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**An assessment of
regional variations in
aerosol indirect
effects**

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A six year satellite-based assessment of the regional variations in aerosol indirect effects

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Received: 29 September 2008 – Accepted: 7 October 2008 – Published: 5 December 2008

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Since aerosols act as cloud condensation nuclei (CCN) for cloud water droplets, changes in aerosol concentrations having significant impacts on the corresponding cloud properties. An increase in aerosol concentration leads to an increase in CCN, with an associated decrease in cloud droplet size for a given cloud liquid water content. Smaller droplet sizes may then lead to a reduction in precipitation efficiency and an increase in cloud lifetimes, which induces more reflection of solar radiation back into space, cooling the atmosphere below the cloud layer. In reality, this relationship is much more complex and is interrelated between aerosol, cloud, and atmospheric conditions present at any one time. MODIS aerosol and cloud properties are combined with NCEP Reanalysis data for eight different regions around the globe between March 2000 and December 2005 to study the effects of different aerosol, cloud, and atmospheric conditions on the aerosol indirect effect (AIE). The first AIE for both anthropogenic and dust aerosols is calculated so that the importance of each can be compared. The unique aspect of this research is that it combines multiple satellite data sets over a six year period to provide a comprehensive analysis of indirect effects for different aerosol regimes around the globe.

Results show that in most regions, AIE has a distinct seasonal cycle, though the cycle varies in significance and period from region to region. In the Arabian Sea, the six-year mean anthropogenic+dust AIE is -0.4 Wm^{-2} and is greatest during the summer months ($< -2.0 \text{ Wm}^{-2}$) during which dust aerosol concentration is greatest, significant concentrations of anthropogenic aerosols are present, and upward vertical motion is also present providing a favorable environment for cloud formation. In the Bay of Bengal, AIE was negligible owing to less favorable atmospheric conditions and a lower concentration of aerosols. In the eastern North Atlantic, AIE was also small ($< 0.1 \text{ Wm}^{-2}$) and in this region dust aerosol concentration is much greater than the anthropogenic or sea salt components. However, elevated dust in this region may also absorb solar radiation and warm the atmosphere, stabilizing the atmosphere as evidenced by

ACPD

8, 20349–20397, 2008

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weak vertical motion during the summer (0.02 Pa s^{-1}) when AOT is greatest. Lower average cloud fraction compared to other regions allows the absorbing effect to offset the cooling effect associated with increasing CCN. The western Atlantic and Pacific oceans have large anthropogenic aerosol concentrations transported from the United States and China respectively and produce modest anthropogenic AIE ($0.7, 0.9 \text{ Wm}^{-2}$) in these regions as expected. Anthropogenic AIE was also present off the West African coast corresponding to aerosols produced from seasonal biomass burning. Interestingly, atmospheric conditions were not particularly favorable for cloud formation compared to the other regions during the times where AIE was observed. Overall, we are able to conclude that aerosol type, atmospheric conditions and their relative vertical distributions are a key factors as to whether or not significant AIE occurs and simple correlations between AOT and cloud properties are insufficient to explain the AIE.

1 Introduction

Each year, approximately 140 Tg of anthropogenic aerosols are injected into the atmosphere with 60% of these aerosols being sulfates produced from industrial pollution (Schulz et al., 2006). Since aerosols often reflect incoming solar radiation back into space, increasing aerosol optical thickness (AOT) leads to a cooling of the surface, which is known as the direct effect. Small-mode ($r_e < 0.25 \mu\text{m}$) and sulfate based aerosols (where r_e is the effective radius of an aerosol particle) often act as cloud condensation nuclei (CCN) for warm-process clouds (e.g. Quaas et al., 2004; Lohmann and Feichter, 2005; Penner et al., 2004). Both observations and modeling studies have shown that as anthropogenic aerosol concentration increases, the number of CCN increase accordingly, leading to a decrease in water cloud droplet size assuming a constant liquid water path through the cloud (e.g. Jones et al., 1994). Increasing the number of cloud droplets, increases cloud albedo, reflecting more solar radiation back into space. This is known as the first indirect, or the Twomey effect (Twomey, 1977; Kaufman and Fraser, 1997; Feingold, 2003). The decrease in droplet size has the addi-

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tional effect of delaying the onset of collision and coalescence in warm clouds, reducing precipitation efficiency and increasing the lifespan and possibly the areal coverage of the cloud, which is labeled as the second indirect effect (Albrecht, 1989; Quaas et al., 2004). Reducing precipitation efficiency also acts to increase water loading, leading to an increase in cloud liquid water path (LWP) and a corresponding increase in cloud thickness, complicating the identification of the Twomey effect in observations (Han et al., 1998; Reid et al., 1998; Schwartz et al., 2002). Both the first and second indirect effects act to cool the atmosphere, possibility offsetting warming due to greenhouse gases (Lohmann and Feichter, 2005). As a result, it is vital that accurate observations and interpretations of the indirect effects be made.

Previous studies have estimated the total aerosol cooling effect (direct and indirect) anywhere between -0.5 to -4.4 Wm^{-2} from anthropogenic aerosols (e.g. Boucher and Lohmann, 1995) though more recent research suggests that this value is likely closer -1.0 Wm^{-2} (Anderson et al., 2003; Lohmann and Feichter, 2005; Forster et al., 2007; Quaas et al., 2008). However, aerosol indirect effects are highly dependent on the aerosol species, their vertical distribution, and meteorological conditions present at the time (e.g. Patra et al., 2005; Matsui et al., 2006; Yuan et al., 2008). Total column aerosol optical thickness (AOT) and cloud properties are both retrieved by the MODIS instrument present onboard the Terra and Aqua EOS satellites thereby providing the opportunity for a detailed analysis of indirect effects from an observational perspective (Quaas et al., 2008). While using similar methods, this research analyzes indirect effects of both anthropogenic and dust aerosols over a much longer time span while also including analysis of corresponding atmospheric conditions. A novel feature of this work is the comparison of indirect effect to vertical velocity (e.g. Feingold, 2003), since the latter is very important to cloud formation. To the authors' knowledge, this relationship has not been fully explored using observations at a global scale.

