Satellite-derived direct radiative effect of biomass smoke over broken low clouds

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Smoke from biomass burning exerts a radiative forcing on climate by reflecting and absorbing solar radiation. For a given aerosol type and surface albedo, the combined effects produce either a net cooling or a net warming. Here, we use a new, satellite-based approach to quantify the top-of-atmosphere direct radiative effect of aerosol layers advected over the partly cloudy boundary layer in the region of the southeastern Atlantic Ocean during July-October, 2006-2007. We demonstrate that the radiative forcing efficiency at top-of-atmosphere is primarily controlled by the fractional area coverage of underlying clouds. This relationship is nearly linear such that it is possible to define a critical cloud fraction at which the radiative forcing efficiency changes sign. For this region and time period, critical cloud fraction is about 0.4, with strong sensitivity to aerosol single scattering albedo and underlying cloud albedo. The regional-mean direct radiative effect is three times higher when spatial covariation between cloud cover and aerosol loading is taken into account. These results demonstrate the importance of cloud prediction for accurate quantification of aerosol direct effects.
Biomass burning aerosols make a significant but poorly quantified contribution to anthropogenic radiative forcing of climate\(^1,2,3\) and may affect regional atmospheric circulation\(^4\). The most significant differences between model estimates of the top-of-atmosphere (TOA) direct climate forcing (DCF) are in regions where biomass burning aerosol dominates the forcing\(^3\). The DCF is the change in top of atmosphere direct radiative effect (DRE\(_{\text{toa}}\)) since pre-industrial times and cannot be determined from modern measurements alone\(^5\). Both DRE\(_{\text{toa}}\) and the absorption within the atmosphere DRE\(_{\text{atm}}\) are sensitive to both aerosol optical properties (chiefly aerosol optical thickness \(\tau\), absorption and size distribution)\(^6\), and also to the albedo of the underlying surface\(^7,8\).

In the absence of clouds, DRE is negative over the ocean due to its low surface albedo even when the aerosol is strongly absorbing\(^9\). However, when absorbing aerosol layers are located above clouds, DRE\(_{\text{toa}}\) can be positive\(^2,10\). While a few modeling studies have attempted to quantify the regional effects of clouds on DRE, e.g.,\(^10\), for the first time we use spaceborne lidar observations of aerosols above clouds together with observed cloud optical properties to quantify the aerosol DRE and the effects of clouds upon it.

Previous intensive observational studies of biomass burning aerosol conducted during field campaigns over North and South America\(^11,12,13\), and over Africa\(^14,15\) are limited either in time or space. Passive remote sensing of aerosol optical properties is routinely conducted at numerous surface sites around the globe (e.g. AERONET project \(^16\)) and from satellites\(^17,18,19\), but such approaches fail or are highly biased in presence of clouds\(^18,20\), which severely limits our ability to quantify the radiative effects of aerosols in regions where aerosols are advected over low level clouds.
Here, we quantify the optical depth $\tau$ and Angstrom exponent $\alpha$ of aerosol layers overlying optically thick clouds over the southern Atlantic Ocean using spaceborne lidar observations from ‘Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation’ (CALIPSO) using the retrieval method by Chand et al.\textsuperscript{21}. An elevated layer is here defined as being a layer with a detectable optical thickness overlying a strongly attenuating cloud that has a top below 3 km, although over much of the domain the low cloud top height is significantly lower than this (see supplementary Figure 1 on the heights of the cloud and elevated aerosol layers). For the elevated aerosol layers we use the CALIPSO layer identification algorithm\textsuperscript{21} to determine the aerosol layer top and base height. Most elevated aerosol layer top heights fall between 2.5-5.5 km and have a mean thickness of approximately 2 km. Most of the clouds (86 \%) are observed below 3 km over the entire domain. The mode of the cloud base and top of these low level clouds are about 0.7 km and 1.3 km, respectively. If the uppermost cloud layer is above 3 km the aerosol optical thickness is assumed to be zero for the purposes of the radiative transfer calculations. Other data-selection details are given in the Method section and in Chand et al.\textsuperscript{21}.

