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# Marine and biomass burning aerosols in the southern Indian Ocean: Retrieval of aerosol optical properties from shipborne lidar and Sun photometer measurements

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[1] We document aerosol extinction properties in the southern Indian Ocean. A unique data set of shipborne measurements has been collected with a dual Rayleigh-Mie lidar aboard the research vessel Marion Dufresne during two campaigns: one around Madagascar during the Southern Hemisphere late summer and one close to the Kerguelen Islands during the biomass burning (BB) season. During this latter, a layer containing a mix of BB and marine aerosols extending up to ~3 km above mean sea level (amsl) has been observed from [31°S, 69°E] to [24°S, 59°E]. Both vertical structure and aerosol optical properties have been retrieved from the inversion of the lidar signals. Sun photometer-derived aerosol optical thickness (AOT) at 355 nm is used to constrain the lidar inversion. We obtain a mean integrated value of backscatter-to-extinction ratio (BER) (extinction-to-backscatter ratio, or so-called lidar ratio, LR) of  $0.039 \pm 0.009 \text{ sr}^{-1}$  ( $26 \pm 6 \text{ sr}$ ) and  $0.021 \pm 0.006 \text{ sr}^{-1}$  ( $48 \pm 12 \text{ sr}$ ) for the marine aerosols layer, and for the mixing between BB and marine aerosols with an uncertainty of  $0.009 \text{ sr}^{-1}$  (6 sr) and  $0.004 \text{ sr}^{-1}$  (9 sr), respectively. Lidar calibration is used to inverse data without any simultaneous Sun photometer measurements (as nighttime data), and the temporal evolution of the optical properties and vertical extension of the BB aerosol plume is documented. The presence of BB aerosols is in agreement with Lagrangian model GIRAFE v3 (reGlonal ReAl time Fire plumEs) simulations, which show the South American and Southern African BB origin of the encountered aerosol layer.

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## 1. Introduction

[2] Aerosols are efficient scatterers of solar radiation. Consequently, they often have a large impact on local and regional visibility, contribute to the planetary albedo and affect regional and global climate [Crutzen and Andreae, 1990; Andreae, 1996]. Spatially and temporally resolved information on the atmospheric burden and radiative properties of aerosol is needed to estimate radiative forcing [Intergovernmental Panel on Climate Change, 2007]. Moreover, many of the pyrogenic particles can act as cloud condensation nuclei (CCN) [Rogers et al., 1991] and thereby change the radiative properties of clouds. Local fossil fuel and agricultural biomass burning (BB) cause a high aerosol loading over the Indian Ocean [Lelieveld et al., 2001]. Aerosol loading and extinction properties have been intensely studied over the region of the northern Indian

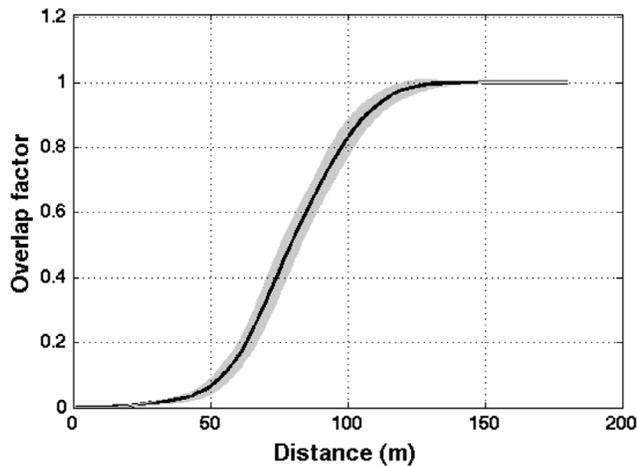
Ocean and Arabian sea in the framework of the INdian Ocean EXperiment (INDOEX) during the winter monsoon [Rajeev et al., 2000; Léon et al., 2001; Pelon et al., 2002; Ramanathan et al., 2001; Rasch et al., 2001; Welton et al., 2002; Chazette, 2003; Forêt et al., 2006].

[3] However, compared to the Northern Hemisphere and to the northern Indian Ocean, the southern Indian Ocean is quite an aerosol-clean area, except during the Southern Hemisphere BB season. The Southern African Regional Science Initiative (SAFARI 2000) experiment showed pronounced smoke and haze exiting off the South-Southeast Africa during the Southern Hemisphere BB season going from July to October. Such plumes are advected over southern Indian Ocean [Annegarn et al., 2002] and can attain Australia [Rosen et al., 2000] and New Zealand [Edwards et al., 2006]. Long-range transports of BB pollution can also occur from South America and Southeast Asia into the southern Indian Ocean during the Southern Hemisphere BB season [Edwards et al., 2006; Duflot et al., 2010]. The resulting aerosol loading causes an increase of the aerosol optical thickness ( $AOT_{\lambda}$ ) and modifies the extinction properties of the low troposphere. Since this seasonal variation of the tropospheric aerosol loading in the southern Indian Ocean

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**Figure 1.** Lidar mean overlap factor  $F$  (black solid line) and its standard deviation (gray shaded area) as a function of the distance from the laser source.

is due to agricultural needs and cultural habits, it occurs every year, and should last still for a long time.

[4] Due to few ground-based measurement points, only big scale means such as MODerate-resolution Imaging Spectroradiometer (MODIS) onboard Terra and Aqua platforms and the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) lidar onboard Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) are available to study the BB aerosol optical properties over the southern Indian Ocean. However, the effect of aerosols on climate strongly depends on their optical properties and tridimensional distribution, and these parameters cannot be easily retrieved from spaceborne instruments, mainly in presence of low AOT, which is often associated with measurements performed with a small signal-to-noise ratio. Ground-based lidars offer the opportunity to measure simultaneously the vertical structure and the extinction coefficient of the atmospheric aerosol layers with a high vertical and temporal resolution, and are complementary to satellite observations.

[5] The data presented here give detailed information on the vertical distribution and optical properties over southern Indian Ocean's marine and BB aerosols. These data have been acquired during two ship-based experiments: one around Madagascar in late austral summer 2009 and another one between La Réunion (21°S, 55°E) and the Kerguelen Islands (50°S, 70°E) in austral winter 2009 during the Southern Hemisphere BB season. In Section 2 the experimental set up involved in this experiment is detailed. The method and the results are presented in Section 3 and discussed in Section 4 taking into account uncertainties on retrievals.

## 2. Data and Numerical Tools

### 2.1. Experimental Set Up and Networks

#### 2.1.1. Rayleigh-Mie Lidar

[6] The Rayleigh-Mie lidar used is a LEOSPHERE ALS450<sup>®</sup> based on a Nd:Yag laser producing pulses with a mean energy of 16 mJ at 355 nm and a frequency of 20 Hz. The system was installed in an air-conditioned box adapted

for use in severe conditions. Lidar measurements have been averaged over 2 min with a vertical resolution of 15 m. The lidar profiles enable to retrieve aerosol optical properties (extinction and backscatter coefficient in synergy with Sun photometer measurements) and atmospheric structures (boundary layer heights, aerosol layers and clouds). It is particularly well-adapted to open ocean experiment and MBL study thanks to its full-overlap height reached at ~120 m (Figure 1). The overlap factor ( $F$ ) has been retrieved using horizontal laser shots both before and after the campaigns as described by Chazette [2003] assuming the atmosphere stable horizontally.

#### 2.1.2. Sun Photometer, AERONET, and Maritime AERONET

[7] AOT measurements were performed in clear-sky condition using a MICROTOSPS II Sun photometer instrument (Solar Light, Inc.). The instrument field of view is about 1°. The AOT ( $AOT_{\lambda}$ ) is measured at five wavelengths ( $\lambda$ ) in the visible spectrum (380, 500, 675, 870 and 1020 nm). The measurements realized with the Sun photometer during the two campaigns are part of the Maritime Aerosol Network (MAN) data. MAN is a component of the Aerosol Robotic Network (AERONET), affiliated with the AERONET calibration and data processing, and deploys ship-based handheld Sun photometers to complement measurements where no islands exist [Smirnov *et al.*, 2009]. The instrument was calibrated at the NASA Goddard Space Flight Center against the AERONET reference CIMEL Sun/sky radiometer. The data presented here have been quality- and cloud-screened following the methodology of Smirnov *et al.* [2000] and the mean uncertainty on the AOT measurements equals 0.015 [Pietras *et al.*, 2002]. The AOT at the lidar wavelength of 355 nm ( $AOT_{355}$ ) was calculated from  $AOT_{380}$  using the Ångström exponent [Ångström, 1964] between 380 and 500 nm. The uncertainty on the retrieved  $AOT_{355}$  has been computed following a similar approach showed by Hamonou *et al.* [1999].