Most previous research has emphasized quantifying the effect of anthropogenic (small-mode) aerosols on cloud properties (Lohmann and Feichter, 2005; Quaas et al., 2008). While these effects are important for climate change studies, the effects of

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5 other types of aerosols (which can also sometimes be anthropogenic in nature) must also be understood because the radiative forcing by an anthropogenic aerosol indirect effect also strongly depends on the natural background conditions (e.g. Bellouin et al., 2008). Traditionally small-mode, hygroscopic aerosols (e.g. sulfates produced from anthropogenic sources) are considered the most efficient CCN beyond naturally occurring sea salt (Jones et al., 1994; Li et al., 1996). Over the ocean, aerosol indirect effects are generally prevalent near locations of large aerosol sources, such as ship tracks and downwind of major pollution sources (Ackerman et al., 2003; Avey et al., 2007; Bennartz, 2007). Since the background aerosol concentration is low, the indirect effect caused by the addition of large concentrations of anthropogenic aerosols is most noticeable over the ocean (Lohmann and Lesins, 2003).

15 The influence of additional aerosol types, such as dust, on CCN and cloud characteristics has been less well documented. Jones and Christopher (2008) observed a positive relationship between total column AOT and cloud droplet effective radius in the Arabian during the northern hemisphere winter. Neither Jones and Christopher (2008) nor Yuan et al. (2008) could precisely determine the exact physical cause although Yuan et al. (2008) hypothesized that the increase in cloud droplet size with AOD could be related to slightly soluble organic particles and/or giant cloud condensation nuclei. Other possible causes include the changes in aerosol species and meteorological conditions that occur throughout the year in this region, or even a microphysical effect of aerosols on cloud liquid water path offsetting the decrease in droplet radius due to an increase in CCN. In the Arabian Sea, anthropogenic aerosols transported over the ocean from India account for the greatest proportion of total aerosol concentration during the winter months, while dust aerosols transported from the Arabian Peninsula in concert with increased sea-salt produced from higher wind speeds are predominant during the summer months (Ramanathan et al., 2001; Ramana and Ramanathan, 2006). Algorithm retrieval errors and sampling effects were found to be an unlikely source for positive aerosol concentration and cloud droplet size correlations (Yuan et al., 2008).

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Using 5 years of January data, Chylek et al. (2006) observed that cloud droplet radius decreases from south to north in the Indian Ocean (15° S to 25° N) corresponding to an increase in anthropogenic aerosol concentration. They considered the month of September as a “clean” case for comparison, despite the presence of large concentrations of dust and sea salt aerosols in the Arabian Sea, which were not accounted for in their analysis. Jones and Christopher (2008) observed that the indirect effect (defined by the inverse correlation between cloud droplet effective radius and AOT) was largest during the summer months, when dust aerosols comprise the largest portion of the total AOT. Dust aerosols are not normally considered as good CCN, but mineral dust has been observed to be effective CCN when coated with anthropogenic aerosols (Levin et al., 1996; Satheesh et al., 2006). Another uncertainty in the indirect effect of dust aerosols is the meteorological conditions that surround the dust plumes. Dust outbreaks in the North Atlantic are also associated with an influx of dry air in the low to mid-levels of the atmosphere with corresponding increases in atmospheric stability (Dunion and Veldon, 2004). Conversely, increased dust aerosol concentrations in the Arabian Sea correspond with the Indian monsoon season where greater amounts of moisture and atmospheric forcing are available to form clouds and precipitation. Given these conditions, greater cloud cover and larger water droplets would be expected (Jones and Christopher, 2008). Thus, any aerosol indirect effects present could be outweighed by the meteorological conditions in which they exist. Part of this research effort will examine the complex interaction between changes in aerosol concentrations vs. humidity and stability conditions to the aerosol indirect effect.

Understanding the changes in aerosol concentrations and the species of those aerosols are key to understanding the relative importance of the indirect effects on the climate. Seasonal changes in aerosol characteristics occur throughout the globe, with the Arabian Sea being a prime example of this change. Previous studies have often focused on analyzing the indirect effect on very small temporal and spatial scales (e.g. Reid et al., 1999) or on a globally averaged scale (e.g. Matsui et al., 2006) often without comparing the effect due to changes in aerosol properties. In both cases, the

indirect effects due to changes in aerosol type and meteorology over long periods of time on a regional scale are generally overlooked.

To evaluate the changes in the aerosol indirect effect as a function of different aerosol species and atmospheric conditions, we selected eight $10 \times 10^\circ$ regions over the ocean, each with a predominant aerosol type (Fig. 1). The North-East Atlantic Ocean (EA) and Arabian Sea (AS) are selected to study the effects of dust aerosols. Large concentrations of dust exist in both regions during the summer months providing the opportunity to analyze their effects on cloud properties. Anthropogenic aerosol from pollution sources are located nearly year-round in the Bay of Bengal (BB), the off the east coasts of China (West Pacific, WP) and the United States (North-West Atlantic, WA). They are also the dominant aerosol type in the Arabian Sea during the winter months. Large concentrations of carbon-based anthropogenic aerosols are present in the South Atlantic off South-East Africa (AF) resulting from biomass burning on the African continent, and we include this region in our analysis. Finally, the relatively pristine regions in the far South Atlantic (SA) and Indian Oceans (IO) that primarily comprise of maritime sea salt are examined to assess possible indirect effects when aerosol concentrations are low.