Figure 1 shows an example of the vertical and along-track structure of clouds and elevated aerosol layers during a night time CALIPSO pass in August 2006. Our aerosol retrieval algorithm indicates that the $\tau$ of the elevated aerosol layers is in places as high as 1.5. The lower detection limit of the $\tau$ retrieval is estimated to be 0.07. Data from each month (Jul-Oct) for years 2006 and 2007 are integrated to obtain a seasonal average over the Atlantic Ocean (7.5°N-22.5°S, 17.5°E-27.5°W).
We use the DISORT radiative transfer model (RTM, see Methods section) to estimate the DRE of elevated aerosol layers overlying clouds and for clear sky conditions over land and ocean. The model inputs are the optical properties (\(\tau\), single scattering albedo \(\varpi\), \(\lambda\), and asymmetry factor \(g\)) and geometrical properties (height, thickness) of the elevated aerosol layer, and the albedo of the surface underlying the aerosol layer (either the cloud or surface albedo).

For the RTM we use \(\tau\) and \(\lambda\) from CALIPSO using a newly developed, above-cloud retrieval (see Methods and reference 21). \(\tau\) is retrieved at 532 nm and \(\lambda\) applies to the wavelength dependence of \(\tau\) between 532 and 1064 nm. The model results depend quite strongly upon \(\varpi\) (550 nm), which cannot currently be determined from spaceborne observations\(^2\). Here we use \(\varpi = 0.85 \pm 0.02\) (regional mean and uncertainty) based on an updated synthesis of remote and in-situ measurements during the Southern African Regional Science Initiative 2000 (SAFARI 2000). For reasons discussed elsewhere,\(^2\) we consider this range of values more reliable than the value of 0.90 derived by Haywood et al.\(^2\) during this same campaign. We set \(g = 0.62 \pm 0.03\) (550 nm), consistent with size distributions of lab-generated and field observed biomass burning aerosols over Southern Africa\(^2\) and South America (Chand et al., unpublished data and reference 24).

The regional distribution of mean \(\tau\) (Fig 2a) clearly indicates that elevated aerosol layers of significant optical thickness are present over the southern Atlantic at distances of well over 2000 km from the South African coast, consistent with advection by the mean flow from the continental biomass burning sources\(^1,2\). Much of the advection appears to be in a zonal direction consistent with the predominantly easterly winds at 600 hPa north of 15°S. Weak meridional advection confines most of the aerosols to south of
The diurnal and seasonal (July-October) mean DRE of the elevated aerosol layers in terms of their impact on total atmospheric column absorption (DRE\textsubscript{atm}, Fig. 2b) and at the TOA (DRE\textsubscript{toa}, Fig. 2c), demonstrate a major impact of elevated aerosols upon the radiative budget of the atmosphere and the climate system. Note that DRE\textsubscript{toa} changes sign reflecting the geographic variability in the underlying cloud fractional coverage $C$. This moves the region of stongest positive DRE\textsubscript{toa} some 5° or so southward from the region with maximum $\tau$ towards the region with the maximum $C$. Conversely, the DRE\textsubscript{toa} just south of the equator over the ocean is reduced because the cloud fraction there is much lower. These results clearly show that the pattern of cloud cover variability beneath the aerosol layers has a first order impact upon the regional distribution of aerosol radiative forcing.

