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Atmospheric Measurement Techniques Discussions

Novel aerosol extinction coefficients and lidar ratios over the ocean from CALIPSO-CloudSat: Evaluation and global statistics

David Painemal[1,2], Marian Clayton[1,2], Richard Ferrare[2], Sharon Burton[2], Damien Josset[3], and Mark Vaughan[2]

[1]Science Systems and Applications Inc., Hampton, VA, 23666 USA
[2]NASA Langley Research Center, Hampton, VA, 23666 USA
[3]US Naval Research Laboratory Stennis Space Center, MS, 39529, USA

Correspondence to: David Painemal (david.painemal@nasa.gov)

Abstract. Aerosol extinction coefficients (σ_a) and lidar ratios (LR) are retrieved over the ocean from CALIOP attenuated backscatter profiles by solving the lidar equation constrained with aerosol optical depths (AOD) derived by applying the Synergized Optical Depth of Aerosols (SODA) algorithm to ocean surface returns measured by CALIOP and CloudSat's Cloud Profiling Radar. σ_a and LR are retrieved for two independent scenarios that require somewhat different assumptions: a) a single homogeneous atmospheric layer (1L) for which the LR is constant with height, and b) a vertically homogeneous layer with a constant LR overlying a marine boundary layer with a homogenous LR fixed at 25 sr (2-layer method, 2L). These new retrievals differ from the standard CALIPSO version 4.1 (V4) product, as the CALIOP-SODA method does not rely on an aerosol classification scheme to select LR. CALIOP-SODA σ_a and LR are evaluated using airborne high spectral resolution lidar (HSRL) observations over the northwest Atlantic. CALIOP-SODA LR (1L and 2L) positively correlates with its HSRL counterpart (linear correlation coefficient r>0.67), with a negative bias smaller than 13.2%, and a good agreement for σ_a (r ≥0.78) with a small negative bias (≤|-9.2 %|). Furthermore, a global comparison of optical depths derived by CALIOP SODA and CALIPSO V4 reveals substantial differences over regions dominated by dust and smoke, in qualitative agreement with previously reported discrepancies between MODIS and CALIPSO AOD.

Global maps of CALIOP-SODA LR feature high values over littoral zones, consistent with expectations of continental aerosol transport offshore. In addition, seasonal transitions associated

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

with biomass burning during June to October over the southeast Atlantic are well reproduced by CALIOP-SODA LR.

1. Introduction

Advances in our understanding of the 3D structure of atmospheric aerosols have been greatly accelerated with the advent of the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP), onboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO, Winker et al., 2009; 2010, 2013). CALIOP has provided the first global view of aerosol distribution in the boundary layer and free troposphere (Winker et al., 2013), progressed our knowledge of the long-range transport of dust (e.g. Liu et al., 2008; Uno et al., 2010; Yu et al., 2015) and smoke (e.g. de Laat et al., 2012; Das et al., 2017; Khaykin et al., 2018), and facilitated the evaluation of chemical transport models (Nowottnick et al., 2015; Koffi et al., 2016), among many other accomplishments in the area of aerosol and cloud research.

CALIOP estimates aerosol extinction coefficients on a global scale with an unprecedented vertical detail. The undetermined problem of solving the lidar equation with two physical unknowns, the aerosol extinction and backscatter coefficients, is addressed in the CALIPSO algorithm by relating both variables via an extinction-to-backscatter ratio, or lidar ratio (LR). This standard technique (e.g. Fernald, 1984) expresses the lidar equation in terms of only one unknown, if LR is prescribed. As aerosol types can be related to specific values of lidar ratios (e.g. Müller et al., 2007), the CALIPSO algorithm utilizes predefined LR assigned to a number of aerosol types, which in turn, are identified using the CALIPSO automated aerosol typing algorithm (Omar et al., 2009; Kim et al., 2018). Thus, the quality of CALIOP retrievals will depend on how well the actual lidar ratios match the pre-tabulated values and to what extent the aerosol typing algorithm properly classifies aerosols. Another source of uncertainty is the detectability limits of the CALIPSO algorithm, which prevents retrieving aerosol properties for tenuous aerosol layers (Rogers et al., 2014; Thorsen et al., 2017). For instance, Toth et al. (2018) found that no aerosol was detected within ~71% of the CALIOP profiles measured during daytime and ~41% of the nighttime measurements. More aerosol detection during nighttime is explained by the absence of solar background noise, which leads to a significantly better signal to noise ratio. The aforementioned factors likely explain discrepancies between CALIOP and other remote sensing datasets such as

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

those from the MOderate resolution Imaging Spectroradiometer (MODIS) and AERONET (e.g. Redemann et al., 2012; Schuster et al., 2012).

Uncertainty reduction in the selection of LR can be attained by constraining the lidar equation solution with an independent estimate of aerosol optical depth (AOD). This implies the minimization of the error between the retrieved AOD (estimated from the retrieved extinction coefficient coefficient) and the target AOD by iteratively adjusting LR. Burton et al. (2010) utilize AOD from the MODIS instruments on board both Aqua and Terra satellites for estimating aerosol extinction from CALIOP for cases in which AOD exceeds 0.15 (0.2) over the ocean (land). Similarly, Royer et al. (2010) applied an equivalent method for estimating LR and extinction coefficients over the Po Valley in Italy. Although CALIOP-MODIS retrievals in Burton et al. (2010) tend to compare better with airborne measurements relative to CALIPSO standard product (Version 2), MODIS AOD is limited to daytime, and MODIS and CALIOP differ in their along-track spatial resolution. These previous studies have proven the value of counting on independent CALIOP retrievals for evaluating CALIPSO's standard data products.

In this contribution, we present a new method in which CALIOP-based lidar ratios and aerosol extinction coefficients over the non-polar oceans are obtained by constraining the retrievals with AOD derived from cross-calibrated CALIOP and CloudSat Cloud Profiling Radar (CPR) surface echos, using the Synergized Optical Depth of Aerosols (SODA) product (Josset et al., 2008). SODA AOD is a suitable dataset, as it is collocated with CALIOP by definition and retrievals are possible during both daytime and nighttime for the period 2006-2011. After November 2011 SODA is only available for daytime, as CloudSat has operated in daylight-only operations mode to conserve power (Gravseth and Piepe, 2013). Our goal is to provide an independent CALIOP dataset that can be used for evaluating specific aspects of the CALIPSO Science Team product, as well as for investigating aerosol-related topics in climate research. We first summarize the algorithm and evaluate the new retrievals against state-of-the-art aerosol observations from the NASA Langley airborne High Spectral Resolution Lidar-1 (HSRL, Sections 3 and 4). Next, we compare the CALIOP-SODA extinction coefficient and AOD with their CALIPSO Science Team Version 4 counterparts. Lastly, we present global maps of lidar ratio and marine boundary layer aerosol optical depth, and provide a physical interpretation for the regional patterns derived from CALIOP-SODA.

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

2. Dataset

2.1.CALIOP

Version 4.1 (V4) CALIOP elastic backscatter lidar measurements at 532 nm and 1064 nm are utilized in this work. For the derivation of CALIOP-SODA retrievals, we use Level 1 lidar attenuated backscatter and the Level 2 Vertical Feature Mask product, with a 333 m horizontal resolution below 8.2 km. CALIOP V4 aerosol extinction coefficients and AOD estimates are taken from the Level 2 Aerosol Profile product at 5 km horizontal resolution. For comparing CALIOP SODA and V4 products, we follow the procedure outlined in Koffi et al. (2016): where the VFM feature classification flags indicate regions of clear air, we set the corresponding extinction coefficients to zero. While these regions are labeled as 'clear air', they are simultaneously assumed to be populated by highly diffuse aerosols that lie well below the CALIOP layer detection threshold.