The aerosol indirect effects due to fine mode (primarily anthropogenic) and dust aerosols for each region are calculated using daily Terra Moderate Resolution Imaging Spectroradiometer (MODIS) aerosol and cloud product over a 6-year period (2000–2006) contained within the Clouds and the Earth's Radiant Energy System (CERES) data in the CERES Single Scanner Footprint (SSF) product. Using this long-term data set, it will be possible not only to compare indirect effects between regions, but also how these effects change as a function of time. Each region has its own unique aerosol and meteorological properties, which are key to understanding the importance of the indirect effect in each region. For comparison, anthropogenic and dust aerosol direct radiative effects are also calculated to provide a comprehensive overview of the aerosol-cloud radiative effects for each region. Using Multi-Angle Imaging Spectroradiometer (MISR) Stereo Height data and Total Ozone Mapping Spectrometer (TOMS) aerosol index (AI), examples of the importance of the vertical profiles of both aerosols

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and clouds relative to the indirect effect are examined. Indirect effects are shown to be highly dependent on the local atmospheric conditions, aerosol type and vertical distributions, with total column aerosol concentrations being a secondary factor in some instances.

2 Data

2.1 Cloud properties

The Clouds and Earth's Radiant Energy System (CERES) Single Scanner Footprint (SSF) FM1, Edition 2B data between March 2000 and November 2005 from the Terra satellite (on a sun-synchronous orbit with an equator-crossing local time of about 10:30 a.m.) were collected for the eight 10×10 degree regions over the global oceans (Fig. 1). Each region represents a unique aerosol – climate regime where aerosol indirect effects are likely to differ. The eight regions chosen are the Arabian Sea (10–20° N; 62–72° E), Bay of Bengal (9–19° N; 85–95° E), South Indian Ocean (10–20° S; 70–80° E), Eastern North Atlantic (10–20° N; 18–28° W), Western North Atlantic (31–41° N; 65–75° W), Eastern South Atlantic (3–13° S; 0–10° E), Central South Atlantic (2–12° S; 33–43° W), and finally the Western North Pacific (32–42° N; 140–150° E). The CERES-SSF product combines the radiative fluxes retrieved from the CERES instrument with aerosol properties from the MOD04 (Collection 4) product (Remer et al., 2005) and cloud (Minnis et al., 2003) properties retrieved from MODIS. At nadir, CERES-SSF footprint resolution is ~20 km with a near daily global coverage. Cloud properties include cloud liquid water path (LWP), water cloud effective droplet radii (R_c), cloud optical thickness (COT), and cloud top pressure (CTP) retrieved from the 3.7 μm (near-infrared) channel (Minnis et al., 2003). For adiabatically stratified water clouds, the theoretical relationship between R_c and LWP is described by Eq. (1) where ρ is the density of liquid water and τ_c is cloud layer optical depth (e.g. Wood and

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Hartmann, 2006).

$$\text{LWP} = \frac{5}{9} \rho \tau_c R_c \quad (1)$$

The MODIS is capable of resolving cloud characteristics at 2 different levels, one nearer to the surface, the other (if it exists) higher in the atmosphere. However, the primary focus of this study is liquid water clouds, so only data from the lower cloud layer are considered. In the regions considered, this layer is usually less than 5 km a.s.l., or below the atmospheric freezing level (0°C). The MODIS algorithm uses visible wavelengths to retrieve cloud optical depth and near IR to mid-IR measurements to retrieve cloud droplet size that are then converted to LWP using Eq. (1). The only constraints placed on the data (outside normal quality control flags) is that MODIS cloud data are only used for pixels over water surfaces and when the MODIS cloud phase parameter indicates that the cloud in question is at least 95% or more comprised of liquid water droplets. Potential effects of aerosols on ice clouds are beyond the scope of this study. Compared to the cloud retrieval in the MOD06 product (Platnick et al., 2003), CERES-SSF generally produces smaller cloud droplet size and cloud optical thickness (COT) values, though the overall patterns are generally similar with overall cloud amounts differing less than 10% (Minnis et al., 2003). Since we are focusing on relative the changes between cloud and aerosol characteristics, a bias one way or the other should not significantly affect the outcome of this research.

Han et al. (1994) and more recently Platnick et al. (2003) provide a review of the various error sources in the retrieval process including calibration, assumptions in atmospheric and surface properties, ambiguous solutions for optically thin clouds calibration, vertical inhomogeneity of clouds and cirrus contamination. The most significant uncertainty related to this research is the uncertainty associated with optically thin clouds (e.g. Nakajima and King, 1990). Under these circumstances, the relationship between retrieved COT and cloud droplet effective radius may break down. However, we cannot ignore optically thin clouds as part of this research as they contribute a large

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portion of the total cloud cover for some regions at certain times of the year (Jones and Christopher, 2008).

In addition to using cloud-top pressure to determine cloud top height, this research also examines cloud layer height using the MISR Stereo height product (Moroney et al., 2002; Kahn et al., 2007). This product identifies cloud (or thick aerosol) layers by stereo-matching images from MISR's nadir camera with those from camera observing at more oblique angles up to 70°. Using observations of the same feature from multiple viewing angles, it is possible to solve for the approximate height of that layer to within ±200 m. The result is a map of either cloud layer or aerosol layer heights for a given MISR swath, which can then be compared to the independent cloud property retrievals. Several examples of MISR data were collected where both significant aerosol and cloud concentrations exist to better examine the importance of vertical profiles of each to the indirect effect. In future work, co-located spaceborne lidar data (e.g. from the CALIPSO satellite) may be used in addition to the MISR data. However, no long time series currently exists, and CALIPSO is only available for the afternoon constellation including the Aqua satellite.