The impact of the underlying cloud is more clearly demonstrated by examination of the radiative forcing efficiency ($\text{RFE}_{\text{toa}}=\text{DRE}_{\text{toa}}/\tau$) of the elevated aerosol layers (Figure 3). There is a remarkably strong correlation between $\text{RFE}_{\text{toa}}$ and the monthly mean value of $C$ ($r^2=0.96$) implying that the cloud fractional coverage is an excellent predictor of the mean RFE in a particular region on a monthly timescale. The $\text{RFE}_{\text{toa}}$ for clear sky conditions is inferred to be -34 W m\textsuperscript{-2} $\tau$\textsuperscript{-1}, whereas the mean value for cloudy sky is 52 W m\textsuperscript{-2} $\tau$\textsuperscript{-1}, indicating an average increase in $\text{RFE}_{\text{toa}}$ of 0.86 W m\textsuperscript{-2} per unit $\tau$ for 1% increase in cloud cover. The critical cloud fraction $C_{\text{crit}}$, for which DRE\textsubscript{toa} changes sign, is 0.40. Based on the Terra data used herein, the average cloud coverage over this region is 0.48, leading to a positive estimate for DRE\textsubscript{toa} (2.4 W m\textsuperscript{-2}) for the region as a whole. Importantly, this is three times as large as that (0.8 W m\textsuperscript{-2}) obtained by assuming that the spatial pattern of $\tau$ (whose seasonal-regional mean value is 0.11) is independent.
of that in $C$, emphasizing the importance not only of the mean cloud fractional coverage but also its spatial distribution with respect to the overlying aerosol layers. There is a tendency for regions with optically thick aerosol layers to be those with a fractional coverage of low clouds exceeding that for the domain as a whole. Thus knowledge of the domain mean cloud cover and mean aerosol optical thickness is insufficient to determine the regional mean DRE, and the covariance between the two must be considered. This is a stringent challenge for global climate models which exhibit considerable deficiencies in their ability to represent the correct optical properties of both clouds and aerosols.

The value of $C_{\text{crit}}$ derived herein (0.40) is sensitive to uncertainties in aerosol optical properties (see Supplementary Figure 3). Increasing $\varpi$ over its uncertainty range of 0.83 to 0.87 leads to an increase of almost 0.1 in $C_{\text{crit}}$ (from 0.37 to 0.47). This constitutes the greatest source of explicitly estimated error in our study. Changing $\tilde{a}$ and $g$ over their uncertainty ranges (1.1-2.1 and 0.59-0.65, respectively) causes $C_{\text{crit}}$ to vary by 0.04 in each case. In contrast, shifting cloud altitude by 1.25 km had only a minor effect on $C_{\text{crit}}$ of 0.01.

Podgorny and Ramanathan used an RT model to estimate the top-of-atmosphere radiative effect of absorbing aerosols over broken low clouds in the Indian Ocean. Assuming thick clouds and moderately absorbing aerosol ($\varpi=0.90$ at 500 nm), they found a similar strong dependence upon cloud fraction, but a lower value of $C_{\text{crit}}$ (0.25) than found herein (0.40). The explanation for this difference appears to be cloud albedo, which is calculated from cloud optical depth within the RT models of both studies. In our study, cloud albedos are $0.50 \pm 0.06$ (mean and standard deviation) based on MODIS-retrieved cloud optical depths of 7.8 $\pm$ 1.8. The calculations by Podgorny and
Ramanathan assumed a cloud optical depth of 15, implying much higher cloud albedo (close to 0.7). This would cause aerosol over cloud have a much stronger warming effect, lowering the balance point, $C_{crit}$. We see from this comparison that accurate knowledge of cloud albedo (in addition to $C$, $\tau$, and $\varpi$) is critical to the accurate determination of aerosol radiative forcing.

Small scale or day-to-day covariation among aerosol and underlying cloud properties (assumed herein to be zero; see Methods section) constitutes an additional, unknown source of error in our estimates. A recent study conducted in the same region of the SE Atlantic demonstrates covariation of $\tau$ and $C$. However, this finding relied on retrieving both quantities from MODIS, and the authors were careful to point out that the cause could be either a physical relationship or an instrumental artifact. If physical, such covariation would tend to increase the warming effect of the aerosol and lower $C_{crit}$.

Covariations among other combinations of key properties ($C$, cloud albedo, $\tau$, and $\varpi$) could also be important and should be investigated in future studies.