2.2.SODA aerosol optical depth

SODA uses the relationship between CALIOP (532 nm and 1064 nm) and CPR (3.1 mm, 94 GHz) surface return signals, along with a correction for the atmospheric transmission at the radar wavelength, to derive AOD at the lidar wavelengths. In short, SODA estimates of AOD rely on the radar-to-lidar ocean surface scattering cross-calibration for cloud-free columns (Josset et al., 2008, 2010). Consequently, SODA can provide a cloud-free AOD without having to rely on an accurate assignment of a particular aerosol type with an appropriate lidar ratio. In addition, the algorithm does not depend on pre-determined aerosol models with a specific particle size distributions and refractive indexes, unlike MODIS. SODA AOD Version 2, based on CALIPSO Version 3 (V3), is developed at the ICARE data and services center (http://www.icare.univ-lille1.fr) in Lille (France) under the auspices of the CALIPSO mission and supported by the French National Centre for Space Studies (CNES). Josset et al. (2013) estimate a systematic error in SODA AOD of 0.015 and 0.059, respectively, for nighttime and daytime AOD. In addition, good agreement between SODA and MODIS has been reported in Josset et al. (2010, 2015), while Dawson et al. (2015) reports a root-mean-square-error of 0.03 between SODA and AERONET AOD and r = 0.59 for AERONET sites near the coast. Further, we also evaluate SODA AOD with HSRL data in Section 4, and compare SODA and MODIS AOD over the global ocean in Section 6. While 1064 nm SODA AOD is also utilized in this study, caution needs to be exercised when using the 1064 nm SODA data due to uncertainty calibrations in CALIPSO V3 (Vaughan et al., 2010).

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

Atmospheric Measurement Techniques Discussions

2.3.HSRL

CALIOP retrievals are evaluated against airborne measurements by the NASA Langley High Spectral Resolution Lidar (HSRL, Hair et al., 2008) at 532 nm. The instrument allows for the independent determination of aerosol extinction and backscatter coefficients at 532 nm (and thus, lidar ratio) using the HSRL technique (Eloranta, 2005). As HSRL measurements at 1064 nm are limited to attenuated backscatter, similar to CALIOP, only 532 nm HSRL retrievals will be utilized in this study. The data used in this study were acquired August 11–27, 2010 while the HSRL conducted a dedicated CALIPSO validation campaign over the Caribbean Sea (Burton et al., 2013; Rogers et al., 2014). As required for all HSRL-CALIPSO validation measurements, the HSRL flight paths during this campaign were spatially matched with coincident CALIPSO ground tracks (Rogers et al., 2014).

3. Derivation of aerosol extinction coefficient and lidar ratio

The method for deriving aerosol extinction coefficient (σ_a) and lidar ratio (LR) is based on Fernald (1984) applied to the CALIOP attenuated backscatter, and is briefly summarized in the following. For CALIOP, the lidar equation is expressed in terms of height z (range) as:

$$\beta_{att}(z) = \left(\beta_m(z) + \beta_a(z)\right) \cdot exp\left(-2\int_0^z \left(\sigma_m(z') + \sigma_a(z')\right)dz'\right) \quad (1)$$

Where β_{att} corresponds to the CALIOP total attenuated backscattering cross section, β_m and β_a denote the molecular (m) and aerosol (a) backscatter coefficients, and σ_m and σ_a are the molecular and aerosol extinction coefficients. Since the molecular contribution can be accurately estimated using atmospheric profiles from numerical weather models, the two unknowns are $\beta_a(z)$ and $\sigma_a(z)$. Equation (1) can be reduced to one unknown by relating extinction and backscatter coefficient via their lidar ratio, that is

$$LR = \frac{\sigma_a(z)}{\beta_a(z)}. \quad (2)$$

It follows that eq. (1) can be expressed in terms of LR and β_m as:

$$\beta_{att}(Z) = \left(\beta_m(z) + \beta_a(z)\right) \cdot exp\left(-2\int_0^z \left(\sigma_m(z') + LR \cdot \beta_a(z')\right)dz'\right) \quad (3)$$

β_m is estimated as a function of air density (from the Goddard Earth Observing System Model, Version 5 GEOS-5), and effect of ozone attenuation is accounted for in σ_m following

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

Vaughan et al. (2005). $\beta_a(z)$ and $\sigma_m(z)$ are, thus, estimated from Eq. (3) as in Fernald (1984), assuming a constant value of LR with height. The LR selection is physically constrained by comparing the retrieved aerosol optical depth ($AOD_{ret} = \int_0^z \sigma_a(z')dz'$) with SODA AOD ($AOD_{SODA}$), and LR is iteratively adjusted until the retrieved AOD matches the SODA AOD to within 0.001 or less (i.e., when $|AOD_{ret} - AOD_{SODA}| \leq 0.001$).

We also consider an additional scenario for solving the lidar equation, which consists of treating the atmospheric column as two layers, that is., the marine atmospheric boundary layer (MBL) and a second aerosol layer of as-yet-undetermined composition. This method is intended to better capture specific events with two predominant aerosol types, namely, smoke over marine aerosols and dust over marine aerosols, which are particularly frequent over the Atlantic Ocean. The LR for the MBL is assumed constant at 25 sr, as suggested by HSRL measurements over the ocean (Burton et al., 2012; 2013). This lidar ratio is slightly higher than the one compiled by Kim et al. (2018) for maritime aerosols (23 sr). $\sigma_a(z)$ and the upper layer LR are iteratively calculated using the Fernald method with the constraint provided by AOD_{SODA}, and LR =25 sr in MBL. MBL height is computed by applying the bulk Richardson number method (McGraw-Spangler and Molod, 2014) to GEOS-5 atmospheric fields.

In sum, aerosol extinction profiles and lidar ratio are calculated using one of two independent assumptions: either a single homogeneous layer (1L method), or a two-layer atmosphere with a prescribed LR=25 sr in the boundary layer and a single free tropospheric LR, which is estimated during the iterative process (2L method).

The CALIOP attenuated backscatter at 333 m resolution is taken from the Level 1 CALIPSO product. Before retrieving LR and σ_a, three contiguous 333-m lidar attenuated backscatter samples are averaged to achieve a 1 km along-track resolution. Similarly, SODA AOD retrieved at 333m is averaged to 1 km resolution. In addition, the Feature Classification Mask product is utilized for identifying cloudy pixels and cases with fully attenuated signal, in which CALIOP-SODA retrievals are not possible.

4. CALIOP-SODA evaluation with airborne HSRL measurements

CALIOP-SODA retrievals of aerosol extinction coefficient, lidar ratio and AOD are evaluated using eight flights during August 2010 over the western Atlantic, for the domain bounded by 70˚W-55˚W and 13˚N-35˚N (Figure 1a). CALIOP-SODA is spatially averaged to

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

match the nominal 5 km horizontal resolution of CALIPSO V4, and only samples with 5-km cloud-free scenes are retained. Both CALIPSO V4 and CALIOP-SODA are then spatially collocated with the aircraft track (Figure 1) for samples with a temporal mismatch of less than 90 minutes (Rogers et al., 2014). Lastly, satellite and airborne observations are spatially averaged to a common 0.5 ˚ resolution (in latitude). Approximately 42 and 46 0.5˚-samples were collocated with HSRL (CALIOP-SODA and CALIOP, respectively).

The HSRL measurements during Caribbean 2010 were characterized by the presence of dust, dust mixed with maritime aerosols, and continental pollution; the occurrence of pure maritime aerosols was confined to the boundary layer (Burton et al., 2013). This aerosol typing is manifested in a lidar ratio of 25 sr below 500 m, and a linear increase with height that reaches values of 40-45 sr in the free troposphere (Fig. 1b). These measurements also provide justification for the use of a lidar ratio of 25 sr in the boundary layer for the 2L method. Before evaluating aerosols extinction coefficients and lidar ratios, we compare SODA AODs and CALIOP V4 AODs against their HSRL counterparts (Fig. 2a). In general, both CALIOP-based retrievals correlate well with the HSRL, with a slightly higher correlation for SODA, and absolute bias between 10-17%), with SODA (CALIOP V4) underestimating (overestimating) AOD. Linear fits of SODA and V4 AOD relative to HSRL (red and blue lines in Fig. 2a) indicate that the SODA bias is relatively constant with AOD whereas a V4 AOD overestimate tends with increase with AOD especially during nighttime. Nighttime and daytime correlations remain approximately the same for both CALIOP V4 and SODA. However, V4 linear correlation coefficient for AOD < 0.3 are slightly lower for daytime (r = 0.78) than nighttime (r = 0.94), whereas SODA daytime/nighttime correlations for low AOD remain high ($r \geq 0.93$). The reduced daytime correlation for CALIOP V4 is expected as the reduced signal to noise ratio due to the solar background signal hampers the algorithm's ability to detect and classify aerosols. Finally, in terms of the root-mean-square error (RMSE), SODA RMSE (24.2% relative to the mean) is smaller than that for CALIOP V4 (31.2%, Table 1).

