) in the anthropogenic and continental sources of submicrometer aerosol concentrations. Also, during the 1999 intensive field phase of INDOEX, Welton et al. (2002) measured *AOD* (at 523 nm) of 0.05 ± 0.03 for marine aerosols in the absence of any continental influence with a peak extinction close to the top of the marine boundary layer. In addition, Rajeev et al. (2000) examined the aerosol distribution through AVHRR[3] observation during February-March 1998. They found that *AOD* at 630 nm appears smaller than 0.1 in the Southern Hemisphere Indian Ocean, but the non-SSA component is estimated to exceed the SSA component because of interhemispheric transport, despite the high SSA production associated with high wind speed values around 20°S. Atmospheric models have also been used to describe shortwave *AOD* and aerosol radiative forcing from January to March 1999 (e.g. Rasch et al., 2001; Collins et al., 2002; Reddy et al., 2004). *AOD* values lower than 0.1 were obtained from 10°S to 30°S, with larger values due to Australian dust and SSA contribution to total *AOD* around 40 %− 60 %. Other studies after INDOEX have analyzed the impact of contamination from Asia to the Southern Indian Ocean (e.g. Pant et al., 2009); Lawrence and Lelieveld (2010) reviewed the current understanding of the southern Asian pollutant outflow (see especially sections 4 and 6 in this reference for the Indian Ocean) and encouraged further studies.

As indicated, the studies mentioned above have focused on relatively short and similar periods (at most three months and during the winter monsoon season). In this paper, we present a 8−year (2005–2012 inclusive) study of *AOD* and aerosol distribution and variability (during a year) in the pristine area indicated above. To the authors knowledge, no study of comparable length has been performed over the pristine Indian Ocean region. To that end, several sources of independent data have been used: spaceborne instruments, ground stations, and model reanalysis. A short description of each is given in Section 2. The period chosen (2005–2012) corresponds to that for which all of these sources of data were available together. The spatial distributions and variabilities of the *AOD* and aerosols are presented in Section 3. In Section 4, probability density functions (pdfs) related to *AOD* are computed. The pdfs may be of interest to aerosol/cloud modeling research since they can provide a statistical basis for theoretical analysis and inform numerical models. These results are then used (Section 5) to assess the *DREA* (direct radiative effect of aerosols) at TOA[4]. Finally, conclusions are given in Section 6. A list of the most important acronyms used is given in the Appendix.

2. Description of datasets and intercomparisons

2.1. Satellite data

Spaceborne instruments are used to measure the aerosol optical depth (*AOD*) at regional and synoptic scales. The *AOD* data comes from retrievals from the POLDER-3/PARASOL[5] instrument. POLDER is a wide field of view imaging radiometer which provides measurements of the spectral, polarization and directionality of solar radiation reflected by aerosols and clouds.

Among the Level 2 products, the aerosol optical thickness data correspond physically to *AOD* at a wavelength (in vacuum)$\lambda = 865$ nm. This product is available from March 2005 to October 2013 with a latitude/longitude (lat/lon) resolution of 1/6° × 1/6°. In the present study, this product has been averaged on a daily 0.75° × 0.75° lat/lon grid, in order to have the same gridding as other data fields for comparisons described further below. This change does not modify substantially the number (10^6) of data points over the area considered. This number is high enough to allow the statistical examination to be further presented throughout this paper. Over oceans, surface reflectance is an important source of uncertainty in aerosol remote sensing, but cloud screening is probably the largest one (Myhre et al., 2004, and references therein). The POLDER cloud screening identification procedure is based on reflectance threshold values, pressure and polarization considerations (e.g. Bréon and Colzy, 1999, for details). Overall, the uncertainty in the *AOD* provided in the Level 2 product is about 0.02 over the area of investigation.

In addition to POLDER data, we have also used data from the spacelidar CALIOP/CALIPSO[6] to examine how aerosols are distributed in the vertical. This instrument provides backscattering profiles at $\lambda = 532$ nm and 1064 nm, including parallel and perpendicular polarized signals at 532 nm. Precisely, we used the Level 3 product associated with monthly averaged aerosol extinction profiles at 532 nm over a uniform spatial grid (2° × 5° lat/lon) with a vertical resolution of 30 m (60 m above 8.2 km) (Winker et al., 2010). In this Level 3 product, cloudy pixels are removed after calibration and range registration to avoid cloud contamination of the aerosol data. See Liu et al. (2000, 2004) for the aerosol-cloud discrimination technique.

2.2. Ground data: aeronet/aeroman

AERONET[7] (Holben et al., 1998, 2001) is an international collaborative network of ground-based Sun-Photometers providing high quality aerosol optical property observations over the long term. The AERONET dataset is used as ground validation of some satellite data. Here, the Level 2.0 data, which are pre- and post-field calibrated, cloud-screened, and quality-assured (Dubovik and King, 2000; Dubovik et al., 2002), have been used. Data are ranked by observation time, with a temporal resolution of around 15 min under cloud-free conditions. Daily-averaged data are also available. The estimated uncertainty in

[1] Indian Ocean Experiment.
[2] Inter Tropical Convergence Zone.
[3] Advanced Very High Resolution Radiometer.
[4] Top of the atmosphere.
[5] Polarisation and directionality of the Earth's Reflectances/Polarization and Anisotropy of Reflectances for Atmospheric science coupled with Observation from a Lidar.
[6] Cloud-Aerosol Lidar with Orthogonal Polarization/Cloud-Aerosol Lidar and Infra-red Pathfinder Satellite Observation.
[7] Aerosol Robotic NETwork.