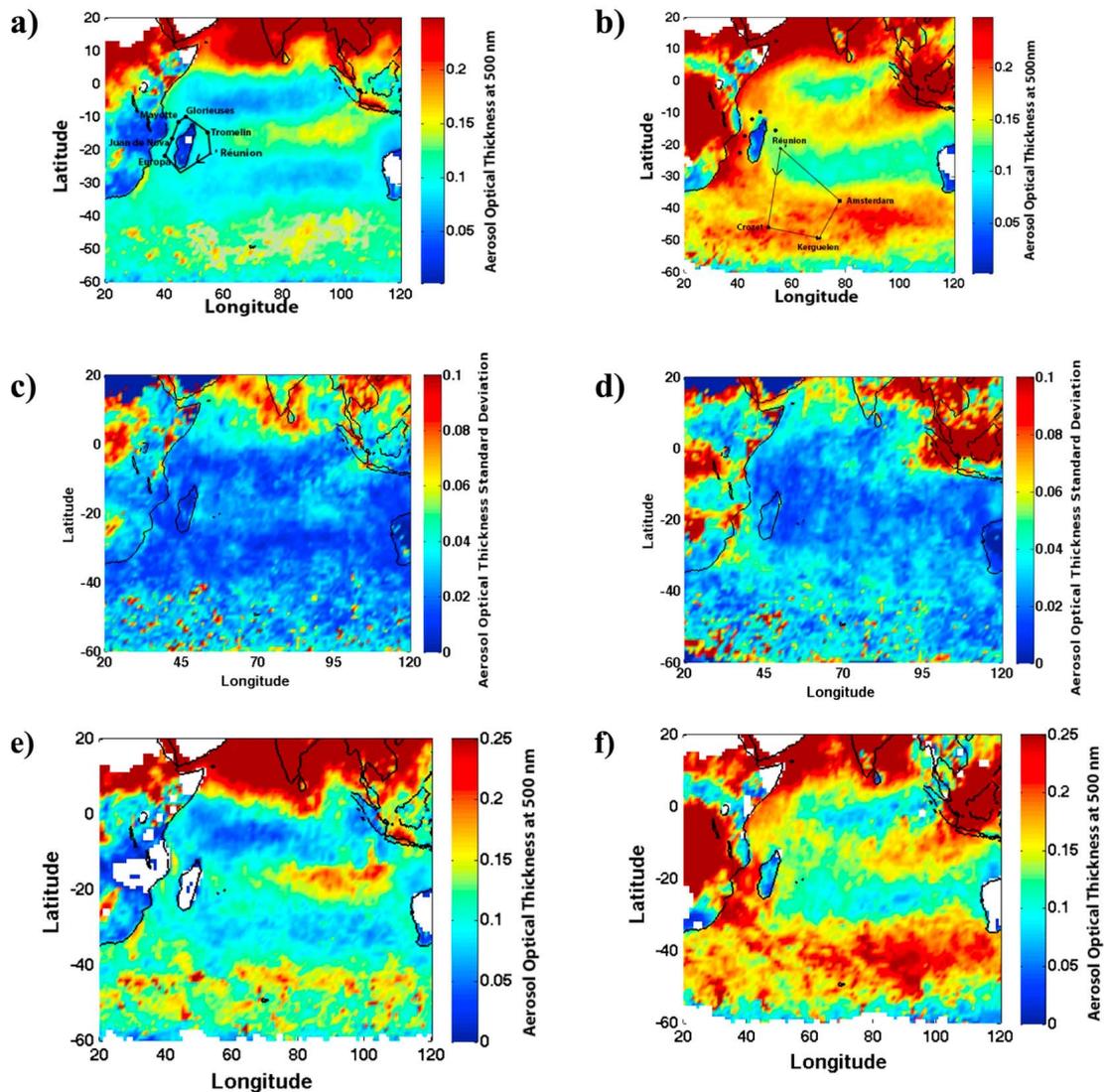
#### 2.1.3. MODIS

[8] The MODIS instrument onboard the Terra and Aqua satellites (<http://modis.gsfc.nasa.gov/>) enables active fire detection based on the high infrared emissions of the fires [Giglio *et al.*, 2003]. This information is available via the FIRMS site (Fire Information for Resource Management System) developed by the University of Maryland (<http://maps.geog.umd.edu/firms/>). FIRMS delivers active fires spots using the MODIS active fire locations processed by the MODIS Rapid Response System using the standard MODIS MOD14 Fire and Thermal Anomalies Product. Each active fire location represents the center of a 1 km pixel that is flagged by the algorithm as containing a fire within the pixel.

[9] We also have used  $AOT_{500}$  as retrieved by MODIS on board the Terra spacecraft with a  $10 \times 10$  km<sup>2</sup> horizontal resolution (at nadir, level 3 data). The retrieval of aerosol properties over both land [Kaufman *et al.*, 1997] and ocean [Tanré *et al.*, 1997] makes use of seven spectral channels in the solar spectrum (0.47–2.1  $\mu$ m).

#### 2.1.4. Ship Plan

[10] Lidar and Sun photometer measurements were performed aboard the French research vessel Marion Dufresne around Madagascar from 19 April to 13 May, 2009, during the so-called Measurement of Aerosols in the Mozambique



**Figure 2.** (a) Monthly mean  $AOT_{500}$  retrieved from MODIS for the month of April over the years 2002 to 2009 and track of the research vessel Marion Dufresne around Madagascar in April–May 2009. (b) Monthly mean  $AOT_{500}$  retrieved from MODIS for the month of September over the years 2002 to 2009 and track of the research vessel Marion Dufresne in August–September 2009. (c) Standard deviation of the monthly mean  $AOT_{500}$  retrieved from MODIS for the month of April over the years 2002 to 2009. (d) Standard deviation of the monthly mean  $AOT_{500}$  retrieved from MODIS for the month of September over the years 2002 to 2009. (e) Monthly mean  $AOT_{500}$  retrieved from MODIS for the month of April 2009. (f) Monthly mean  $AOT_{500}$  retrieved from MODIS for the month of September 2009.

Channel (MACAM0Z) campaign, and from La Réunion (21°S, 55°E) to Kerguelen Islands (50°S, 70°E) from 21 August to 17 September, 2009, during the so-called Kerguelen Aerosols Measurement from African Sources and pUmes Trajectory Reverse Analysis (KAMASUTRA) campaign. Figures 2a and 2b show the tracks of the two campaigns superimposed on the mean AOT at 500 nm retrieved over Indian Ocean from MODIS Aqua in April (Figure 2a) and September (Figure 2b) from 2002 to 2009. Such information gives climatology of the aerosol loading in the area for the two periods. One can see that the aerosol loading in April (Figure 2a) around Madagascar is low ( $AOT_{500} < 0.1$ ); this is not the Southern Hemisphere BB season and the free

troposphere is quite clean of aerosol. The main contribution in the central Indian Ocean is due to emissions from the Indian subcontinent and Southeast Asia with a mean  $AOT_{500} \sim 0.15 \pm 0.04$ .

[11] A more important aerosol loading is noticed between La Réunion and Kerguelen Islands in September (Figure 2b) characterized by higher mean  $AOT_{500} \sim 0.2$  and a larger footprint on the southern part of the Indian Ocean. Note that the contribution of Indonesia is enhanced compared with the previous period. The two plumes are mainly due to the contributions of biomass burning that occur during this season. One can notice the stream of air masses loaded in BB smoke exiting off the South-Southeast Africa toward

south Australia [Annegarn *et al.*, 2002] and the one going from Indonesia-Malaysia to the La Réunion area during the Southern Hemisphere BB season [Duflot *et al.*, 2010].

[12] Figures 2c and 2d show the standard deviation of the monthly mean  $AOT_{500}$  over the years 2002–2009: this gives the temporal variability of the aerosol loading patterns from one year to another for the two periods. The standard deviation of  $AOT_{500}$  along the MACAMOZ campaign's track (Figure 2c) is low ( $<0.03$ ), which suggests that the low aerosol loading in this area in April is quite constant from one year to another. Figure 2e shows that the situation is quite similar to the climatological pattern for April 2009. The standard deviation of  $AOT_{500}$  along the KAMASUTRA campaign's track (Figure 2d) is below 0.05; as the mean  $AOT_{500}$  in the plume exiting off the South-Southeast Africa toward the Southwestern Indian Ocean is larger than 0.15 and can reach 0.3 in the plume center, this suggests that this plume is present every year in September. Figure 2e shows that the situation is very close to the climatological pattern for September 2009.

[13] Consequently, the MACAMOZ campaign around Madagascar in April 2009 was focused on testing the instrumental set up and on retrieving the optical properties of the marine aerosols trapped in the marine boundary layer (MBL) while the KAMASUTRA campaign between La Réunion and Kerguelen Islands in September 2009, taking place during the Southern Hemisphere BB season, was focused on retrieving the optical properties of the BB aerosols.

## 2.2. GIRAFE-FLEXPART

[14] The GIRAFE (reGional ReAl time Fire plumes) v3 model combines the MODIS fire counts system with the FLEXPART 6.2 Lagrangian model: at each fire pixel location detected by MODIS, a number of numerical particles is emitted and advected by the FLEXPART 6.2 code. The  $0.5 \times 0.5$  gridded fire-counts are corrected for spatial variability in the frequency of satellite overpasses and missing observations. FLEXPART version 6.2 is a Lagrangian particle dispersion model [Stohl *et al.*, 2005], which simulates the transport and dispersion of linear tracers and treats advection and turbulent diffusion by processing the trajectories of a multitude of particles. The FLEXPART model was driven by global wind field data from ECMWF, with a horizontal resolution of  $1 \times 1$  degree and 60 vertical levels, and a temporal resolution of 4 h. During the measurement campaign, daily GIRAFE simulations over 5 days in forecast mode were performed in order to anticipate the crossing of a BB plume. The particles were injected above fire pixels for 6 h. As recommended by Dentener *et al.* [2006], particles were emitted up to 2000 m agl (above ground level) for each fire pixel detected by MODIS.

## 3. Data Analysis

[15] In this section, the lidar calibration is first explained (Section 3.1). We then present the method used to inverse the lidar signal using the AOT simultaneously measured by Sun photometer (Section 3.2). Only marine aerosols were encountered during the MACAMOZ campaign around Madagascar during the early winter season. We thus use the acquired data to characterize the MBL and the optical properties (i.e., the vertical profile of aerosol extinction

coefficient and the representative backscatter-to-extinction ratio  $BER_\lambda$  at the wavelength  $\lambda$ , as well as the lidar ratio  $LR_\lambda$  – which is the inverse of the  $BER_\lambda$  of the marine aerosols in this part of the Indian Ocean (Section 3.2.1). A BB aerosol layer was encountered during the KAMASUTRA campaign toward Kerguelen Islands during the Southern Hemisphere BB season. We use the AOT simultaneously measured by Sun photometer to characterize the vertical expansion of the BB aerosol loaded layer, as well as its optical properties (Section 3.2.2.1). In Section 3.2.2.2, we use the instrumental constant computed in Section 3.1 to inverse nighttime lidar profiles (and other times without Sun photometer) and to access the evolution with time of the aerosols optical properties and extinction profiles during the crossing of the encountered BB aerosol plume.

### 3.1. Lidar Calibration

[16] After correction of the sky radiance, the lidar equation gives the range-corrected signal  $S(z)$  for the emitted wavelength of  $\lambda = 355$  nm as a function of the range  $z$ , the total (molecular and aerosol) backscatter  $\beta_{355}(z)$  and extinction coefficients  $\alpha_{355}(z)$  [Measures, 1984]:

$$S(z) = \frac{C_\lambda}{(z_0 - z)^2} \cdot F(z) \cdot \beta_{355}(z) \cdot \exp\left(-\frac{2}{\cos\theta} \cdot \int_{z_0}^z \alpha_{355}(z') \cdot dz'\right) \quad (1)$$

where  $\theta$  is the pointing angle,  $C_\lambda$  is a constant which characterizes the overall efficiency of the lidar system implemented at the altitude  $z_0$  and  $F(z)$  is the overlap factor (see Figure 1). In analog mode the instrumental constant is a function of optical efficiency, reception area, laser energy, amplification and photomultiplier gain.