2.2 Aerosol properties

Cloud properties are combined with MODIS derived aerosol optical thickness (AOT) and fine mode fraction (FMF) at 0.55 μm wavelength. MODIS AOT is derived from clear-sky 500 m pixels and aggregated to 10 km footprint used by the MODIS level 2 aerosol product (MOD04). Where at least some number of clear-sky pixels exist within each 10 km footprint, a total and small-mode AOT value for that footprint is reported along with the fraction of that 10 km footprint covered by clouds. The cloud fraction ranges from 0 (indicating completely clear) to 1.0, indicating totally cloudy scenes. Aerosol retrievals are often possible for cloud fractions up to 0.95. MODIS resolution AOT is then converted to CERES resolution product using a point spread weighting function. CERES resolution AOT is reported unless nearly 100% cloud cover exists over an entire CERES footprint, which occurs <10% of the time. The accuracy of the

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MODIS AOT product over oceans is $\pm 0.03 \pm 0.05\tau$ with the uncertainty for the FMF being somewhat greater (Remer et al., 2005).

FMF is a measure of aerosol size with large values of FMF indicating mostly small-mode (e.g. largely anthropogenic) aerosols present, and low values indicating mostly coarse-mode (e.g. coarse sea salt and/or mineral dust) present (Kaufman et al., 2005b). FMF is used as a tool to determine the effect of aerosol type on cloud characteristics. Predominately small mode ($D < 0.25 \mu\text{m}$) aerosols are often considered anthropogenic in origin while larger, coarse mode ($D > 1 \mu\text{m}$) aerosols are considered naturally occurring (dust and sea salt). Previous research indicates that the Twomey effect should be greatest in the presence of small-mode aerosols in relatively moist environments (e.g. Heintzenberg et al., 1997; Quaas et al., 2004). While this may be the case in certain circumstances, the complexities of the aerosol-cloud droplet interactions mean that no one process can often be singled out. AOT, FMF, R_c , and LWP will be compared to determine the nature and changes of the indirect effects in each region as a function of time.

The TOMS instrument can also be used to study aerosol concentration and type using ultra-violet (UV) observations (Hsu et al., 2000). In particular, dust and biomass burning aerosols absorb radiation at UV wavelengths when compared to a clear-sky or even a maritime aerosol background (Torres et al., 2002). Highly absorbing carbonaceous and dust aerosols above the boundary layer are most sensitive to UV radiation. TOMS-AI is less sensitive to anthropogenic aerosols resulting from pollution. In general, AI is not sensitive to either aerosol types below the boundary layer top ($H < \sim 2 \text{ km}$). The relationship between UV radiation and absorbing aerosol concentration is defined in terms of the TOMS aerosol index (AI), which is the difference between the UV observations and model calculations from a pure molecular atmosphere with the same surface and measurement conditions. Positive AI values indicate the presence of UV-absorbing aerosols in the mid and upper troposphere, while near zero and negative values are indicative of non-absorbing, scattering, small-mode, and/or aerosols near the surface.

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2.3 Meteorology

Monthly mean, global surface wind speed and direction, and relative humidity at 1000, 850, and 700 hPa levels were obtained from National Center for Environmental Prediction (NCEP) Reanalysis data. The NCEP Reanalysis contains global meteorological conditions with a 2.5 degree horizontal resolution and a 17 level vertical resolution at 6 h time intervals (Kalnay et al., 1996). The reanalysis data set reliability captures synoptic scale dynamic and thermodynamic features, though often misses smaller scale phenomena. Total column humidity is included within the CERES-SSF product co-located with the CERES footprint as derived from ECMWF operational meteorological analysis. Daily NCEP data is substituted when analyzing specific examples of indirect effects.

3 Methodology

All statistics such as correlations and regression coefficients between aerosol and cloud properties are computed using the pixel-level data (with a daily resolution temporally and ~20 km spatially) within each $10 \times 10^\circ$ region for each month of data. Pixel-level data for each one month period are averaged to form monthly averaged values from which times series of aerosol, cloud and atmospheric conditions are constructed. Due to the relatively low resolution of the NCEP data, only monthly averaged atmospheric conditions are examined. Anthropogenic and dust direct and indirect radiative effects are calculated using a modified form of the methods outlined by Quaas et al. (2008). As part of this process, the total AOT (τ) must be separated into its maritime (sea-salt), dust, and small mode constituents. For the purposes of this research, small mode aerosols are considered to be primarily anthropogenic in origin and are labeled as such. The Kaufman et al. (2005) method is employed to calculate the portion of AOT resulting from each aerosol type (Jones and Christopher, 2007). This method assumes that maritime AOT is primarily a linear function of wind-speed and

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that maritime (τ_{ma}), dust (τ_{du}), and anthropogenic (τ_{an}) aerosols each have characteristic FMF values that can be used as separation points between each aerosol type. Characteristics FMF values used here are the same as those employed by Jones and Christopher (2007), which do not vary as a function of region, but are allowed to vary as a function of time. Uncertainties in this method are explained in detail in Jones and Christopher (2007 and 2008), where it is noted that the component AOT values have an uncertainty of between 30 and 50%. Quaas et al. (2008) used the method outlined by Bellouin et al. (2005) to calculate the anthropogenic portion of the AOT. However, this method does not discriminate between sea-salt and dust aerosols, preventing the calculation of dust direct and indirect effects. Uncertainties of up to 50% exist both direct and indirect methods (Quaas et al., 2008).