The evaluation of CALIOP-SODA lidar ratio and aerosol extinction coefficient is summarized in the following. For LR, we use the column-effective lidar ratio, calculated as:

$$LR_{HSRL} = \frac{\sum_{z=z0}^{6km} \sigma_a(z)}{\sum_{z=z0}^{6km} \beta_a(z)} \qquad (4)$$

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

For evaluating CALIOP SODA 1L LR, LR_{HSRL} in eq. (4) is estimated using the last range bin above the ocean surface (37.8 m) as the lower bound, z_0. In addition, the comparison between CALIPSO SODA 2L LR and LR_{HSRL} is performed by recomputing LR_{HSRL} using the MBL height for z_0 in eq. 4. Since valid HSRL extinctions retrievals are only derived for heights above 270 m from the surface, we have assumed a constant extinction coefficient for the layer below 270 m, with values taken from the lowest height with available retrievals (~ 270 m). The comparison depicted in Fig. 2b, yields r = 0.67-0.74 between both CALIOP-SODA methods (1L and 2L) and HSRL, with a negative mean bias smaller than 13%, and RMSE of up to 8.1 sr (Figure 2b and Table 2).

Mean vertically-resolved aerosol extinction coefficients from SODA, CALIOP V4, and HSRL are depicted in Figure 3a and b for daytime and nighttime observations, respectively. The agreement between HSRL (red) and CALIOP-SODA 1L and 2L (overlapped gray and black) is remarkable throughout the lower troposphere, with a maximum overestimation of 0.027 km^{-1} (50%) near 500 m. CALIOP-SODA 1L and 2L yield identical results, which is likely the effect of a shallow marine boundary layer (<500 m). In contrast, CALIOP V4 (blue) consistently overestimates the airborne measurements for heights below 1 km during both daytime and nighttime, with magnitudes up to 0.102 km^{-1} (100%) relative to the HSRL during nighttime and 0.078 km^{-1} (140%) during the day. This overestimate is explained by the CALIPSO V4 constant lidar ratio of 37 sr for dusty marine aerosol, which is generally higher than the lidar ratio retrieved by both the HSRL and SODA for Caribbean 2010 (Figure 2b). Interestingly, both CALIOP-SODA and CALIOP V4 correlate well with the HSRL, with correlations around 0.80 (Table 3). The RMSE for CALIOP V4 is also higher than that for CALIOP-SODA especially below 1 km, with maxima around 0.12 km^{-1} (155%) and 0.06 km^{-1} (83%) for CALIOP V4 and CALIOP-SODA, respectively (Fig 3c). Aerosol extinction coefficient statistics for the atmospheric column below 3.0 km (Table 2) corroborate the overall smaller bias and RSME of CALIOP-SODA relative to V4.

5. Global analysis

5.1. Comparison between CALIOP-SODA and CALIOP-V4

Five months of collocated SODA and CALIOP V4 Level 2 data during June-October 2010 were compared over non-polar oceanic regions with the goal of identifying main differences in aerosol extinction coefficient profiles. This period was selected because of the high global

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

climatological AOD observed over the ocean by CALIOP (e.g. Yu et al., 2010). We first averaged 1km CALIOP-SODA to the V4 Level 2 nominal resolution (5km). Then, CALIOP-SODA and CALIOP V4 retrievals were further reduced by averaging the retrievals to a common 25 km resolution. Lastly, we reduced the potential effect of overcast scenes in the retrievals by limiting the comparison to 25-km samples with less than 2/3 (67%) of cloudy coverage. Cloud cover was derived from the 333 m Vertical Feature Mask and determined as the ratio between profiles with at least one cloudy feature in the atmospheric column to the total. To circumvent CALIOP's narrow field of view, we calculated the statistics in 6˚x3˚ (longitude x latitude) grids.

We first focus on the AOD difference (ΔAOD) between CALIOP V4 and SODA at 532 nm and 1064 nm, for day and nighttime (Figure 4). Daytime 532 nm ΔAOD maps reveal higher V4 AOD than SODA for the northeast Atlantic (NEA) and the Indian Ocean (IO), whereas V4 AOD is smaller than SODA over the southeast Atlantic (SEA) and over vast regions of the open ocean. As will be discussed in Section 6, these differences are similar to those observed between CALIOP V3 and MODIS (Redemann et al., 2012). Overall, nighttime differences in 532 nm AOD appear to diminish especially for the SEA and the northwest Pacific (NWP), while the positive ΔAOD remains high over IO and NEA.

We also show ΔAOD for the 1064 nm channel in Figure 4 (lower panels). The largest ΔAOD values are mostly confined to the NEA and IO domains, with higher values for SODA AOD, while nighttime ΔAOD are similar to its daytime counterpart.

Matched CALIOP-SODA and CALIOP V4 mean vertical profiles of aerosol extinctions over the regions defined in Figure 4 (black boxes) are shown in Figs. 5 and 6, for the 532 nm and 1064 nm channels, respectively. CALIOP-SODA derived using the 1L (CAL_S 1L) and 2L (CAL_S 2L) methods are depicted in black and blue, while CALIPSO V4 (CAL_{V4}) is in red. The main differences, in agreement in AOD differences in Figure 4, are found: a) over IO and NEA where CALIPSO V4 extinction profiles are higher than CALIOP-SODA, and b) over SEA, with lower V4 extinctions than CALIOP-SODA. Even though the main V4-SODA differences in extinction decrease during nighttime, especially over the SEA, the nighttime differences for NEA and IO remain nearly the same. Interestingly, the higher CALIOP V4 extinction for NEA and IO resembles the CALIPSO V4 overestimation during Caribbean 2010 (Fig. 3). CALIOP-SODA and V4 profiles differences for regions with small AOD differences, such as the south Pacific (SP) and the northwest Pacific (NWP), are modest. Another interesting aspect is the generally higher

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

variability of daytime CALIPSO V4 relative to SODA, manifested in the high standard deviations in Figure 5 (error bars). This indicates that SODA retrievals are more stable than CALIPSO V4 especially during the daytime, due to the AOD constraint. Moreover, the high solar background substantially degrades CALIPSO aerosol detection capabilities, affecting the retrieved extinction. Lastly, CALIOP-SODA differences between 1L and 2L are small, and typically confined to a layer below 700 m, where 2L tends to be smaller than 1L. This is explained, as in Section 4, by a relatively shallow mixed-layer height (< 500 m), where LR = 25 sr for the 2L method.

For completeness, we show in Figure 6 the aerosol extinction profiles for the 1064 nm channel. CALIOP-SODA and V4 profiles yield smaller differences relative to their 532 nm counterpart, in agreement with ΔAOD (Figure 4).

5.2. Maps of CALIOP-SODA Lidar ratio (LR) at 532 nm

Figures 7 and 8 depict global maps of 532 nm LR derived from the 1L (LR_{1L}) and 2L (LR_{2L}) assumptions, temporally averaged from March to August (MAMJJA, boreal spring-summer) and September to February (SONDJF, boreal autumn-winter) of 2010 from the 25-km averaged retrievals with cloud fraction less than 67%. Daytime 532 nm LR exhibits a clear spatial pattern with high values (>45 sr) in coastal regions especially off the southwestern African coast and east of China. The lowest values are observed over the western and central equatorial Pacific, with ratios less than 30 sr, which are typical of clean maritime environments (e.g. Burton et al., 2013). Semiannual transitions are primarily found near the continents, namely, the Southeast Atlantic, Mediterranean Sea, Indian Ocean, and off the coast of eastern Asia. Nighttime LRs are similar to their daytime counterparts, but with slightly higher values and a rather heterogeneous pattern, likely attributed to the reduced cloud-free sampling at night due to the increased cloud cover, especially over subtropical regions where stratiform and shallow cumulus clouds are abundant.