[17] In order to calibrate the lidar (i.e., compute the instrumental constant  $C_\lambda$ ), we used simultaneously (5 min around the lidar acquisition time) Sun photometer-derived  $AOT_{355}$ . In clear sky condition, above the aerosol layers where only molecular scattering occurs at the altitude  $z_m$ , the lidar equation becomes for a vertically pointing lidar:

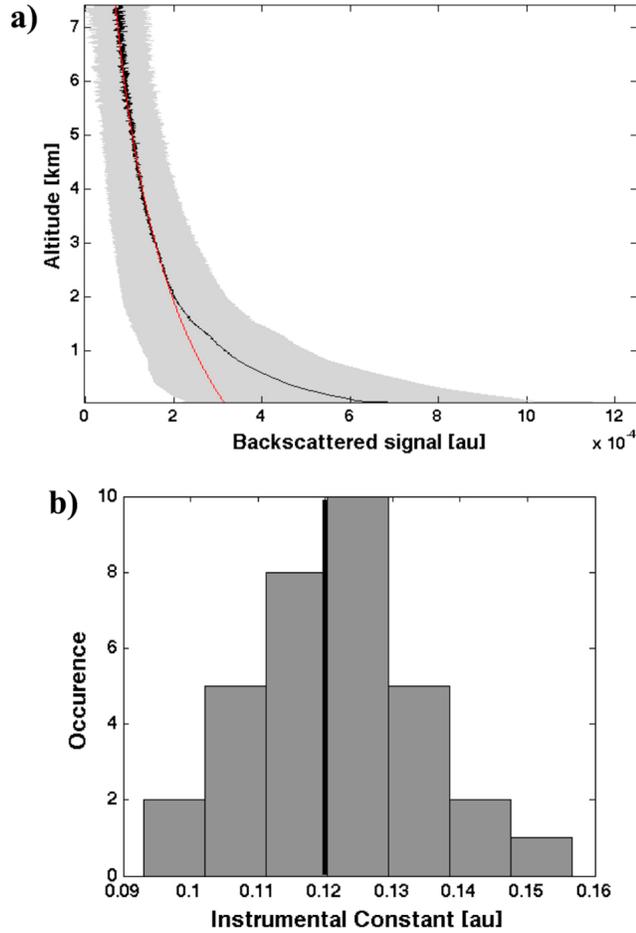
$$S(z_m) = \frac{C_\lambda}{(z_0 - z)^2} \beta_{355}^m(z_m) \cdot \exp\left[-2 \left( \underbrace{\tau_{355}^a(z_0, z_m)}_{AOT_{355}} + \tau_{355}^m(z_0, z_m) \right)\right] \quad (2)$$

where  $\beta_{355}^m(z_m)$  is the molecular backscatter and  $\tau^m(z_0, z_m)$  the optical thickness at 355 nm between  $z_0$  and  $z_m$  which are determined from climatological radiosounding profiles (pressure, temperature) using the polynomial approximation proposed by Nicolet [1984].

[18] Then,  $C_\lambda$  is given against the Sun photometer measurements by:

$$C_\lambda = \frac{(z_0 - z)^2}{\beta_{355}^m(z_m)} S(z_m) \cdot \exp[2(AOT_{355} + \tau_{355}^m(z_0, z_m))] \quad (3)$$

Figure 3a shows the mean vertical profile of the apparent backscatter coefficient derived from the lidar signals used for the calibration superimposed on the simulated pure molecular backscatter coefficient. The altitude reference at which only molecular scattering occurs was taken between 3 and 4 km.



**Figure 3.** (a) Mean (black line) and standard deviation (gray area) of the vertical profile of the apparent backscatter coefficient used for the lidar calibration. The red line shows the simulated pure molecular scattering signal used for the calibration. (b) Histogram of the retrieved  $C_\lambda$ . The mean  $C_\lambda$  (0.12 [au]) is given by the black vertical bar.

[19] Figure 3b shows the histogram of the retrieved  $C_\lambda$ . The mean  $C_\lambda$  is 0.12 [au] with a standard deviation of 0.014 [au]. The mean  $C_\lambda$  will be used in Section 3.2.2.2 to inverse lidar signals without any simultaneous Sun photometer measurements.

### 3.2. Lidar-Derived Aerosol Optical Properties

[20] In this section, Sun photometer measurements are used to constrain the lidar inversion [Chazette, 2003]. The backscatter lidar equation is underdetermined due to its dependence on two unknowns: aerosol backscatter (or extinction) coefficient and  $BER_{355}$  [Klett, 1985; Chazette, 2003]:

$$\beta_{355}^m(z) = \frac{(z - z_0)^2 \cdot S(z) \cdot Q(z)}{(z_r - z_0)^2 \cdot \frac{S(z_r)}{\beta_{355}^m(z_r)} + \frac{2}{BER_{355}} \cdot \int_z^{z_r} (z' - z_0)^2 \cdot S(z') \cdot Q(z') \cdot dz'}$$

$$\text{with } Q(z) = \exp \left[ 2 \cdot \left( \frac{3}{8\pi \cdot BER_{355}} - 1 \right) \cdot \int_z^{z_r} \alpha_{355}^m(z') \cdot dz' \right] \quad (4)$$

where  $z_r$  is the reference altitude corresponding to the integration constant of the Bernoulli differential equation.

[21] Hence, lidar data are inverted to retrieve both the vertical profile of the aerosol extinction coefficient and  $BER_{355}$  using the Bernoulli's differential form of the propagation equation [Klett, 1985] and a dichotomous approach where  $BER_{355}$  is increased (decreased) if the lidar-derived optical thickness is larger (lower) than the Sun photometer-derived  $AOT_{355}$  simultaneously acquired. The dichotomous approach is stopped when the difference between estimated and calculated optical thickness becomes less than the error on  $AOT_{355}$ . It is noteworthy that this method gives access to a height-independent  $BER_{355}$  value.

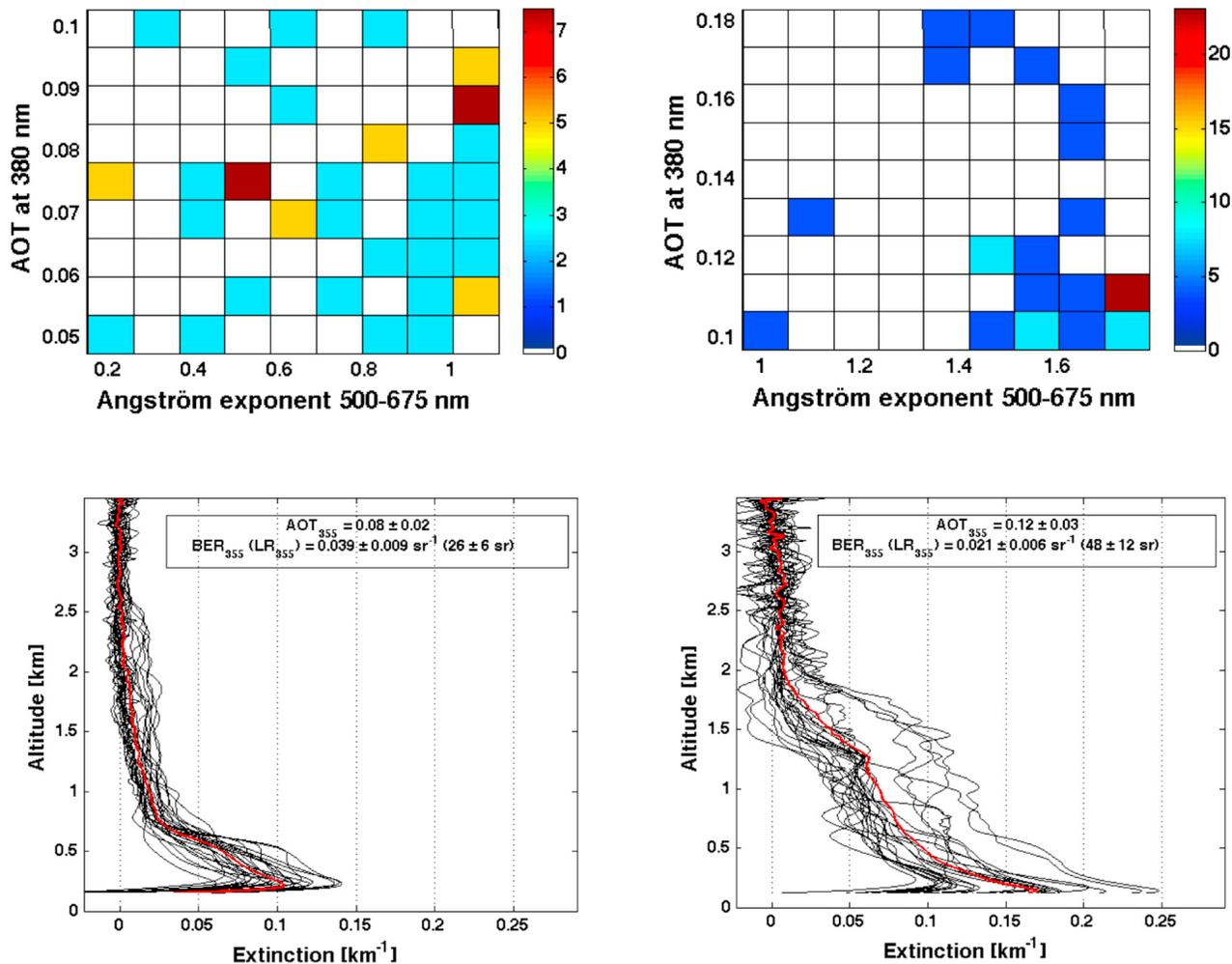
[22] Hereafter, we investigate the data acquired during the MACAMAZ and KAMASUTRA campaigns to document the optical properties of the marine and BB aerosols. We used two criteria to discriminate the cases containing only marine aerosols from the cases containing marine and another type of aerosols: 1) the shape of each lidar range-corrected signal (corrected from the sky background and the solid angle) is compared to the shape of a simulated signal assuming only Rayleigh backscattering; 2) the simultaneously (5 min around the lidar acquisition time) acquired Sun photometer-derived  $AOT_{355}$  is compared to a threshold equal to 0.10. If the Sun photometer-derived  $AOT_{355}$  is below this threshold and if the shape of the simultaneously acquired lidar signal matches the shape of the pure Rayleigh simulated signal except in the MBL (supposed to be less than 1 km thick), then the case is classified as containing only marine aerosols. Using this approach, it appears that no BB aerosols events have been detected during the MACAMAZ campaign, and that a BB aerosols plume was crossed during the KAMASUTRA campaign. Thus, we use the data acquired during the MACAMAZ campaign to document only the optical properties of the marine aerosols (Section 3.2.1), and we use the data acquired during the KAMASUTRA campaign to document the optical properties of the BB and marine aerosols (Section 3.2.2).