Dust and anthropogenic direct radiative effect are calculated using the incoming solar radiation derived from the satellite overpass time, solar zenith angle, and earth-sun distance, and then applying a diurnal adjustment factor (D) (Bellouin et al., 2005; Jones and Christopher, 2007). Unlike Quaas et al. (2008), we apply the diurnal adjustment method used by Remer and Kaufman (2005) and further by Jones and Christopher (2007). Differences between these adjustments are small. The diurnally averaged direct radiative effect due to anthropogenic aerosols (ΔF_a) can be expressed as the change in planetary albedo due to anthropogenic aerosols ($\Delta\alpha_a$) multiplied by the incoming solar radiation at the top of the atmosphere (F_s) as shown in Eq. (2).

$$\Delta F_a = \Delta\alpha_a F_s \quad (2)$$

The change in planetary albedo due to anthropogenic aerosols is expressed by Eq. (3).

$$\Delta\alpha_a = \frac{d\alpha}{d\ln(\tau)} [\ln(\tau) - \ln(\tau - \tau_{an})] \quad (3)$$

Combining Eqs. (2) and (3) and using the empirical relationship derived for $d\alpha/d\ln(\tau)$, the anthropogenic direct radiative effect becomes

$$\Delta F_a = -(1 - f)a_2 [\ln(\tau) - \ln(\tau - \tau_{an})] F_s D \quad (4)$$

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where a_2 is a constant defined by Quaas et al. (2008) and f is the total cloud fraction. These and the following constants are computed as a function of season over several ocean domains and the appropriate values for each region defined here are used. To calculate dust direct radiative effect (ΔF_d), the term $\ln(\tau - \tau_{an})$ is simply replaced by $\ln(\tau - \tau_{du})$ forming Eq. (5).

$$\Delta F_d = -(1 - f)a_2[\ln(\tau) - \ln(\tau - \tau_{du})]F_s D \quad (5)$$

This research does not compute direct radiative effects using CERES shortwave radiance observations like previous research that use CERES-SSF data (e.g. Jones and Christopher, 2007). The primary reason is that we want to compare direct and indirect effects of aerosols in the same region, and using compatible methods greatly simplifies this process. Also, the method used here already takes into account the effect of cloud-cover eliminating the need for any sort of bias adjustment (Christopher and Jones, 2008).

The cloud albedo effect (or first indirect effect) is a function of the relationship between the number density of liquid water droplets in a cloud (N_d) and the aerosol optical thickness. Number density is not reported directly within the CERES-SSF product and must be calculated using cloud optical thickness (τ_c) and effective droplet radius (r_e). Assuming adiabatic conditions, Brenguier et al. (2000) derive this relationship to be

$$N_d = \gamma \tau_c^{1/2} r_e^{-5/2} \quad (6)$$

where $\gamma = 1.37 \times 10^{-5} \text{ m}^{-0.5}$. The first anthropogenic indirect effect (ΔF_{ia1}) can be expressed by Eq. (7), where $A(f, \tau_c)$ is empirical function relating albedo to cloud fraction and cloud optical thickness. This function is explained in detail in the Appendix of Quaas et al. (2008).

$$\Delta F_{ia1} = -f A(f, \tau_c) \frac{1}{3} \frac{d \ln(N_d)}{d \ln(\tau)} [\ln(\tau) - \ln(\tau - \tau_{an})] F_s D \quad (7)$$

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When the correlation between N and τ is positive (e.g. more aerosols=more cloud droplets), the indirect effect becomes negative, cooling the atmosphere. If the correlation is negative, then ΔF_{ia1} becomes positive, opposite to the expected first indirect effect. To calculate the dust aerosol indirect effect (ΔF_{id1}), the same substitution that is made for the direct effect (Eq. 4) is made to Eq. (7). The term $d \ln(N_d)/d \ln(\tau)$ represents the linear regression fit between the natural logarithm of cloud droplet number density and aerosol optical thickness. This value is calculated on a month-by-month basis and is unique to each region studied. Uncertainties in this relationship are the greatest contributor to uncertainty in the reported in direct effects using this method.

Some clues about the second indirect effect or cloud lifetime effect were also derived by Quaas et al. (2008). However, given the large uncertainties present in the relationship between cloud fraction, cloud liquid water path, and going from number density to aerosol optical thickness, we chose to primarily focus our results on the first aerosol indirect effect. The term “aerosol indirect effect” in the following discussion referred to the first AIE component only unless otherwise stated.

Many uncertainties in both observations and cloud-aerosol interactions exist that complicate the interpretation of the resulting AIE values. While the MODIS algorithm uses strict cloud-clearing thresholds when calculating AOT, some cloud contamination does remain (Remer et al., 2005; Yuan et al., 2008). When this occurs, AOT is overestimated in the vicinity of clouds when partly cloudy conditions exist within a MODIS pixel (Koren et al., 2007). Similarly, Rayleigh scattering from nearby clouds may also lead to spuriously high AOT retrievals (Mauger and Norris, 2007; Marshak et al., 2008). Both these observational biases would result in an increased correlation between AOT and COT, leading to an overestimation of AIE (primarily the second AIE). However, sensitivity studies by Yuan et al. (2008) showed that these uncertainties did not play a major role when estimating AIE. Other uncertainties may be a result of aerosol hygroscopicity. Some aerosols species, such as sea salt and sulfate, are hygroscopic meaning that they will grow in size in high humidity environments (Feingold et al., 2003). High humidity environments are of course present in the vicinity of clouds. Thus, the same aerosol

concentrations will produce a higher AOT near clouds since the aerosols have swelled in size due to the moisture. While this may be indeed occurring, the uncertainty in aerosol retrievals in near clouds would make observations of this effect difficult. Since this effect is likely small, and may not be observed in a pure form anyway, it is unlikely to cause significant impacts when interpreting overall AIE. However, this may significantly affect whether or not this research classifies the AIE as dust or anthropogenic. Sulfate aerosols in pure form are assumed to be mostly small-mode and anthropogenic. However, under some circumstances, anthropogenic aerosols can be larger and when this occurs anthropogenic AIE will be falsely classified as dust AIE. Since this distinction has never been attempted previously from an observational perspective, the relative magnitude of anthropogenic vs. dust AIE must be considered highly uncertain. (More confidence does exist in the combined dust + anthropogenic values).