Comparing the two layer assumptions, LR_{2L} (Figure 9) is higher than LR_{1L}, especially for lidar ratios > 40 sr. This result is expected, as the prescribed MBL lidar ratio of 25 sr for 2L tends to be lower than the lidar ratio for any aerosol type that would be found above the marine boundary layer, and therefore lower than the column average or column effective lidar ratio. Therefore, to match the SODA AOD, the lidar ratio above the MBL in the 2L case must be larger than the column effective value that the 1L case derives. Overall, LR_{1L} and LR_{2L} differences are relatively

small (~ 5 sr), which, as we will show in the next section, is associated with the shallow MBL height estimated from the bulk Richardson number method, and therefore a relatively small fraction of aerosol that is controlled by the assumed marine lidar ratio in the 2L method.

5.3. Fractional CALIOP-SODA AOD at 532 nm in the marine boundary layer

CALIOP-SODA aerosol extinctions are further utilized for quantifying AOD in the boundary layer. We first show in Figure 9 the 2010 semiannual total SODA AOD for daytime (left) and nighttime (right) CALIPSO overpasses. Consistent with several studies (e.g. Kittaka et al., 2011; Redemann et al., 2012), high AOD primarily occur over the eastern Atlantic, in connection with biomass burning and dust emissions from southern and equatorial Africa. A second region of interest encompasses most of the Asian coastal region, where a combination of pollution and dust give rise to high AOD (Itahashi et al., 2010).

Before presenting MBL AOD, we show the MBL height maps (Figure 10), with typical heights below 800 m, and littoral maxima up to 1150 m in northern Africa and Eurasia. Next, we compute MBL AOD by numerically integrating CALIOP-SODA aerosol extinction coefficient from the surface to the MBL height. MBL AOD in Figure 11 shows a dissimilar pattern relative to its total AOD counterpart (Figure 9), manifested in a less dominant role of the southeast Atlantic. In addition, coastal Africa, Eurasia, and North America exhibit peaks in MBL AOD (>0.12) during boreal spring-summer. A second region with high AOD encompasses the extratropical oceans poleward of 45 ˚S/N, with a particularly consistent zonal band with high AOD in the Southern Ocean. As expected, 2L MBL AOD is lower than 1L due to the 2L assumption of a lidar ratio = 25 sr in the MBL. Except for the subtropical ocean, which features shallow MBL and low MBL AOD, a spatial modulation of the marine boundary layer in the MBL AOD is unclear. It is important to mention that estimates of the AOD apportioned in the boundary layer will depend on the MBL dataset utilized in the calculations. An alternative MBL height estimation derived from CALIOP attenuated backscatter (McGrath-Sprangler and Denning, 2013) yields similar if not slightly higher values than our GEOS-based MBL. However, MBL estimates based on thermodynamical vertical profiles (temperature, relative humidty) from meteorological analyses produce significantly higher MBL (von Engeln and Teixeira, 2013), closely matching the cloud top height of stratiform and shallow cumulus clouds. Thus, the MBL used here is expected to primarily represents the mixed-layer height (von Engeln and Teixeira, 2013).

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

The fraction of MBL AOD relative to the total is depicted in Fig. 12. The extratropical bands show the highest fraction of MBL AOD, accounting for up to 0.73 (73%) of the total AOD. Low fractions are found in the subtropics and tropics, with the lowest AOD fraction over the eastern Atlantic and the west-central Pacific. Interestingly, vast areas over the ocean feature AOD fractions of less than 40%, suggesting a significant contribution of free tropospheric aerosols to the total AOD. These results are qualitatively consistent with the results Bourgeois et al. (2018) using CALIPSO version 4.1.

6. Discussion

Due to the limited airborne and ground-based observations of lidar ratios over the ocean, a global assessment of CALIOP-SODA retrievals is challenging. One of the few global satellite-based estimates of lidar ratio is reported in Bréon (2013) who estimated LR utilizing the retrieved scattering phase function at 180° angle derived from the Polarization and Directionality of the Earth's Reflectances (POLDER) satellite instrument and a prescribed aerosol model. POLDER LR is somewhat comparable to CALIOP-SODA (Figure 8-9) with both retrievals yielding high LR over the coasts of eastern Africa and Eurasia, and a notable increase in LR over the Indian Ocean in boreal autumn-winter. In addition, both POLDER and CALIOP-SODA produce LR < 30 sr over the open ocean. On the other hand, LR from POLDER tend to be slightly higher, with a typical range between 30-70 sr. Bréon (2013) also indicates that because POLDER retrievals rely on scattered photon measurements, LR might be biased low in regions dominated by absorbing aerosol, such as the southeast Atlantic. A somewhat different method of retrieving LR from SODA AOD, documented in Josset et al. (2011), consists of analytically solving the lidar equation. The only available global analysis of LR using the technique in Josset et al. (2011) is documented in Dawson et al. (2015) for maritime aerosols only, reporting values between 20-40 sr.

As different aerosol types can be, to some extent, characterized by their lidar ratio, the reliability of CALIOP-SODA LR retrievals is qualitatively assessed by analyzing the consistency between the CALIOP-SODA LR spatial pattern and the regional occurrence of aerosol types. Burton et al. (2012), using HSRL measurements over North America and the adjacent Atlantic Ocean, provide the following lidar ratios for a number of aerosol types: the highest LR (45-80 sr) are typically attributed to smoke and urban aerosols, LR of 25-50 sr and 40 sr are associated with dust and polluted maritime aerosols (respectively), and maritime aerosols are characterized by lidar

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

ratios of less than 30 sr. Following the classification of Burton et al. (2012), CALIOP SODA lidar ratios (Fig. 7-8) will be interpreted in the context of major variations in atmospheric aerosols over the global oceans. For simplicity, we will primarily interpret daytime LR_{1L} in Figures 7a and c. Two distinct maxima in CALIOP-SODA LR (LR > 50 sr) for spring-summer (Figure 8a) are found over the southeast Atlantic and the Mediterranean Sea. The LR peak in the southeast Atlantic is explained by the well-documented biomass burning season over southern Africa, with massive fires events from May to September during the dry season (Roberts et al., 2009), and smoke being transported offshore by the prevailing winds during July to October (Adebiyi et al, 2015). The high spring-summer LR over the Mediterranean Sea is also expected given the southward pollution transport from Europe which is maximized in summer (Duncan and Bey, 2004). A major LR maximum in autumn-winter is observed south of India, over the Bay of Bengal and part of the Arabian Sea. This pattern is concomitant with the pervasive presence of pollution and biomass burning during the winter and pre-monsoon season (October to April, Krishnamurti et al., 2009). In contrast, during the monsoon season (June-September), dust aerosols become the dominant species over the Bay of Bengal (Das et al., 2013), which is manifested in the substantial reduction in SODA LR in spring-summer. Regions with intermediate CALIOP-SODA LR (35 sr< LR< 50 sr) are located over broad regions of the eastern Pacific, as well as narrow littoral bands over eastern Asian and the east coast of North America. These regions are likely influenced by a combination of maritime aerosols and pollution from the continents. Lastly, the regions with the lowest LR are located over the tropical ocean, where AOD is the lowest (Figure 10). LR over the Southern Ocean is typically around 30-35 sr, which is rather surprising for such a remote region with limited continental influence, where maritime aerosols (and associated lidar ratios near 25 sr) are expected to be the dominant aerosol type. Interestingly, Omar et al. (2009) noted a large number of cases in which the CALIPSO aerosol classification algorithm detected continental clean aerosol, which in turn, the CALIPSO algorithm assigns a 532 nm lidar ratio of 35 sr (version 3) and 53 sr (version 4.1). Lastly, the interpretation the 1064 nm CALIOP-SODA is not attempted here due to the lack of independent measurements and calibration uncertainties associated with the use of CALIPSO V3 for deriving SODA AOD. A future release of SODA based on CALIPSO V4 will benefit from the improved calibration of V4, which is estimated to be within 3% (Vaughan et al., 2018).