#### 3.2.1. MACAMAZ Campaign: Optical Properties of the Marine Aerosols

[23] Figure 4 (top left) shows the synthesis of the Sun photometer measurements during the whole MACAMAZ campaign. We use two independent variables:  $AOT_{380}$  and Ångström exponent ( $a$ ) between 500 and 675 nm. Mean  $AOT_{380}$  is 0.08 and mean  $a$  is 0.63. One can notice that all  $AOT_{380}$  are below 0.1 and most  $a$  are between 0.2 and 1, which correspond to typical values of optical thickness and Ångström exponent for marine aerosols [Smirnov *et al.*, 1995] in open ocean. Despite the occurrence of some cases with  $a > 1$  - which could correspond to BB aerosols events - no such event has been encountered using the approach described here above.

[24] Figure 5a shows the histogram of the  $BER_{355}$  ( $LR_{355}$ ) retrieved during the MACAMAZ campaign for the whole troposphere. The mean  $BER_{355}$  ( $LR_{355}$ ) is  $0.039 \text{ sr}^{-1}$  (26 sr) with a variability of  $0.009 \text{ sr}^{-1}$  (6 sr). The sources of uncertainties are discussed in section 4.2.

[25] The mean value of  $BER_{355} = 0.039 \text{ sr}^{-1}$  (mean  $LR_{355} = 26 \text{ sr}$ ) is used to assess the vertical profile of aerosol extinction coefficient in the MBL during the campaign (Figure 4, bottom left). No extinction is given below the full-overlap height (120 m, Figure 1). One can observe that the MBL aerosol layer extends up to  $\sim 0.7 \text{ km}$  above mean sea level (amsl). This result agrees with the works of Flamant



**Figure 4.** (left) MACAMOZ campaign; (right) KAMASUTRA campaign. (top) Cross histogram of the AOT at 380 nm and the Ångström exponent between 500 and 675 nm (relative occurrence in [%], 26 measurements for each campaign). (bottom) Aerosol extinction coefficient profiles (black) and mean aerosol extinction coefficient profile (red).

*et al.* [1998], *Russell et al.* [1998], and *Welton et al.* [2002] showing a MBL height in open ocean between  $\sim 0.45$  and  $\sim 1$  km.

### 3.2.2. KAMASUTRA Campaign: Optical Properties of the Biomass Burning Aerosols

[26] The KAMASUTRA campaign was characterized by almost steady cloudy conditions. However, using the same approach as described here above (Section 3.2), a BB aerosols plume event was identified during the campaign between 14th and 16th September from (31°S, 69°E) to (24°S, 59°E). We focus in this section on the data collected during the crossing of this BB plume. We use two different methods to constrain the lidar data inversion: we first use simultaneously acquired Sun photometer measurements (Section 3.2.2.1) and we then use calibration values to derive AOT at times without Sun photometer, such as nighttime (Section 3.2.2.2).

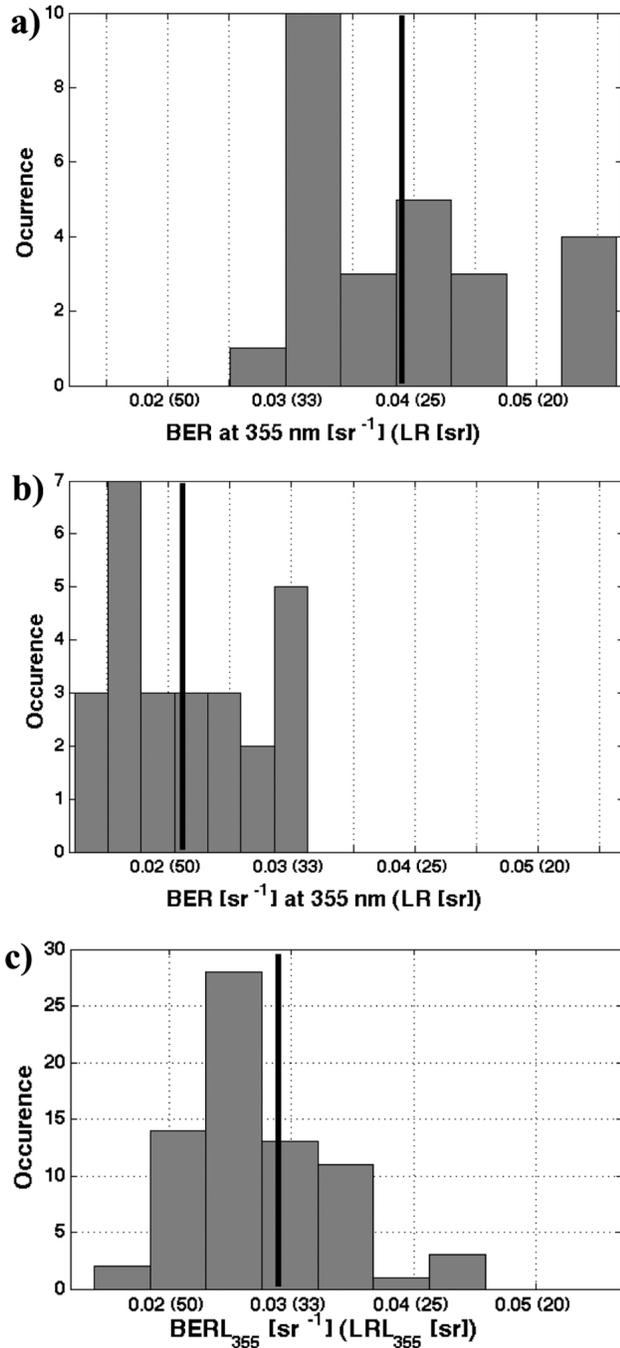
[27] We assume hereafter that the marine aerosols are mixed with the BB aerosols above, i.e., that the MBL is not isolated from the free troposphere. The validity of this

hypothesis depends on the amount of turbulence at the layer interface [*Russell et al.*, 1998]. As we do not have data giving information on the (un)stability of the interface MBL-free troposphere, we chose to consider the most observed case in open ocean: the mixing between the two layers [*Russell et al.*, 1998; *Rasch et al.*, 2001; *Bates et al.*, 2002; *Welton et al.*, 2002; *Forêt et al.*, 2006]. Thus, we consider in the following a single aerosol layer containing mixed marine and BB aerosols.

#### 3.2.2.1. Using Sun Photometer Measurements

[28] Figure 5 (top right) shows the synthesis of the Sun photometer measurements during the BB aerosols plume event using  $AOT_{380}$  and  $a$  as two independent variables. Over this period, the vessel track crossed a BB aerosol plume associated with  $AOT_{380} = 0.12 \pm 0.03$  and  $a = 1.52 \pm 0.21$ .

[29] Figure 5b shows the histogram of the  $BER_{355} (LR_{355})$  retrieved during the crossing of the BB aerosol plume (14–16 September) using the Sun photometer-derived  $AOT_{355}$  as a constraint for the inversion. A mean  $BER_{355} (LR_{355})$  value of



**Figure 5.** (a) Histogram of the  $BER_{355}$  ( $LR_{355}$ ) retrieved during the MACAMOZ campaign. The mean  $BER_{355}$  ( $0.039 \text{ sr}^{-1}$ ) (mean  $LR_{355} = 26 \text{ sr}$ ) is given by the black vertical bar. (b) Histogram of the  $BER_{355}$  retrieved during the crossing of the BB aerosol plume (14–16 September – KAMASUTRA campaign). The mean  $BER_{355}$  ( $0.021 \text{ sr}^{-1}$ ) (mean  $LR_{355} = 48 \text{ sr}$ ) is given by the black vertical bar. (c) Histogram of the  $BERL_{355}$  ( $LRL_{355}$ ) retrieved during the crossing of the BB plume (14–16 September – KAMASUTRA campaign). The mean  $BERL_{355}$  ( $0.029 \text{ sr}^{-1}$ ) (mean  $LRL_{355} = 34 \text{ sr}$ ) is given by the black vertical bar.

$0.021 \text{ sr}^{-1}$  (48 sr) with a variability of  $0.006 \text{ sr}^{-1}$  (12 sr) has been calculated for the whole troposphere. As previously, the sources of uncertainties are discussed in section 4.2. This value is significantly different from the previous  $BER_{355}$  ( $LR_{355}$ ) value determined during MACAMOZ campaign for marine aerosols ( $BER_{355} = 0.039 \pm 0.009 \text{ sr}^{-1}$  or  $LR_{355} = 26 \pm 6 \text{ sr}$ ). Comparing Figures 5a and 5b, one can see that the distributions of retrieved  $BER_{355}$  ( $LR_{355}$ ) values are different, with little to no overlap between the two distributions.

[30] The mean value of  $BER_{355} = 0.021 \text{ sr}^{-1}$  ( $LR_{355} = 41 \text{ sr}$ ) is used to assess the vertical profile of aerosol extinction coefficient during the crossing of the BB aerosol plume (14–16 September). The mean aerosol extinction coefficient profile observed during the BB aerosol event is given in Figure 5 (bottom right). No extinction is given below the full-overlap height (120 m, Figure 1). One can observe that the marine+BB aerosol (MIX aerosol) layer extends up to  $\sim 2 \text{ km}$  amsl. This result is in accordance with the results of Ramanathan *et al.* [2001] showing that aerosol layers can extend as high as 3 km above the Indian Ocean.