A recent comparison of global models showed that the southeast Atlantic region exhibits extremely large inter-model differences in all-sky direct radiative forcing. Even in the annual mean, the modeled values of radiative forcing over this region vary from -1 to +2 W m$^{-2}$. These estimates are poorly constrained by traditional aerosol retrievals from passive remote sensing since these methods are restricted to clear-sky situations. As shown herein, above-cloud aerosol retrievals from CALIPSO combined with cloud retrievals from passive satellites provide a powerful new set of tools for adjudicating among the discordant model estimates and, ultimately, improving understanding of aerosol radiative forcing and its dependence upon underlying clouds.
References


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Method
Based on the active remote sensing observations from the 532 nm and 1064 nm channels on Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO), we developed a new method to quantify the aerosols optical depth (τ) and Ångstrom exponent (å) of aerosol layers above clouds\(^{26}\). Here we applied this technique regionally over the southern Atlantic Ocean (7.5N-22.5S, 17.5E-27.5W) to produce monthly mean τ and å estimates at 5°×5° resolution for two biomass burning seasons (July-October of 2006 and 2007). Data from the Moderate Resolution Imaging Spectroradiometer (MODIS) on the NASA Terra satellite are used to provide monthly mean estimates of ice-cloud fractional coverage (used as a data selection screen), water-cloud fractional coverage \(C\), and, water-cloud visible albedo, where the latter derives from water-cloud optical thickness retrievals\(^{29}\) and two-stream radiative transfer theory (Coakley, J.A., and P. Chýlek J. Atmos. Sci., 32, pp 409, 1975). Cloud data from MODIS provide essentially complete coverage of each 5°×5° blocks every day; however, the daily, spatial coverage by CALIPSO is much lower. For this reason, we use both daytime and nighttime CALIPSO data and we aggregate them to monthly averages for each 5°×5° blocks prior to combining them with MODIS data to calculate direct radiative forcing. Thus, our method assumes (i) that above-cloud aerosol in this region does not exhibit significant diurnal variation, (ii) that the above-cloud aerosols detected by CALIPSO along its orbit are representative of the entire 5°×5° block, and (iii) that there is no day-to-day co-variation of cloud fraction or properties with above-cloud aerosol properties. These are reasonable assumptions but should be tested in follow-up studies.
The direct radiative forcing efficiency (DRE) of elevated aerosol layers is calculated using the DISORT radiative transfer code. Calculations are performed for each 5x5 degree block at monthly-mean resolution. Because we do not explicitly model the effects of cirrus clouds, blocks where ice-cloud fraction is greater than 5% were excluded from the analysis. This excludes about 40% of the available blocks. Calculations run without this screening are noisier (e.g. the correlation between DRE and $C$ is somewhat lower than indicated in Figure 3) but yield almost identical values of domain-mean DRE and $C_{crit}$ (see supplementary figure 4). We perform separate calculations for aerosols in a clear sky situation (assuming an ocean surface albedo of 0.06 and a land surface albedo 0.25) and above a cloudy surface having the observationally-determined mean albedo. We apply the mean cloud and aerosol heights observed by CALIPSO over the entire domain. We perform additional calculations to determine the sensitivity of domain-mean DRE and $C_{crit}$ to the fixed values of $\bar{\sigma}$, $g$, $\dot{a}$, cloud height, and aerosol height (see the supplementary figures). These calculations indicate that DRE and $C_{crit}$ are most sensitive to $\bar{\sigma}$ followed by $g$ and $\dot{a}$. As long as the aerosol layer is separated from the cloud layer by at least a few hundred meters, changes in cloud or aerosol height have negligible effects.
Figure 1 Profiles of 532nm backscatter return signal from the CALIPSO lidar showing the vertical distribution of aerosols and underneath clouds. Aerosols optical thickness (AOT) is shown by the plate at right top corner. Strong backscattering (>0.001 sr\(^{-1}\) km\(^{-1}\)) is associated with aerosol and/or cloud layers (as indicated by arrows). Observations from 2006 and 2007 indicate that such events are very frequent during the biomass burning season (particularly August and September) over the west coast of Africa between the equator and 20°S.
Figure 2 Regional distributions of aerosols and its radiative impacts with winds and cloud fraction. Maps showing seasonal (July-October, 2006/2007) mean values of: (a) aerosol optical thickness \( \tau \) (including zeros when no elevated aerosol layer is detected), and National Centers for Environmental Prediction (NCEP) winds at 600 hPa; (b) Atmospheric direct radiative effect (DRE\(_{\text{atm}}\); column absorption); (c) Direct radiative effect at top of the atmosphere (DRE\(_{\text{toa}}\) shown by colors and cloud fraction shown by contour lines); and (d) the direct radiative forcing efficiency, RFE (RFE=DRE\(_{\text{toa}}\)/\( \tau \)). The \( \tau \) and cloud fraction are retrieved from CALIPSO satellite and MODIS-Terra satellite, respectively.
Figure 3 Correlation of aerosol direct radiative forcing efficiency (RFE) with cloud fraction. Radiative forcing efficiency at top of the atmosphere (squares), and within the atmosphere (circles) as a function of cloud fraction. \( C_{\text{crit}} \) is the cloud fraction when the top of atmosphere (TOA) RFE changes sign.