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

An aspect that deserves further discussion is the reliability of SODA AOD, as it is essential for constraining the lidar equation in our study. In this study we find a high linear correlation between SODA and HSRL AOD (r=0.96), with no clear relationship between SODA biases and AOD magnitudes, and a SODA-to-HSRL RSME comparable to the one estimated between SODA and AERONET in Dawson et al. (2015). The differences between SODA and the CALIPSO V4 AOD (Figure 4) also support inferences based on comparisons between MODIS and CALIPSO Science Team AOD over the ocean (Redemann et al., 2012; Kim et al., 2013). Analogous to our results, Redemann et al. (2012) found an underestimation of CALIOP Version 3 of MODIS collection 5 over the southeast Atlantic in July, and overestimation over the Bay of Bengal and the zonal band near the equatorial Atlantic Ocean. Redemann et al. (2012) and our results both point to an overestimation of CALIPSO V4 AOD over oceanic regions dominated by dust, and underestimation in regions dominated by smoke. To verify that SODA-CALIPSO V4 differences are mainly attributed to CALIPSO V4 biases, we perform an additional comparison using Aqua-MODIS Level 2 550 nm AOD (MYD04_3K product), taken from the latest Collection 6.1 (Levy et al., 2013) for the June to September period of 2010. Cloud-free 3-km MODIS AOD pixels are collocated with CALIPSO track and averaged to approximately 25 km (along track) to match the averaged 25 km SODA retrievals. Next, MODIS-SODA mean differences are averaged every 6˚x3˚ grid, and depicted in Figure 13. The MODIS-SODA differences in Figure 13 are typically within the [0.06 -0.06] range, with negligible values over the eastern Atlantic and the northwest Pacific. Although ΔAOD reaches up to 0.12 over the Indian Ocean, these differences are smaller than those between CALIPSO V4 and SODA (Figure 4, upper left panel). Overall, MODIS further corroborates that CALIPSO V4 AOD is biased over regions dominated by smoke and dust. We note that the plausible oceanic CALIOP V4 bias dependence on aerosol types suggested in our study might not be applicable over land, where AOD for dust is underestimated by CALIPSO (e.g. Schuster et al., 2012).

7. Concluding remarks

One year of a new CALIOP-based aerosol extinction coefficient and lidar ratio dataset has been presented, with the goal of providing a flexible dataset for climate research as well as independent retrievals that can be helpful for refining CALIPSO Science Team algorithms. The new retrievals build on the CALIPSO V4 total attenuated backscatter and cloud mask data

products. However, the method that we used to invert the lidar equation differs fundamentally from the CALIOP standard aerosol product, as it does not rely upon an aerosol classification module to prescribe the lidar ratio. We evaluated CALIOP-SODA AOD, LR, and extinction using airborne HSRL retrievals over the western Atlantic, and found excellent agreement, with statistically significant correlations and biases less than 27 %. Given these encouraging results, we envision potential uses of CALIOP-SODA lidar ratios for evaluating CALIOP V4 aerosol properties. This can be done similar to Dawson et al. (2015), by stratifying CALIOP-SODA LR as a function of CALIOP V4 aerosol types and their assigned lidar ratio.

Although the retrievals presented here are limited to cloud-free atmospheric columns due to the constraint imposed by SODA AOD, it is possible to adapt the algorithm to make use of above-cloud satellite AOD retrievals (e.g., Jethva et al., 2014; Liu et al., 2015). In this regard, above-cloud AOD using CALIOP can be derived by combining the integrated attenuated backscatter and depolarization ratio (Hu et al., 2007; Liu et al., 2015), with corrections for the multiple-scattering depolarization relationship implemented by SODA (Deaconu et al., 2017). Efforts to retrieve above-cloud lidar ratio and extinction profiles over the southeast Atlantic using the above cloud AOD are currently underway (Ferrare et al., 2018).

CALIOP-SODA 1L retrievals are expected to perform better for relatively homogeneous atmospheric profiles characterized by a single aerosol type. Alternatively, SODA 2L retrievals are likely to be advantageous for specific regions where massive aerosol plumes from the continent are transported offshore at high altitudes through convective processes, in such a way that MBL aerosols are detached from the layer above and the assumption MBL LR=25 sr (maritime) is a good approximation. This is probably the case over the southeast Atlantic during the biomass burning season or for episodic dust transport over the tropical Atlantic. However, the CALIPSO Science Team product will continue providing the best available global dataset for monitoring complex aerosol profiles, continental processes, and aerosols in the upper troposphere.

Data availability. CALIPSO version 4.1 is available at https://eosweb.larc.nasa.gov, and SODA aerosol optical depth at http://www.icare.univ-lille1.fr.

Competing interests. The authors declare that they have no conflict of interest.

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Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

Atmospheric Measurement Techniques Discussions

Figures

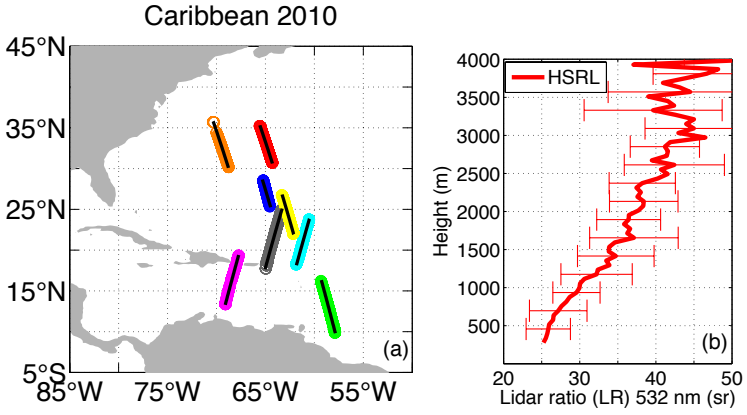

Figure 1: a) Flight tracks during the 2010 field campaign. Black solid lines correspond to the matched CALIPSO tracks. b) Mean HSRL lidar ratio (532 nm) as a function of altitude and one standard deviation (error bar) for all the flight tracks in Fig. 1a.

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
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Atmospheric Measurement Techniques Discussions

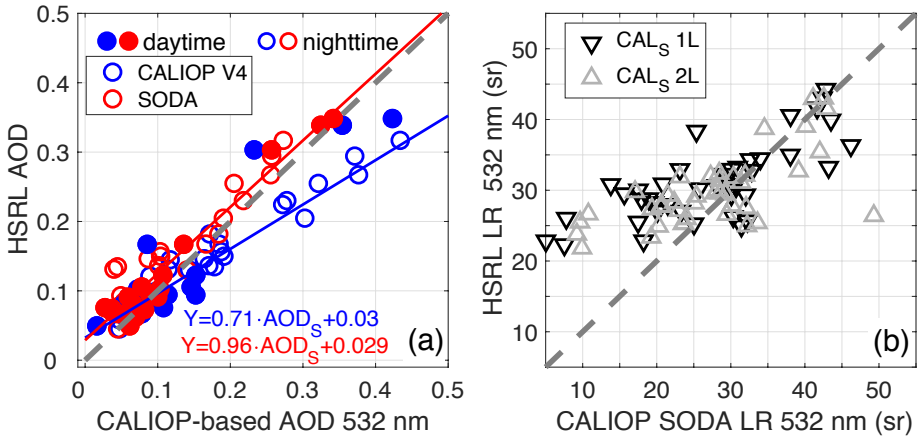

Figure 2: a) Scatterplot between SODA (red) and CALIPSO V4 (blue) against HSRL AOD at 532 nm. Filled and open circles indicate daytime and nighttime observations, respectively. Blue and red lines (and equations) are the linear fit for V4 and SODA AOD (AOD_{v4} and AOD_S) relative to HSRL. b) Comparison between CALIPSO SODA (CAL_S) lidar ratio based on the 1-layer (1L) and 2-layer (2L) assumption with the HSRL column-effective lidar ratio from Eq. 4 (black and gray symbols, respectively). Gray dashed line is the one-to-one relationship.