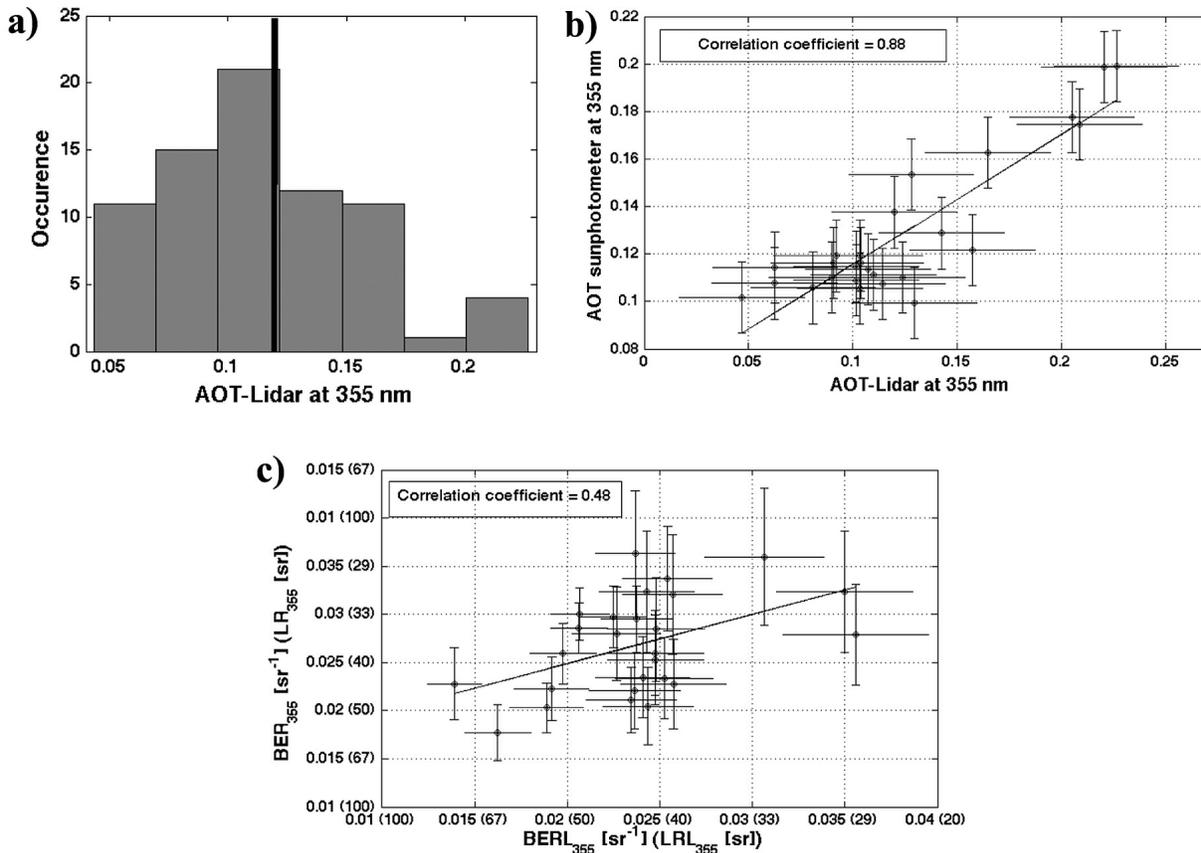
### 3.2.2.2. Using Calibration Values to Derive AOT

[31] In this section, lidar data are inverted using the same dichotomous approach as in the previous section, except that the constraining  $AOT_{355}$  is not the Sun photometer-derived AOT acquired simultaneously with the lidar signal, but the AOT-lidar ( $AOTL_{355}$ ) retrieved via lidar calibration (Section 3.1):

$$AOTL_{355} = \frac{1}{2} \left[ \ln \frac{C_\lambda \cdot \beta_{355}^m(z_m)}{S(z_m) \cdot (z_0 - z)^2} \right] - \tau_{355}^m(z_m) \quad (5)$$

We still focus here on the section of the track crossing the BB aerosol plume. Figure 6a gives the histogram of the retrieved  $AOTL_{355}$ . A mean  $AOTL_{355}$  value of 0.12 with a variability of 0.05 has been calculated. Figure 6b shows the correlation plot between  $AOTL_{355}$  and  $AOT_{355}$ . The correlation coefficient equals 0.88 (26 points), which shows that the two approaches give consistent results. Moreover, whereas Figure 6b shows some discrepancies between  $AOTL_{355}$  and  $AOT_{355}$  values due to uncertainties on  $C_\lambda$  (see Section 4.2 for discussion on uncertainties), the mean  $AOTL_{355}$  value equals the mean  $AOT_{355}$  one (0.12).

[32] Derived  $AOTL_{355}$  values allow us to inverse lidar data without any simultaneous Sun photometer measurements, especially lidar data acquired during nighttime. Such an extrapolation to nighttime measurements is possible because the lidar is implemented in an air-conditioned box, which avoids drifts of laser energy due to variation of temperature. We can thus reasonably suppose that the calibration constant does not change between daytime and nighttime measurements. Figure 5c shows the histogram of the  $BERL_{355}$  ( $LRL_{355}$ ) calculated with the  $AOTL_{355}$  retrieved during the 14–16 September period.  $BERL_{355}$  ( $LRL_{355}$ ) values have been averaged over ten values in order to reduce the statistical error on the retrieved  $BERL_{355}$  ( $LRL_{355}$ ) (Section 4.2), which gives one  $BERL_{355}$  ( $LRL_{355}$ ) value every  $\sim 20 \text{ min}$ . Cloudy profiles were removed from the data set. For 80% of the remaining profiles, the procedure has been convergent and a mean  $BERL_{355}$  ( $LRL_{355}$ ) value of  $0.029 \text{ sr}^{-1}$  (34 sr) with a variability of  $0.006 \text{ sr}^{-1}$  (7 sr) has been calculated for the whole troposphere. Such a value is within the error bar of the



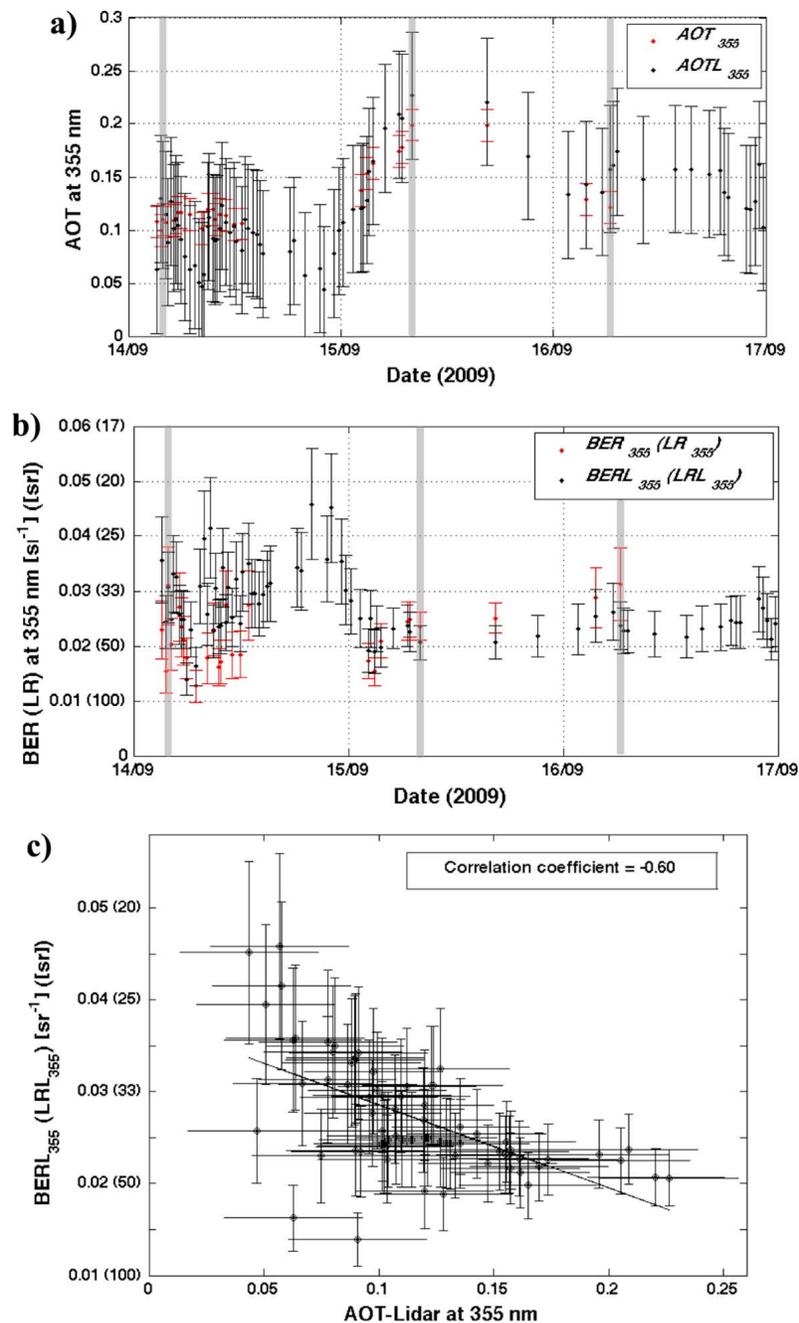
**Figure 6.** (a) Histogram of the  $AOTL_{355}$  derived from lidar measurements and calibration. The mean  $AOTL_{355}$  (0.12) is given by the black vertical bar. (b) Correlation plot between the  $AOTL_{355}$  derived from lidar calibration values versus the  $AOT_{355}$  retrieved from Sun photometer measurements. The correlation coefficient equals 0.88 (26 points). (c) Correlation plot between  $BERL_{355}$  ( $LRL_{355}$ ) versus the  $BER_{355}$  ( $LR_{355}$ ). The correlation coefficient equals 0.48 (26 points). All data were acquired during the crossing of the BB aerosol plume (14–16 September). See Section 4.2 for error bars calculation.

one deduced from the synergy between lidar and Sun photometer (see Section 4.2 for uncertainties calculation). However, the correlation coefficient between  $BER_{355}$  ( $LR_{355}$ ) and  $BERL_{355}$  ( $LRL_{355}$ ) is 0.48 (26 points, Figure 6c), which is lower than the one between  $AOTL_{355}$  and  $AOT_{355}$  (0.88). This can be explained by the high sensitivity of the BER retrieval technique to the value of the AOT used to constrain the inversion (see Section 4.2). Investigating the distribution of the retrieved  $BERL_{355}$  ( $LRL_{355}$ ) (Figure 5c), one can notice some cases with  $BERL_{355} > 0.030$  ( $LRL_{355} < 33$ ), which implies that the crossed plume is not uniformly loaded in aerosols. These high (low) values of  $BERL_{355}$  ( $LRL_{355}$ ) may also result from underestimated  $AOTL_{355}$  (Figure 6b).

[33] Figure 7a shows the evolution with time of  $AOT_{355}$  and  $AOTL_{355}$  during the crossing of the BB plume (values of error bars are explained in Section 4.2). One can see that  $AOT_{355}$  and  $AOTL_{355}$  values are within each other's error bars.  $AOTL_{355}$  values stay between 0.05 and 0.13 on the 14/09, increase to 0.2 on the 15/09 and stay around 0.15 on the 16/09. Figure 7b shows the evolution with time of the retrieved  $BER_{355}$  ( $LR_{355}$ ) and  $BERL_{355}$  ( $LRL_{355}$ ) during the crossing of the BB plume (see Section 4.2 for the error bars calculation).