Both direct and indirect effects are also a function of solar zenith angle. The same albedo perturbation (whether from aerosols or clouds) produces a greater radiative effect during the summer months when solar zenith angle is lower. If all things are equal, then both DRE and AIE will increase somewhat during summer when solar radiation is more direct. Using data from Jones and Christopher (2008), DRE varies ~30 percent between solar zenith angles of 20° and 60°. It will be shown that things are decidedly not equal and that much larger variations do exist that cannot be taken into account by changes in solar zenith angle of $\pm 15^\circ$. Other, less significant, uncertainties in near-cloud aerosol observations are also present and are discussed in further detail by Yuan et al. (2007). As noted previously, uncertainties in cloud property retrievals are greatest for optically thin clouds ($COT < 4$ or $LWP < 20 \text{ gm}^{-2}$); thus, confidence in AIE increases as cloud thickness increases.

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4 Results

4.1 Regional direct and indirect effects

A suite of general circulation models estimate that anthropogenic direct aerosol radiative effects range from -1.9 to -0.3 Wm^{-2} globally (Lohmann and Feichter, 2005; Forster et al., 2007). More recent observational studies indicate that the total anthropogenic aerosol effect is likely on the small side of the range previously reported, and possibly negligible in certain regions (Matsui et al., 2006; Quaas et al., 2008). No corresponding statistics for the indirect effects of dust aerosols are known to the authors. While this research does not report globally averaged values, but instead focuses on regional differences in both the dust and anthropogenic indirect effects to determine under what conditions these effects are most likely to occur (Fig. 1). Table 1 lists dust and anthropogenic direct and indirect effects derived using the methods described in Sect. 3. All radiative effect values reported here are diurnally averaged with no clear-sky bias adjustment required (Christopher and Jones, 2007). Monthly averaged cloud top pressure ranges between 824 hPa in the Bay of Bengal to as low as 853 hPa in the southern Atlantic. These values correspond to heights between 1 and 2 km above sea-level (a.s.l.), which is well below the freezing level in any of these regions. If the aerosols and clouds are located at different atmospheric levels, then false indirect effects may be observed. Examples of both conditions are examined for some regions using MISR stereo height data.

The largest aerosol direct effects are associated with the highest aerosol concentrations (Table 1). The 6-year average of total column AOT is greater than 0.25 for regions AS, BB, EA, AF, and WP. The combined dust and anthropogenic direct effects range between -3.0 and -4.0 Wm^{-2} . Corresponding indirect effects do not necessarily correspond to higher AOT. For BB and EA, both the dust and anthropogenic indirect effects are negligible, indicating that other factors besides aerosols are significantly contributing to droplet growth (however, adiabatic N is used rather than R_c for the computation of AIE). In EA, the primary aerosol species is dust, which has usually not been thought

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of as good CCN (Levin et al., 1996). Depending upon their properties, dust aerosols may also absorb solar radiation warming the layer where dust is located, increasing atmospheric stability. This increase in stability inhibits cloud formation, which in turn prevents the occurrence of the aerosol indirect effect (Matsui et al., 2006; Mauger and Norris, 2007). Thus, the weak aerosol indirect effects in EA are not completely unexpected. BB is more of a mystery since sulfates account for a large proportion of the total AOT, with anthropogenic AOT, DCF, and cloud property retrievals all being similar to those observed in EA. One key difference is that the concentration of elevated dust aerosols is much less in BB compared to EA. Another is that upward vertical motion is greater between 900 and 700 hPa, where most of the liquid water cloud in both regions exist. The importance of these differences relative to indirect effects is explained in greater detail below.

In AF, average AOT is 0.32, but the dust+anthropogenic direct radiative effect is only -3.1 Wm^{-2} , somewhat less than expected for this aerosol concentration. However, this region contains the largest proportion of black carbon aerosols, which also absorb solar radiation and warm the atmosphere, which in turn reduces shortwave radiative efficiency while increasing atmospheric stability (Matsui et al., 2006). The anthropogenic indirect effect is -0.37 Wm^{-2} with the dust effect being -0.03 Wm^{-2} . Similar results were observed in CO for anthropogenic direct and indirect effects, though dust aerosol concentrations and effects are larger. This is consistent with observations by Ramana and Ramanathan (2006), which show a complex mix of aerosol species in this region.

Indirect effects are also observed to be over the open ocean regions, IO and SA, which have total (anthropogenic+dust) indirect effects of -0.8 and -0.2 Wm^{-2} respectively and are comparable to the corresponding direct radiative effect values (Table 1). However, the total AOT in both regions is low ($\tau=0.11$) indicating that the aerosols that are present are very effective CCN. Approximately 50% of the total AOT is comprised of maritime sea salt aerosols that are not accounted for in the direct and indirect calculations presented here. Indirect effects from dust aerosols are present for both regions, despite the lack of a nearby dust source. It is possible that some maritime sea salt

aerosols are being misclassified as dust using the Kaufman et al. (2005) algorithm. Aerosol concentrations, direct and indirect effects all vary significantly as a function of the atmospheric conditions present (Jones and Christopher, 2008; Yuan et al., 2008). The temporal variation of these conditions and how it relates to aerosol indirect effects is examined in the following section. While the values of the indirect effects in each region have large uncertainties, these are still not enough to explain the seasonal variability in these effects observed in several regions. Below, we discuss this variation for each region as a function of aerosol concentration and atmospheric conditions and their relative importance to AIE.

4.2 Seasonal variation in AIE

(i) Arabian Sea (AS)

Aerosol indirect effects vary substantially as a function of time in all regions. These variations are a function of changes in AOT, atmospheric moisture content, and synoptic uplift. In the Arabian Sea, the maxima in cloud fraction and COT during JJA correspond well with maxima in 850, 700 hPa humidity and 850 hPa vertical velocity (Figs. 2–5a). As such, the atmospheric environment is most favorable for deep cloud formation during this period. AOT is also maximized at the same time, primarily from an influx of elevated dust aerosols and increased production of sea-salt due to higher wind speeds (Jones and Christopher, 2008). Since both AOT is high and conditions for cloud formation are favorable, indirect effects would seem more likely to occur. While dust aerosols themselves make poor CCN, they may be coated with sulfate aerosol already present in the Arabian Sea (Levin et al., 1996). Also, the sea salt aerosols being produced can act as excellent CCN. The available observations do not resolve which of these aerosol species is most important to the indirect effects in the Arabian Sea, but the authors believe that both likely have some impact.