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

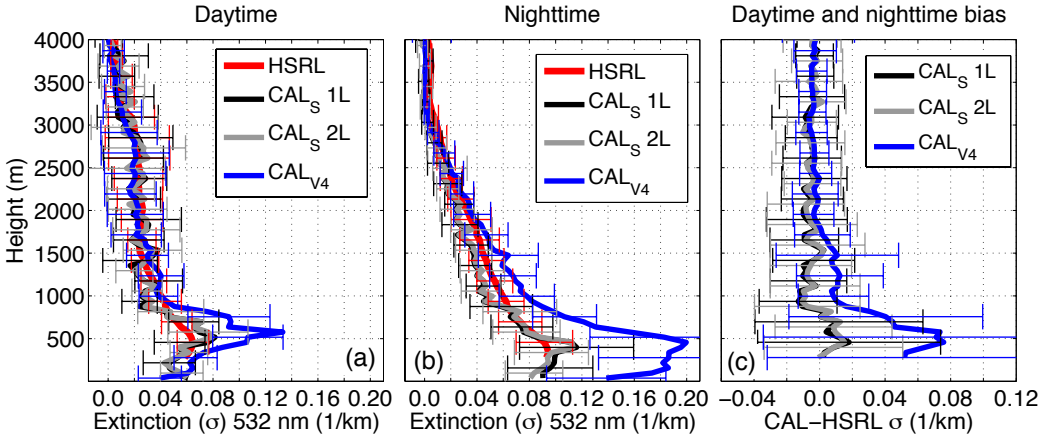

Figure 3: Mean aerosol extinction coefficient profile from the HSRL (red), CALIPSO SODA 1L (black), 2L (gray), and CALIPSO standard V4 product (blue) during a) daytime and b) nighttime. c) Total mean bias of CALIPSO-based extinction relative to the HSRL: CALIPSO SODA 1L (black) and 2L (gray), CALIPSO V4 (blue). Error bars in Fig. 3 a and b denote one standard deviation, and RMSE in Fig. 3c.

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

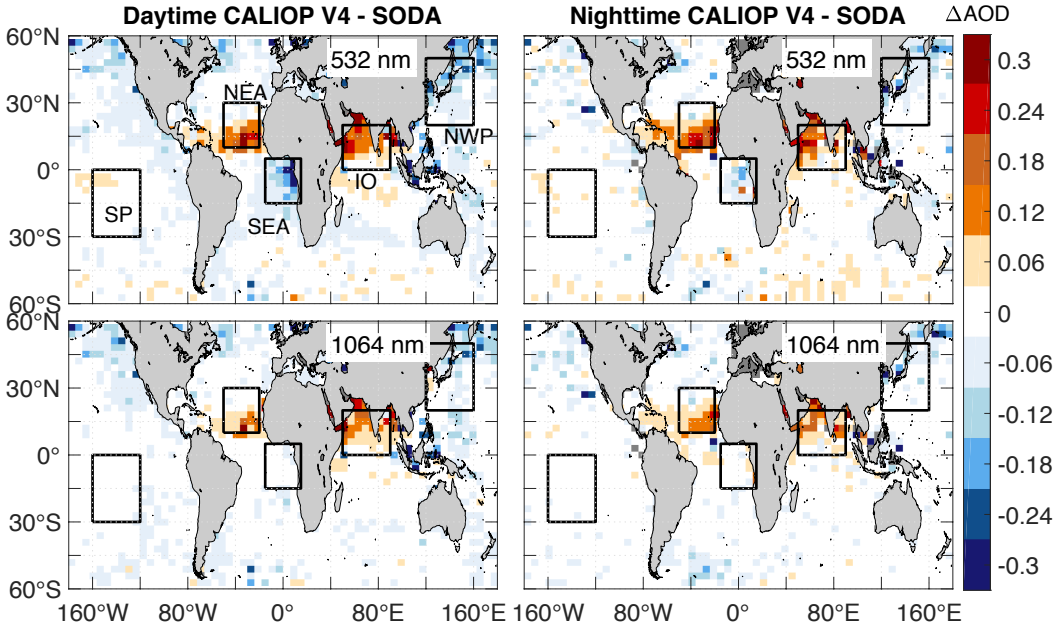

Figure 4: Mean AOD difference between CALIOP V4 and SODA for five months of 2010 for daytime (left) and nighttime (right), and the 532 nm (upper panels) and 1064 nm (lower panels) channels. Boxes denote specific regions in which the extinction coefficient profiles are further compared in Figure 5: South Pacific (SP), southeast Atlantic (SEA), Indian Ocean (IO), northeast Atlantic (NEA), and northwest Pacific (NWP).

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

Atmospheric Measurement Techniques Discussions

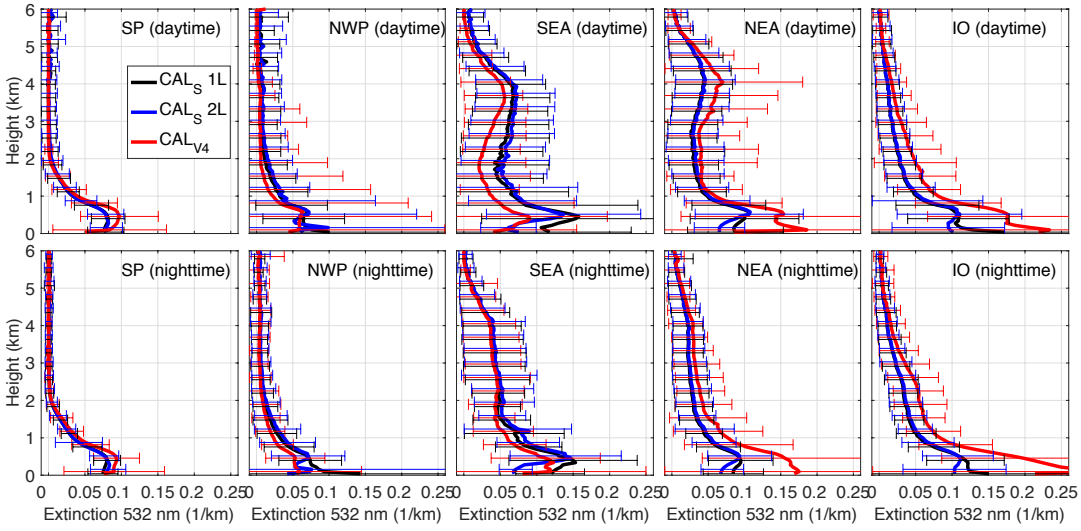

Figure 5: Mean aerosol extinction coefficient at 532 nm for the five regions defined in Fig. 4. Upper and lower panels correspond to daytime and nighttime retrievals. CALIPSO-SODA profiles are in black (1L) and blue (2L), and CALIPSO V4 is in red.

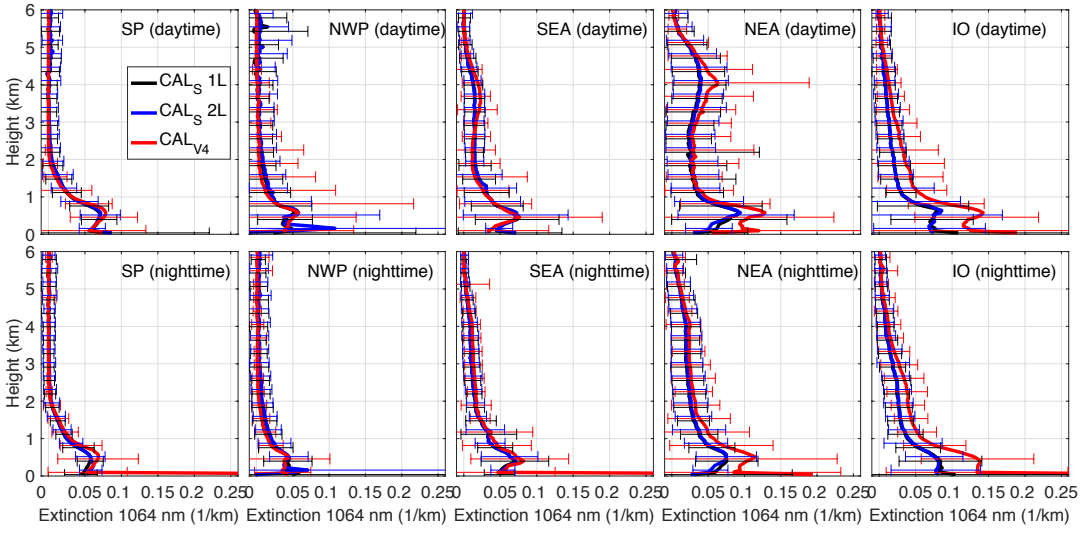

Figure 6: As in Figure 5 but for the 1064 nm channel.