One can see that  $BER_{355}$  ( $LR_{355}$ ) and  $BERL_{355}$  ( $LRL_{355}$ ) values are within each other's error bars.  $BERL_{355}$  ( $LRL_{355}$ ) evolves mostly between  $\sim 0.025$  and  $\sim 0.045$   $\text{sr}^{-1}$  ( $\sim 22$ – $40$  sr) on the 14/09, decreases (increases) to  $\sim 0.02$   $\text{sr}^{-1}$  ( $\sim 50$  sr) on the 15/09, and finally stays between  $\sim 0.02$  and  $\sim 0.03$   $\text{sr}^{-1}$  ( $\sim 33$ – $50$  sr) on the 16/09. It seems therefore that the part of the plume crossed on the 15/09 is the one containing the more BB aerosols (highest  $AOTL_{355}$  and lowest  $BERL_{355}$ ). One can also note that  $BERL_{355}$  ( $LRL_{355}$ ) decreases (increases) while  $AOTL_{355}$  increases: the correlation coefficient between  $BERL_{355}$  and  $AOTL_{355}$  is  $-0.60$  (72 points, Figure 7c). This suggests that the increase of the optical thickness is mainly due to the increase of the BB aerosol concentration.

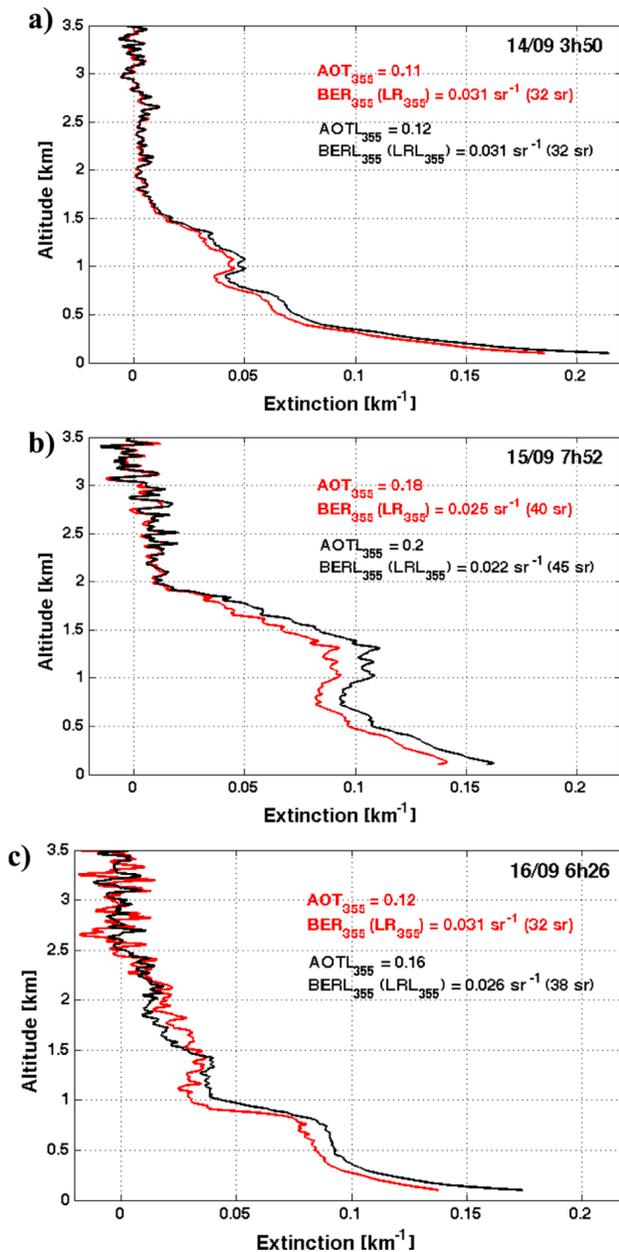
[34] Three extinction profiles (one per day) have been selected and are shown in Figure 8 (no extinction is given below the full-overlap height of 120 m as shown in Figure 1). We chose extinction profiles coinciding with Sun photometer measurements to compare the results obtained from the two retrieval methods (Sun photometer measurements versus calibration values). The related time,  $AOTL_{355}$ ,  $AOT_{355}$ ,  $BERL_{355}$  and  $BER_{355}$  are shown for each profile. One can first



**Figure 7.** (a) Temporal evolution of the retrieved  $AOT_{355}$  and  $AOTL_{355}$  during the crossing of the BB plume. The error bars are also given. (b) Temporal evolution of the retrieved  $BER_{355}$  and  $BERL_{355}$  during the crossing of the BB plume. The gray bars stand for the extinction profiles shown in Figure 8. (c) Correlation plot between the  $BERL_{355}$  ( $LRL_{355}$ ) versus the  $AOTL_{355}$ . The correlation coefficient equals  $-0.60$  (72 points). See Section 4.2 for error bars calculation.

notice that the shapes of the extinction profiles retrieved using the calibration values to derive the  $AOTL_{355}$  are in quite good accordance with the shape of the extinction profiles retrieved using Sun photometer measurements ( $AOT_{355}$ ). However, the selected profiles show a slightly higher extinction when retrieved via  $AOTL_{355}$ . This can be explained by the fact that the related  $AOTL_{355}$  are slightly higher than the related  $AOT_{355}$  for the selected cases.

[35] The height of the aerosol layer is  $\sim 1.8$  km on the 14/09 profile (Figure 8a),  $\sim 3$  km on the 15/09 profile (Figure 8b) and  $\sim 2.5$  km on the 16/09 profile (Figure 8c). This result is also in accordance with the results of Ramanathan *et al.* [2001] showing that aerosol layers can extend as high as 3 km above the Indian Ocean. The 15/09 profile (Figure 8b) shows an enhanced aerosol layer between  $\sim 0.7$  and  $\sim 1.4$  km amsl, which could be related to the highest  $AOTL_{355}$  and



**Figure 8.** Aerosol extinction coefficient profiles retrieved from lidar and Sun photometer measurements (red) and aerosol extinction coefficient profiles retrieved from lidar measurements using calibration values to derive  $AOTL_{355}$  (black) on (a) 14/09 3h50, (b) 15/09 7h52, and (c) 16/09 6h26. Times are Universal Time (UT).

lowest (highest)  $BERL_{355}$  ( $LRL_{355}$ ) observed on the 15/09 (Figure 7). This layer is therefore likely to contain mostly BB aerosols.

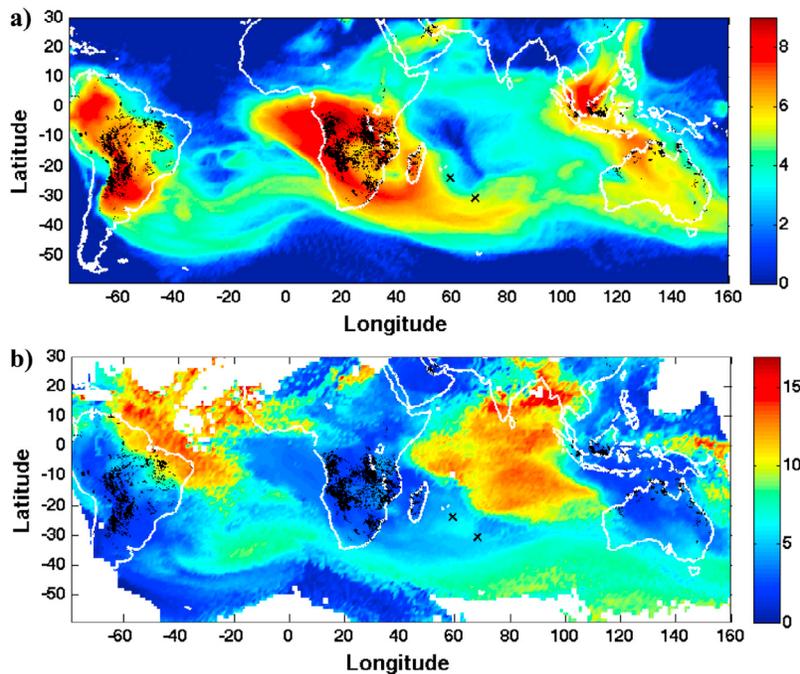
## 4. Discussion

### 4.1. Origin of the Aerosol Loaded Air Masses: Comparison With the GIRAFE Model

[36] The mean tropospheric lifetime of BB aerosols is usually estimated being in the 4 to 7 days range [Andreae,

1995; Rasch *et al.*, 2000; Edwards *et al.*, 2006]. In order to take into account long-range transports of aerosol plumes, seven days forward GIRAFE simulations were performed for each day from 7 to 15 September in order to compare GIRAFE results and lidar-Sun photometer observations for the 14–16 September period and to identify the origin of the air masses. Particles were emitted up to 2000 m agl (Section 2.2) for each fire pixel detected by MODIS (Section 2.1.4 and 2.2). Geographical and height (agl) distributions of the particles remaining in the troposphere between the 14 and 16 September have been averaged in a  $1^\circ \times 1^\circ$  gridded map on this three day period (Figure 9). Note that the geographical distribution of the particles (number of particles in a  $1^\circ \times 1^\circ$  square) is given in a log scale. The extreme ship positions during the investigated period ( $[31^\circ\text{S}, 68^\circ\text{E}]$  and  $[24^\circ\text{S}, 59^\circ\text{E}]$ ) are highlighted by the black crosses. Five main BB plumes can be identified in Figure 9: one plume exiting off southeast South America, crossing the South Atlantic ocean and joining the second plume exiting off South-Southeast Africa and Madagascar toward Australia, a third one exiting off Indonesia-Malaysia toward the southwestern Indian Ocean, a fourth exiting off northwest Australia and joining the Indo-Malaysian plume, and a fifth one exiting off northwest southern Africa toward South America. This simulated BB plume distribution is in accordance with previous studies on Southern Hemisphere pollution transportation patterns such as those by Annegarn *et al.* [2002] and Edwards *et al.* [2006]. Moreover, the long-range transport pathway at high tropospheric altitude from Southeast Asia and Indonesia-Malaysia toward the southwestern Indian Ocean is clearly visible in Figure 9b, showing a transportation altitude of  $\sim 13$  km [Dufлот *et al.*, 2010].