Correlation between R_c and AOT was indeed most negative in JJA with a correlation coefficient of -0.3 averaged over JJA for all years. Similar results were observed by

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Patra et al. (2007), also indicating maximum indirect effects during the summer months. This and all following correlations $>\pm 0.1$ are statistically significant to a 99% or greater level using paired Student's T test. The correlation between R_c and AOT is weakly positive during the winter resulting in both anthropogenic and dust AIE being small, despite the monthly averaged R_c also being small compared to the overall mean value (11.0 vs. 14.2 μm). The effective radius is also greatest during the summer months, when aerosol concentrations are greatest. This differs from the finding by Chylek et al. (2006) who observed smaller droplet radii in September (high AOT) compared to larger radii in January (low AOT). However, their spatial domain was substantially larger than used here; thus, their statistics are not necessary valid for the smaller Arabian Sea domain used by this research. The reason for the generally larger water droplets in the summer has little to do with the aerosol themselves, but more a response to more favorable atmospheric conditions for cloud development as evidenced by greater moisture concentrations and upward motion during the monsoon season (Fig. 3a).

While total AOT being greatest during the summer months, anthropogenic direct radiative effect is maximized during November and December when the anthropogenic portion of the AOT is greatest with values exceeding -5.0 Wm^{-2} (Fig. 5a). However, the anthropogenic indirect effect at this time is negligible. Here, atmospheric humidity is less and vertical motion is weak, inhibiting cloud formation leaving relatively thin clouds as evidenced by the decrease in COT (Figs. 4a, 6a). Comparison of thin ($\text{LWP} < 20 \text{ gm}^{-2}$) vs. thick ($\text{LWP} > 20 \text{ gm}^{-2}$) clouds shows that the correlation between AOT and R_c is more negative for the thick cloud sample. As a result, AIE is greater for thicker clouds, consistent with Nakajima and King (1990) and the vertical motion results observed here. This is true for this and all other regions analyzed by this research.

As the anthropogenic component of AOT decreases and dust increases, the dust direct radiative effect extends up to -2.5 Wm^{-2} . Both anthropogenic and dust indirect effects are maximized in the summer, with values near -1.0 Wm^{-2} . The question remains as to whether or not the dust indirect effect reported here is actually from pure dust aerosols. It is possible that that some maritime sea salt aerosols are being

classified as dust by the algorithm. Dust coated with sulfates may also be part of the dust component. Between June and August (JJA), sustained upward vertical motion from near the surface up to at least 600 hPa may be aiding both by transporting some sea salt and anthropogenic aerosol high enough into the atmosphere to interact with some of the dust aerosols present (Fig. 6b). Anthropogenic aerosols in the vicinity of clouds may be increased in size due to high humidity (Feingold et al., 2003; Loeb and Manalo-Smith, 2005), which could also result in a misclassification of these aerosols as dust. Finally, it is important to determine whether or not the elevated dust aerosols are co-located with cloud heights. To examine this last question, cloud height data from the MISR Stereo height product were analyzed for 8 August 2003 corresponding to the time period when the indirect effect in the Arabian Sea is maximized. MISR estimated cloud heights in the Arabian Sea generally range between 1 and 3 km a.s.l., while significant aerosol concentrations are also evident from over-laid MODIS AOT data (Fig. 7). While aerosol heights are not directly sampled, the average TOMS-AI values for this day (1.8) indicates the presence of elevated dust aerosols well above 1 km a.s.l. Daily NCEP Reanalysis of vertical velocity (ω) at 15° N and 67.5° W shows that upward motion is maximized between 850 and 700 hPa, with ω remaining negative up to 400 hPa (Fig. 8). Since both aerosols and clouds exist in the same atmospheric column where upward motion favorable for cloud formation is present, indirect effects are quite likely to occur. Evidence for this was observed in correlation between AOT and R_c for this day, which was -0.35 .

(ii) Bay of Bengal (BB)

In the Bay of Bengal, a similar pattern exists as in the Arabian Sea. AOT is maximized in the summer due to increase dust aerosol transport and some increase in sea-salt production. The magnitude of the increase in dust aerosols is much smaller than that observed in the Arabian Sea ($\tau_{du}=0.05$ vs. 0.11), and small mode aerosols account for larger proportion of the total AOT. Comparisons of model and observed aerosol speciation by Jones and Christopher (2007) suggest that in this case, the small mode aerosols

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consist primarily of sulfate. The mean anthropogenic AOT component is nearly identical for both regions ($\tau_{an}=0.16$). As in the Arabian Sea, anthropogenic direct radiative effect is maximized during the winter with values between -6 and -8 Wm^{-2} (Fig. 5b). The corresponding increase in summertime dust aerosol direct effect is much smaller (-1.0 Wm^{-2}). Cloud fraction, cloud droplet effective radius, AOT, relative humidity, and upward vertical velocity are all again maximized in the summer. Unlike the Arabian Sea, no corresponding increase in the either anthropogenic or dust aerosol indirect effects are readily apparent (Figs. 2b,5b). The most significant difference in Arabian Sea vs. Bay of Bengal conditions is the relative heights of the aerosol and cloud layers between both regions. On average, cloud top pressures are approximately 30 hPa higher than in the Arabian Sea, but the aerosols themselves are more concentrated nearer the surface.