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

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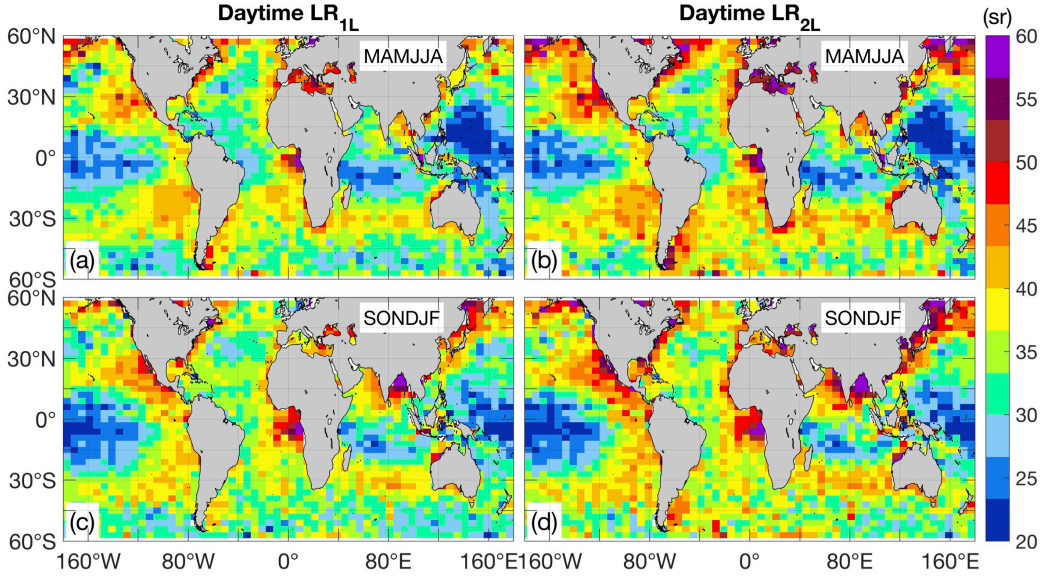
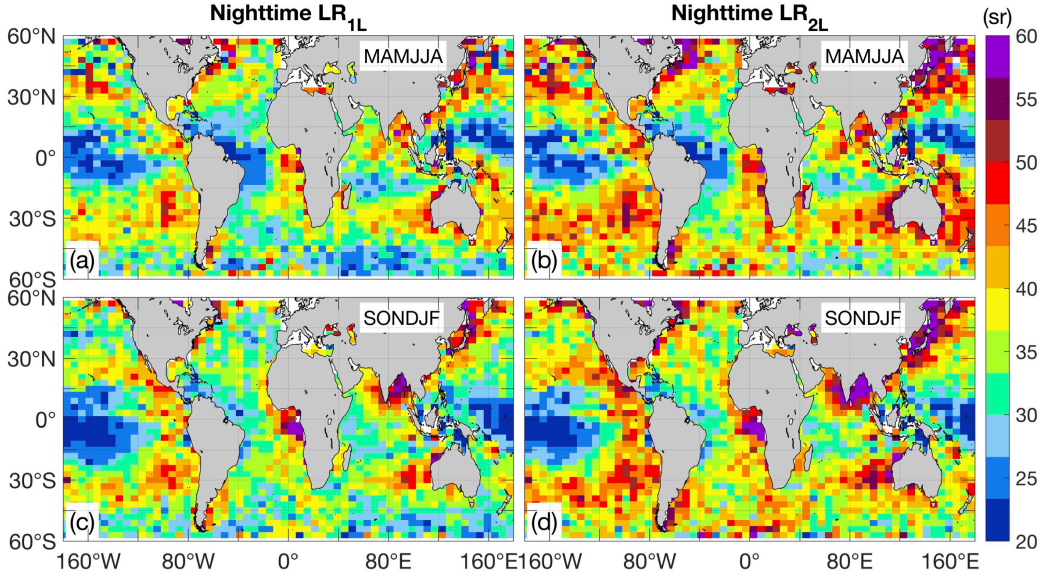


2

3 Figure 7: Semi-annual daytime 532 nm lidar ratios. a) LR_{1L} for spring-summer, b) LR_{2L} for
4 spring-summer, c) LR_{1L} for autumn-winter, and d) LR_{2L} for autumn-winter.

5



6

7 Figure 8: As in Figure 7 but for nighttime.

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Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

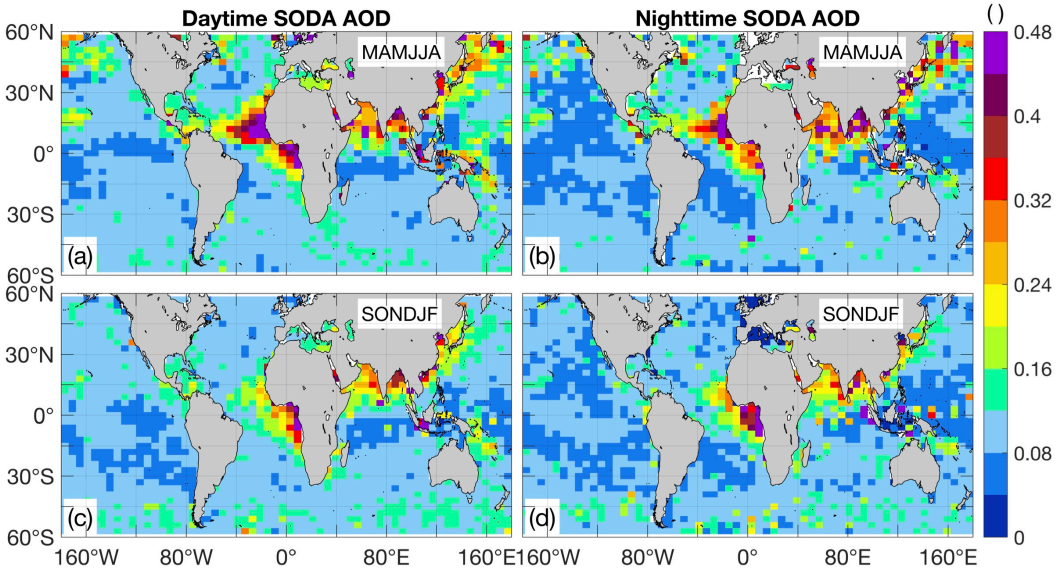

Figure 9: SODA AOD for daytime (a and c) and nighttime (b and d), spring-summer (MAMJJA) and autumn-winter (SONDJF).

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

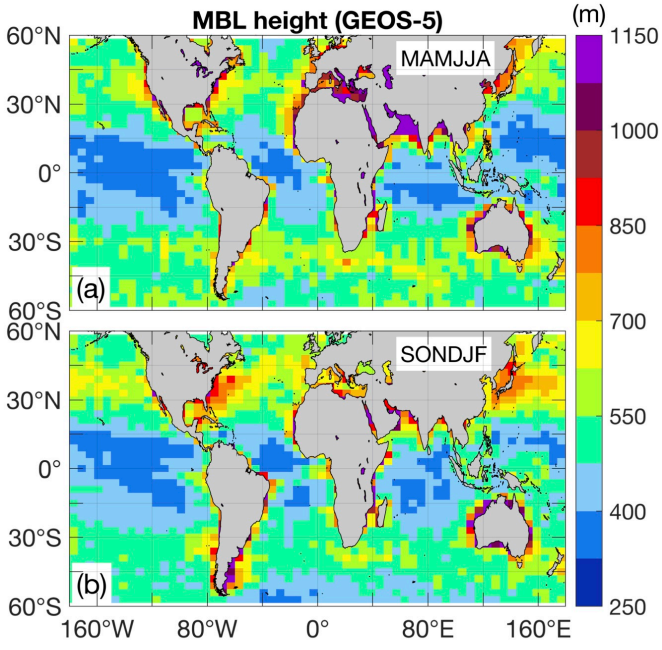

Figure 10: Daytime marine boundary layer height for a) spring-summer, and b) autumn-winter.