[37] One can see that the ship is on the edge of the BB plume during this period and almost at the junction between the BB plumes coming from South America, Southern Africa and Southeast Asia. This confirms the BB origin of the encountered aerosol plume and can explain the relatively low  $AOT_{355}$  measured during the crossing of the plume ( $\sim 0.1$ – $0.2$ ). To assess the relative contribution of each of the BB emission areas (South America, Southern Africa and Asia-Oceania) to the detected BB aerosol plume, we performed the same set of simulations separately for each of these three areas and counted the number of particles forming the detected plume. The resulting relative contributions are: 50% from South America, 44% from Southern Africa and 6% from Asia-Oceania. This agrees with the work of Dufлот *et al.* [2010] showing that the area contributing the most to the pollutant concentration in the Réunion Island area ( $21^\circ\text{S}, 55^\circ\text{E}$ ), which is  $\sim 600$  km to the location of the detected BB plume, is South America in September. However, the relative contributions by area calculated in our present work should be considered carefully for the following reasons: 1) the same number of particles was emitted from each fire, independently from the type of biomass burnt; 2) emitted particles were uniformly distributed in altitude and emitted at the same injection height (2 km agl), independently from the location of the fire; 3) the processes affecting aerosols during their transportation were not taken into account, such as chemical reactions, growth and resulting deposition. These two last limitations of the model should also be taken into account in the exploitation of the simulated vertical distribution of the aerosols. One can see in Figure 9b that the simulated mean



**Figure 9.**  $1^\circ \times 1^\circ$  averaged particles (a) geographical [arbitrary unit] and (b) height [km] distributions for particles remaining in the atmosphere between 14 and 16 September, 2009 for 7 days GIRAPE simulations from 7 to 15 September, 2009. Black dots are fires detected by MODIS and black crosses are extreme ship positions on 14 and 16 September, 2009 ( $[31^\circ\text{S}, 69^\circ\text{E}]$  and  $[24^\circ\text{S}, 59^\circ\text{E}]$ ).

particles height between the two extreme ship positions is around 6 km amsl, which is not in accordance with the aerosol layer height observed from lidar measurements (up to  $\sim 2.7$  km, Figures 4 and 8). It seems therefore that the GIRAPE model is a powerful tool to simulate the horizontal distribution of BB aerosols, but is not suitable to simulate their height distribution. A new version of GIRAPE (v4) using the FLEXPART 8.2 model should be released soon. More detailed settling parameterization for aerosols is implemented in the FLEXPART 8.2 model, which should improve the ability of GIRAPE to simulate BB aerosols height distribution.

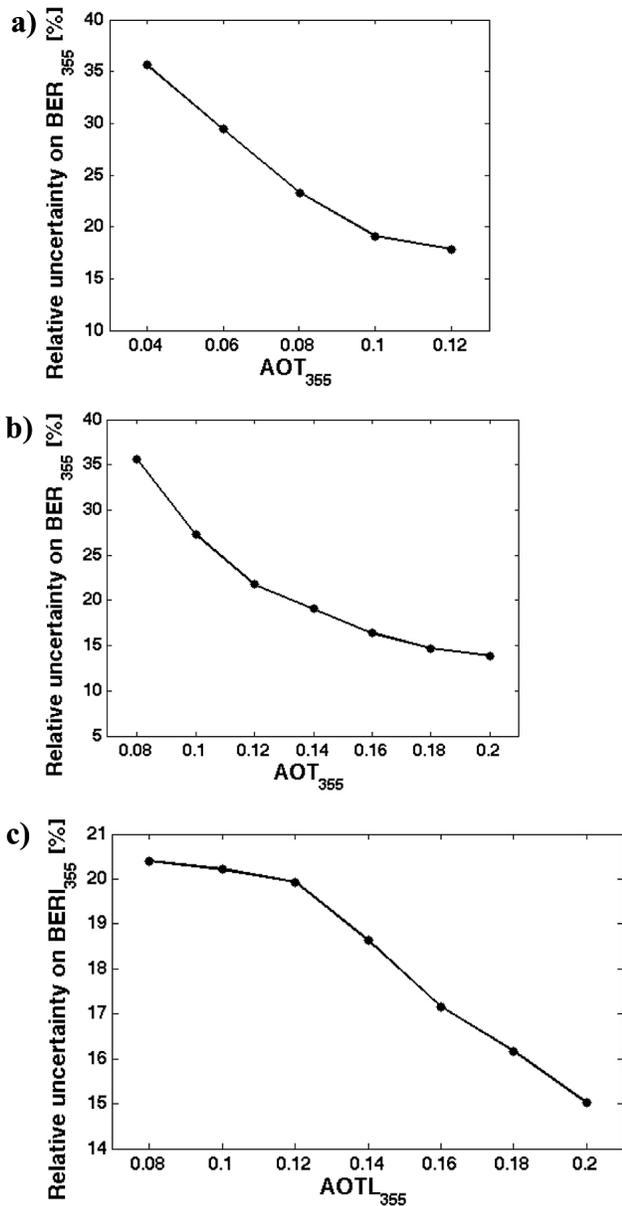
#### 4.2. Uncertainties on the Retrieved BER Values

[38] A direct-inverse model has been developed in order to assess uncertainties on the retrieval of marine and biomass burning aerosol optical properties.

[39] In case of marine aerosol (Section 3.2.1) a Gaussian aerosol layer has been simulated between 0 and 0.7 km amsl with an  $AOT_{355}$  of 0.08 and a  $BER_{355}$  of  $0.039 \text{ sr}^{-1}$  corresponding to the mean values observed during MACAMOZ campaign. Four sources of uncertainties have been identified: i) the uncertainty on the a priori knowledge of the vertical profile of the molecular backscatter signal, ii) the uncertainty on the lidar signal in the altitude range used for the normalization, iii) the statistical fluctuations in the lidar signal associated with random detection processes and iv) the uncertainty on AOT used to constraint lidar inversion (0.015, Section 2.1.2). The uncertainty on the a priori knowledge of the molecular contribution has been assessed to be lower than 2% [Chazette *et al.*, 2010] using a comparison

between several vertical sounding (i.e., radiosounding). The uncertainty on the lidar signal in the altitude range used for the normalization has been assessed to be lower than 6% [Royer *et al.*, 2011]. The statistical fluctuation in the lidar signal is here neglectable as Signal-to-Noise ratio of range-corrected signal is  $\sim 100$  at the reference. The uncertainty due to AOT value has been assessed using a Monte Carlo approach as in Chazette *et al.* [2001] with 1000 realizations assuming a normal probability density function with a standard deviation of 0.015 around different values of  $AOT_{355}$ . Figure 10a shows the relative uncertainty on the retrieved BER for marine aerosols as a function of the  $AOT_{355}$ . The standard deviation of 0.015 on  $AOT_{355}$  has been found to be the major source of uncertainty (between 87 and 97% of total uncertainty). The different sources of uncertainty have been supposed to be independent which lead to an overall uncertainty corresponding to 18 to 36% ( $\sim 0.007$  to  $0.014 \text{ sr}^{-1}$ ) of error on  $BER_{355}$  values for marine aerosols for a mean  $AOT_{355}$  of 0.12 and 0.04 respectively. The mean uncertainty on  $BER_{355}$  for marine aerosols equals  $0.009 \text{ sr}^{-1}$  (6 sr).

[40] In case of the mixing between BB and marine (MIX) aerosols (Section 3.2.2) a Gaussian aerosol layer has been simulated between 0 and 2 km amsl with an  $AOT_{355}$  of 0.12 and a  $BER_{355}$  of  $0.021 \text{ sr}^{-1}$  corresponding to the mean values observed during the crossing of the BB plume during the KAMASUTRA campaign. The same four sources of uncertainties as in the case of marine aerosols have been identified. Figure 10b shows the relative uncertainty on the retrieved BER for MIX aerosols as a function of the  $AOT_{355}$ . The standard deviation of 0.015 around the values of



**Figure 10.** (a) Relative uncertainty on the retrieved  $BER_{355}$  as a function of the  $AOT_{355}$  for the marine aerosols. (b) Relative uncertainty on the retrieved  $BER_{355}$  as a function of the  $AOT_{355}$  for the MIX aerosols. (c) Relative uncertainty after temporal averaging on the retrieved  $BERL_{355}$  as a function of the  $AOTL_{355}$  for the MIX aerosols.

$AOT_{355}$  has been found to be the major source of uncertainty (between 80 and 97% of total uncertainty). The different sources of uncertainty have been supposed to be independent which lead to an overall uncertainty corresponding to 14 to 36% ( $\sim 0.003$  to  $0.008$   $sr^{-1}$ ) of error on BER values for MIX aerosols. The mean uncertainty on  $BER_{355}$  ( $LR_{355}$ ) for the mixed marine and BB aerosols equals  $0.004$   $sr^{-1}$  (9 sr).

[41] In case of  $BERL_{355}$  retrieved thanks to the instrumental constant  $C_\lambda$  through  $AOTL_{355}$  for MIX aerosols (Section 3.3), a Gaussian aerosol layer has been simulated between 0 and 2 km amsl with an  $AOTL_{355}$  of 0.12 and a

$BERL_{355}$  of  $0.029$   $sr^{-1}$  corresponding to the mean values retrieved during the crossing of the BB plume. One additional source of uncertainty has to be considered in this case: relative uncertainty on  $C_\lambda$  (12%) which leads to an uncertainty on  $AOTL_{355}$  of 0.06. In order to reduce the induced uncertainty on the retrieved  $BERL_{355}$ , lidar profiles have been averaged over ten profiles. Uncertainty on  $AOTL_{355}$  has been found to be the major source of uncertainty ( $>98\%$  of total uncertainty on  $BERL_{355}$ ).  $BERL_{355}$  have been averaged over 10 profiles in order to reduce the uncertainty of the retrieval. Figure 10c shows the final relative uncertainty after temporal averaging on the retrieved  $BERL_{355}$  for MIX aerosols as a function of the  $AOTL_{355}$ . The different sources of uncertainty have been supposed to be independent which lead to an overall uncertainty corresponding to 15 to 20% ( $\sim 0.004$  to  $0.006$   $sr^{-1}$ ) of error on BER values for MIX aerosols. The mean uncertainty on  $BERL_{355}$  ( $LR_{355}$ ) for the mixed marine and BB aerosols equals  $0.005$   $sr^{-1}$  (6 sr).

### 4.3. Comparison With Other Studies

[42] Table 1 shows retrieved  $BER_\lambda$  ( $LR_\lambda$ ) values from previous studies for BB and marine aerosols, as well as the used lidar wavelengths and the measurement (for marine aerosols) and source (for marine+BB aerosols) areas. Table 1 is not exhaustive, but rather a summary of what can be found in the literature about  $BER_\lambda$  ( $LR_\lambda$ ) for marine aerosols and for BB aerosols coming from Southern Africa and South America. The values presented in Table 1 are from direct ground based lidar measurements that do not rely on assumptions of particle composition, shape, or size (except for data from *Catrrall et al.* [2005]). Note that no values of  $BER_{355}$  for marine aerosols or for BB aerosols coming from Southern Africa have been found in the literature. Considering the weak spectral dependence (low  $a$ ) of marine aerosols in the UV and visible spectral domains, the values retrieved in the frame of this study are in agreement with the results of previous works. The mean  $BER_{355}$  ( $LR_{355}$ ) of  $0.039 \pm 0.009$   $sr^{-1}$  ( $26 \pm 6$  sr) we derived for marine aerosols is nearly identical to the mean of those previously retrieved, which go from  $BER_{523} = 0.030 \pm 0.005$   $sr^{-1}$  ( $LR_{523} = 33 \pm 6$  sr) [*Welton et al.*, 2002] to  $BER_{532} = 0.043 \pm 0.009$   $sr^{-1}$  ( $LR_{532} = 23 \pm 5$  sr) [*Müller et al.*, 2007], both in the tropical northern Indian Ocean.