July-October, 2006-2007

<table>
<thead>
<tr>
<th>CF(_{\text{ice}}) &lt;= 0.05</th>
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<tbody>
<tr>
<td>ssa = 0.85</td>
</tr>
<tr>
<td>g = 0.62</td>
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<tr>
<td>N = 337</td>
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TOA

\[ a = -34.86 \]
\[ b = 86.12 \]
\[ r^2 = 0.96 \]

Atmosphere

\[ a = 59.30 \]
\[ b = 31.97 \]
\[ r^2 = 0.51 \]
Supplementary Figure 1 Frequency distribution of aerosol and underneath clouds.

Histograms of the top and bottom altitudes of aerosol layers (a, b) and cloud layers (c, d).
Supplementary Figure 2 Atmospheric radiative forcing by ‘above-cloud’ aerosols for clear, cloudy and all sky conditions. Scatter plots of aerosols optical depth (AOD) w.r.t. radiative forcing at the top of the atmosphere (TOA), bottom of the atmosphere (BOA) and within the atmosphere (TOA-BOA) for (A) clear sky conditions (A); (B) cloudy sky conditions; and (C) all sky conditions. The text in the right side boxes shows the intercepts, slopes and correlation coefficient ($r^2$) of the regression lines. The radiative model is run using angstrom exponent (AE)=1.6, single scattering albedo (ssa)=0.85 and asymmetry factor (g)=0.62; as shown on the top right corners of the respective plates. The equations shown at the bottom of the panel (C) are used to get the radiative forcing at TOA and BOA in all sky conditions.
Supplementary Figure 3. Sensitivity of aerosol direct radiative forcing efficiency to changes in aerosol and cloud properties. Scatter plots of water cloud fraction with respect to direct radiative forcing efficiency (RFE) showing sensitivity at the top of the atmosphere (TOA) and within the atmosphere. The sensitivity is derived by varying (a) single scattering albedo (ssa); (b) angstrom exponents (AE); (c) asymmetry factors (g); and (d) cloud top altitude (Zcloud). The vertical arrows show the impact on critical water cloud fraction $C_{crit}$ at the top of the atmosphere when the parameter in equation is changed. In each plate the lower two regressions are for RFE at TOA and the upper two are for the RFE within the atmosphere.
Supplementary Figure 4. Correlation of aerosol direct radiative forcing efficiency (RFE) with liquid water cloud and cirrus (ice) cloud fraction. Scatter plots of water cloud fraction with respect to direct radiative forcing efficiency (RFE) at the top of the atmosphere (TOA) and within the atmosphere for different data screening criteria based on ice cloud fraction $\text{CF}_{\text{ice}}$: (a) $\text{CF}_{\text{ice}} \leq 1$, (b) $\text{CF}_{\text{ice}} \leq 0.2$, (c) $\text{CF}_{\text{ice}} \leq 0.1$ and (d) $\text{CF}_{\text{ice}} \leq 0.05$. The text in the right sides boxes of the plates show the intercepts, slopes and correlation coefficients ($r^2$) of the regression lines. The radiative model is run using angstrom exponent (AE)=1.6, ssa=0.85 and asymmetry factor (g)=0.62; as shown on the top right corners of the respective plates. The value of ice cloud fraction ($\text{CF}_{\text{ice}}$) used for the screening and number of data points (N) are also shown on the top-left corners of the plates.