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Discussion started: 6 November 2018

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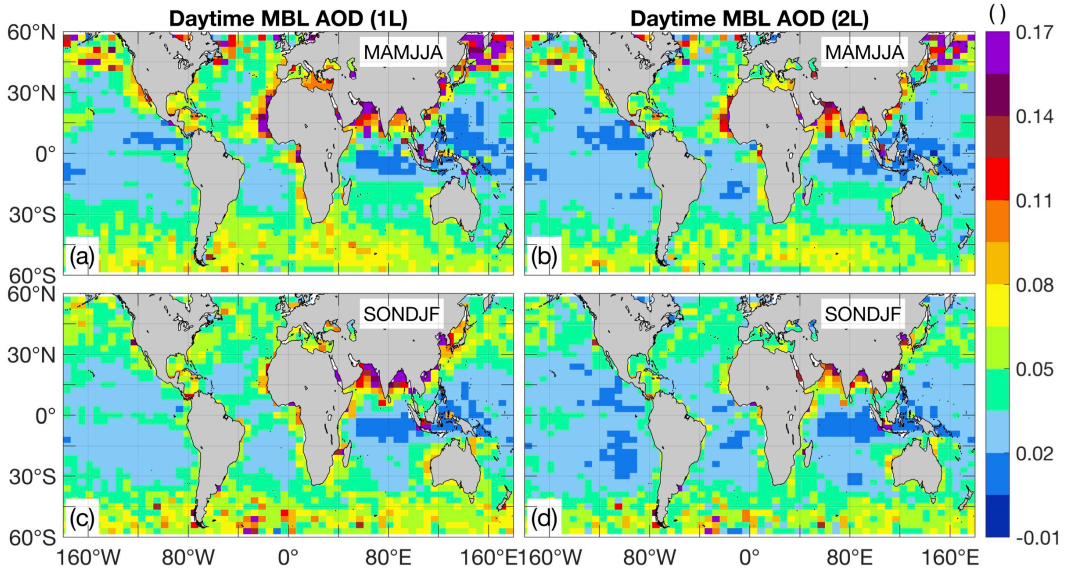


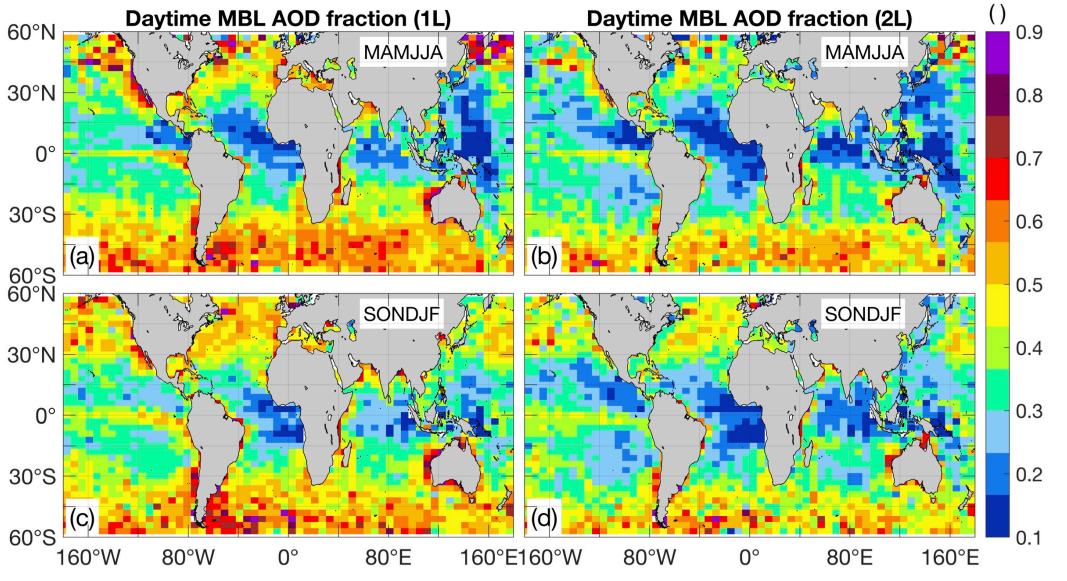
2

3 Figure 11: Daytime MBL 532 nm AOD based on 1L (left) and 2L (right).

4

5

6



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Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

Atmospheric Measurement Techniques Discussions

Figure 12: Fraction of daytime AOD contributed by the marine boundary layer.

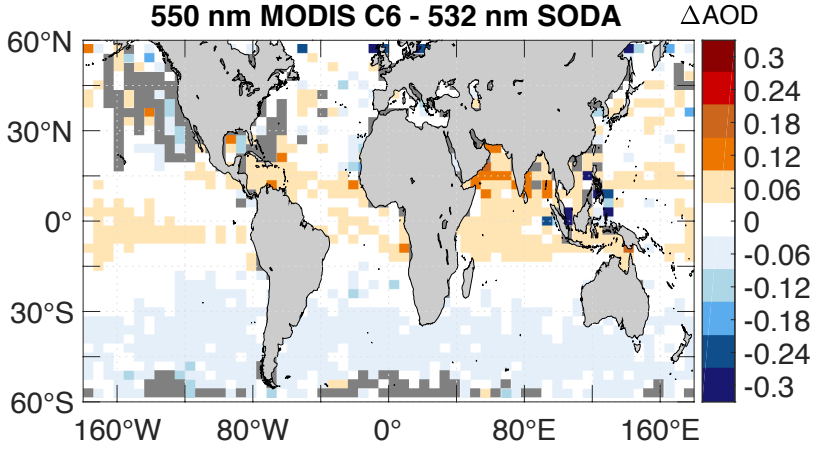

Figure 13: Mean AOD difference between matched 550 nm MODIS C6 and 532nm SODA daytime AOD for five months of 2010.

Atmos. Meas. Tech. Discuss., https://doi.org/10.5194/amt-2018-363
Manuscript under review for journal Atmos. Meas. Tech.
Discussion started: 6 November 2018

Atmospheric Measurement Techniques Discussions

Tables

Table 1: Linear correlation coefficient (*r*), mean bias, and RSME between HSRL and SODA and CALIOP Standard V4 AOD. Percentages are calculated relative to the mean HSRL AOD.

CALIOP-based AOD	***r***	**Mean bias**	**RSME**
SODA	0.96	-0.024 (-17%)	0.035 (24.2%)
Standard V4	0.94	0.014 (10%)	0.044 (31.2%)

Table 2: As in Table 1 but for CALIOP-SODA lidar ratio

CALIOP SODA LR	***r***	**Mean bias**	**RSME**
1 layer (1L)	0.67	-2.4 sr (-8.1%)	7.4 sr (24.8%)
2 layer (2L)	0.74	-3.9 sr (-12.5%)	8.1 sr (26.0%)

Table 3: As in Table 1 but for V4 and SODA aerosol extinction coefficient in the lower troposphere (below 3.0 km).

CALIOP-based extinction	***r***	**Mean bias**	**RMSE**
CALIOP V4	0.82	0.013 km^{-1} (33.0%)	0.043 km^{-1} (106.0%)
SODA 1 layer (1L)	0.78	-0.0037 km^{-1} (-9.2%)	0.028 km^{-1} (72.6%)
SODA 2 layer (2L)	0.79	-0.0029 km^{-1} (-7.0%)	0.028 km^{-1} (73.8%)