[43] Previous retrieved  $BER_{355}$  ( $LR_{355}$ ) values for Eastern European BB aerosols range from  $0.015 \pm 0.006$   $sr^{-1}$  ( $70 \pm 30$  sr) [*Amiridis et al.*, 2009] to  $0.017$   $sr^{-1}$  (59 sr) [*Balis et al.*, 2003]. *Chazette et al.* [2007] give  $BER_{355}$  ( $LR_{355}$ ) of  $0.008 \pm 0.003$   $sr^{-1}$  ( $125 \pm 35$  sr) over southern Niger. For Southern African BB aerosols, previously retrieved  $BER_\lambda$  ( $LR_\lambda$ ) values range from  $0.016 \pm 0.003$   $sr^{-1}$  ( $63 \pm 10$  sr) [*Campbell et al.*, 2003] to  $0.017 \pm 0.002$   $sr^{-1}$  ( $60 \pm 8$  sr) [*Catrrall et al.*, 2005] at  $\lambda = 523$  and  $550$  nm, respectively.

[44] One can see that our retrieved  $BER_{355}$  ( $LR_{355}$ ) for BB aerosols mainly coming from Southern Africa and South America and mixed with marine aerosols is within the range of the BER (LR) retrieved at  $355$  nm for BB aerosols coming from Eastern Europe, but slightly above (below) the range of the previously retrieved  $BER_\lambda$  ( $LR_\lambda$ ) at higher  $\lambda$  (523 and 550 nm) for Southern African BB aerosols. This can be explained by the fact that, oppositely to these previous studies, the detected BB aerosol plume is encountered more

**Table 1.** Retrieved  $BER_\lambda$  (and  $LR_\lambda$ ) From Previous Ground Based Studies for BB and Marine Aerosols

Reference	Wavelength [nm] <sup>a</sup>	Marine Aerosols		BB Aerosols	
		BER [sr <sup>-1</sup> ] (LR [sr]) <sup>b</sup>	Measurement Area <sup>c</sup>	BER [sr <sup>-1</sup> ] (LR [sr]) <sup>d</sup>	Source Area <sup>e</sup>
<i>Flamant et al.</i> [1998]	532	0.041 ± 0.008 (24 ± 5)	North Atlantic		
<i>Franke et al.</i> [2001]	532	>0.033 (<30)	Northern Indian Ocean		
<i>Müller et al.</i> [2001]	532	0.040 (25)	Northern Indian Ocean		
<i>Welton et al.</i> [2002]	523	0.030 ± 0.005 (33 ± 6)	Tropical Indian Ocean		
<i>Catrrall et al.</i> [2005]	550	0.036 ± 0.006 (28 ± 5)	Global	0.017 ± 0.002 (60 ± 8)	South America and Southern Africa
<i>Müller et al.</i> [2007]	532	0.043 ± 0.009 (23 ± 5)	Northern Indian Ocean	0.019 ± 0.004 (53 ± 11)	Siberia/Canada
<i>Müller et al.</i> [2007]	<b>355<sup>f</sup></b>			0.022 ± 0.006 (46 ± 13)	Siberia/Canada
<i>Balis et al.</i> [2003]	<b>355</b>			0.017 (59)	Eastern Europe
<i>Campbell et al.</i> [2003]	523			0.013 ± 0.003 (63 ± 10)	Southern Africa
<i>Chazette et al.</i> [2007]	<b>355</b>			0.008 ± 0.003 (125 ± 35)	Niger
<i>Amiridis et al.</i> [2009]	<b>355</b>			0.015 ± 0.006 (70 ± 30)	Eastern Europe
This study	<b>355</b>	0.039 ± 0.009 (26 ± 6)	Southwestern Indian Ocean	0.021 ± 0.006 (48 ± 12)	South America and Southern Africa

<sup>a</sup>The wavelength  $\lambda$  at which the  $BER_\lambda$  (and  $LR_\lambda$ ) were retrieved.

<sup>b</sup>The mean  $BER_\lambda$  (and  $LR_\lambda$ ) for marine aerosols with the associated standard deviation.

<sup>c</sup>The marine aerosols measurement area.

<sup>d</sup>The mean  $BER_\lambda$  (and  $LR_\lambda$ ) for BB aerosols with the associated standard deviation.

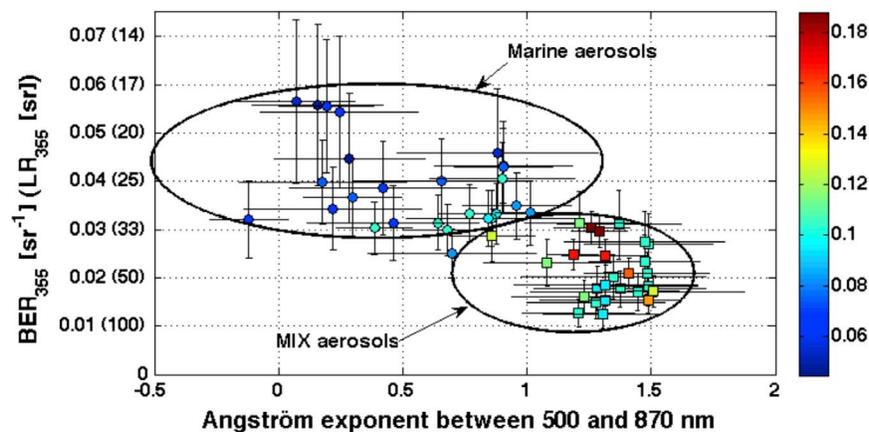
<sup>e</sup>The identified BB aerosols source area.

<sup>f</sup>Boldfacing indicates studies that used the same lidar wavelength as this study.

than 2500 km from its emission area, and thus can be affected by changing of the particles optical properties during the transportation. *Müller et al.* [2007] retrieved a  $BER_{355}$  ( $LR_{355}$ ) value of  $0.022 \pm 0.006 \text{ sr}^{-1}$  ( $46 \pm 13 \text{ sr}$ ) – which is very close to our value – over Europe for BB aerosol plume coming from Siberia/Canada. They state that long-range transport of aerosols seems to increase (decrease) the BER (LR). This may be due, in our case, to mixing with marine particles over the South Atlantic and Southern Indian Ocean, particle growth due to uptake of water or precursor gases, photochemical reactions, and particle growth because of coagulation. This can explain the difference with the  $BER_{355}$  ( $LR_{355}$ ) retrieved over Southern Niger by *Chazette et al.* [2007]. We must also consider that the present study is actually the first report (to our knowledge) on the characterization of the aerosols optical prop-

erties over the southern Indian Ocean from ground-based mobile lidar measurements, thus reference data are missing.

[45] Figure 11 displays a summary of the Ångström coefficient between 500 and 870 nm ( $a$ ) versus the  $BER_{355}$  ( $LR_{355}$ ) retrieved from the synergy between the lidar and the Sun photometer (Section 3.2.1 and 3.2.2.1). The  $BER_{355}$  ( $LR_{355}$ ) boundary between the “marine” and “MIX” (BB+marine) types is the minimum (maximum) BER (LR) value found in the literature for marine aerosols (BER =  $0.030 \text{ sr}^{-1}$  and LR = 33 sr found by *Welton et al.* [2002] at 523 nm). The signatures of marine and MIX aerosols are well identified because little to no overlap is observed. Marine aerosols  $BER_{355}$  ( $LR_{355}$ ) and  $a$  range from 0.030 to  $0.057 \text{ sr}^{-1}$  (18–33 sr) and from 0.10 to 0.95, respectively, while MIX aerosols  $BER_{355}$  ( $LR_{355}$ ) and  $a$  range from 0.012 to  $0.030 \text{ sr}^{-1}$  (33–83 sr) and from 0.86 to 1.51, respectively. This is in accordance with the work of



**Figure 11.** Ångström exponent between 500 and 870 nm versus retrieved BER at 355 nm for marine aerosols (MACAMOZ campaign, circles) and MIX aerosols (KAMASUTRA campaign, squares). Error bars on  $BER_{355}$  are from uncertainty described in Section 4.2. Error bars on Ångström exponent are from uncertainty on AOT and have been computed following a similar approach showed by *Hamonou et al.* [1999]. The gray scale gives related AOT at 380 nm.

Cattrall *et al.* [2005] showing a clear separation between the BER related to BB and marine aerosols. One can verify also in Figure 11 that the AOT related to BER of MIX aerosols are higher than the AOT related to BER of only marine aerosols.

## 5. Conclusion

[46] Lidar observations were performed on the research vessel Marion Dufresne during two ship-based opportunity experiments: one around Madagascar in late austral summer 2009 and one between La Réunion (21°S, 55°E) and the Kerguelen Islands (50°S, 70°E) in austral winter (Southern Hemisphere BB season) 2009. A BB aerosol layer has been encountered between (31°S, 69°E) and (24°S, 59°E) in mid September 2009. The Sun photometer-derived  $AOT_{355}$  has been used as a constraint to determine the mean  $BER_{355}$  ( $LR_{355}$ ) in the marine aerosol layer as well as in the mixed BB+marine aerosol layer, and leads to mean value of  $0.039 \pm 0.009 \text{ sr}^{-1}$  ( $26 \pm 6 \text{ sr}$ ) and  $0.021 \pm 0.006 \text{ sr}^{-1}$  ( $48 \pm 12 \text{ sr}$ ), respectively. The mean aerosol extinction profile has been calculated which reveals an aerosol layer extending up to  $\sim 3 \text{ km}$  amsl. Lidar calibration is used to inverse data without any simultaneous Sun photometer measurements (as nighttime data) and the time evolution of the aerosol extinction properties and vertical extension are documented. We used the GIRAFE/FLEXPART model to confirm the BB origin of the detected aerosol layer: the encountered aerosol plume comes mainly from BB occurring in South America and Southern Africa. However, the mean height of the encountered plume as simulated by GIRAFE is not in agreement with the altitude of the detected aerosol layer. Further studies should be performed with the next version of GIRAFE-FLEXPART (v4), which includes additional aerosols parameterization. This first (to our knowledge) direct ground-based mobile measurement of the aerosols characteristics and spatial distribution in the southern Indian Ocean can be useful for reducing uncertainties in the global climate models for the southern hemisphere.

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