In the Bay of Bengal at least two cloud layers are present in an example from 23 July 2003 (Fig. 9). The most predominate cloud type appears to be cirrus clouds resulting from small pockets of convective activity with heights between 5 and 10 km a.s.l. Below this layer exist a few maritime stratus and cumulus clouds, but their overall areal coverage is low. NCEP data at 15° N and 90° W shows a bimodal distribution of vertical velocity with two levels at which upward motion is maximized, 850 and 300 hPa (Fig. 8). Relative humidity is also high (86%) at this level indicating the presence of this second cloud layer. Mean TOMS-AI is much lower (0.4) than in the Arabian Sea, which is a result of decreased elevated dust aerosol concentrations compared to the Arabian Sea. Total column AOT is also lower primarily due to the decrease in the dust component (Table. 2). As previously noted, aerosols in the Bay of Bengal are generally anthropogenic in nature with the greatest concentration likely present below the maritime boundary later ($\sim 1 \text{ km}$). Thus, most of the clouds in this example appear to be located above the primary aerosol layer, limiting the probably of the indirect effect occurring. During the summer months, upward vertical motion in the Bay of Bengal between 850 and 700 hPa is only half that observed in the Arabian Sea. Conversely, upward motion above 400 hPa is slightly greater. This is consistent with the example

above, and provides further evidence that cloud formation at least in this case is less favored in the Bay of Bengal. The weak upward motion coupled with significant aerosol concentrations not present where upward motion is maximized is a key reason indirect effects are not as apparent as they are in the Arabian Sea.

5 (iii) Eastern North Atlantic (EA)

In the eastern North Atlantic, aerosols primarily originate from dust over the Saharan Desert and are advected westward by the prevailing winds (Dunion and Veldon, 2004; Kaufman et al., 2005a). Dust aerosol concentration is maximized in the late spring and summer months, leading to a seasonal cyclone in AOT and dust direct radiative effect (Figs. 2–5c). Cloud fraction remains approximately constant at 60% with the exception of decreases occurring during the months of December and January (Fig. 3c). While cloud fraction does not change much otherwise, COT does increase during the summer months in association with the northward movement of the ITCZ. During this time, atmospheric moisture is maximized and overall vertical velocities are near zero. This period also corresponds to active tropical cyclone development associated with low pressure systems moving westward off the African coast. By contrast, very low humidity (<30%) exists during the winter months with strong subsidence also present. There does exist a seasonal cycle in the indirect effect, with a positive correlation present during the winter months, where AOT and moisture content are lower. In the summer the correlation is often near zero. This cycle is consistent with that observed in the Arabian Sea, with the exception that the AOT – R_c correlation never quite becomes consistently negative. The overall dust and anthropogenic aerosol indirect effects are small (0.03 and 0.05 Wm^{-2}), but with slightly negative values (< -1.0Wm^{-2}) reported during the summer months of each year with the exception of 2004. Another commonality with the Arabian Sea region is a statistically significant correlation between monthly averaged 850 hPa velocity and AIE, with greater AIE occurring with upward motion is maximized. It appears that the aerosol indirect is indeed occurring, but that it is only able to offset the conditions that lead to cloud droplet size increasing with increasing

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AOT.

One possible reason for the indirect effect in summer not being larger is the mid-level sinking and dry air associated with large dust outbreaks (Dunion and Veldon, 2004). Depending upon their properties, dust aerosols absorb solar radiation, thereby warming mid-levels of atmosphere leading to stabilization, suppressing convection and cloud formation (Ackerman et al., 2000; Koren et al., 2008; Matsui et al., 2006). It follows then that if cloud coverage is low for a given region, then the stabilization effect of absorbing aerosols is more likely to occur offsetting the cooling effects of indirect effects associated with the clouds that are present (Koren et al., 2008). For this region, cloud fraction shows no significant increase in the summer when AOT is maximized; thus, the offsetting stabilization effect is more dominant. So while AOT increases due to the increase in dust aerosol concentration, the atmospheric conditions collocated with the maximum AOT may not favor deep-layer cloud development. Also, recall that aerosols and clouds must exist at the same atmospheric level to interact. Dust aerosols, unlike most pollution-based aerosols, may be present several kilometers high in the atmosphere, well above most maritime stratus. Recall that the Arabian Sea also contains significant concentrations of dust aerosols, but dust AOT in that region is only one half that in EA ($\tau_{du}=0.11$ vs. 0.2). As a result, the likelihood of dust decreasing the favorability of the environment towards cloud formation is less. This coupled with the large-scale synoptic forcing also present in the Arabian Sea is why dust aerosols are more likely to be CCN there than compared to the North Atlantic.

MODIS/MISR observations on 23 July 2004 indicate large dust aerosol concentrations west of the African coast (Fig. 10). Convective clouds extending up to 10 km a.s.l. according to MISR stereo height retrievals are present south of an 850 hPa low pressure center (-25° W, 14° N). However, the average cloud top pressure for liquid water clouds during this month is 842 hPa. Dust aerosols that may be above the freezing level (~ 550 hPa) may act also as ice condensation nuclei (DeMott et al., 2003). This effect, however, is outside the scope of this research. Still, significant dust aerosol concentrations are likely present in the 1–3 km layer, co-located with the majority of the cloud

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cover, but little evidence for the indirect effect was observed for this day. In fact, the correlation between R_c and AOT was positive (0.23). As with the overall monthly mean, vertical motion is weak during this day, with even some subsidence being observed as low as 700 hPa. This acts to suppress cloud formation, and as a result would limit indirect effects. This is in contrast to the Arabian Sea, which also has large elevated dust concentrations in the summer months, but also has consistent upward motion to aid in the formation and growth of clouds.

(iv) Eastern South Atlantic (AF)

In the eastern South Atlantic, aerosols primarily originate from biomass burning consisting of large amounts of carbon based aerosols, especially between August and October (e.g. Lindsay et al., 1996). These aerosols are transported westward from central and southern Africa, leading to high AOT values (>0.6) in the region of study. This corresponds to an anthropogenic direct radiative effect of -8.0 Wm^{-2} , which is very consistent from year to year (Fig. 5d). A secondary maximum in dust aerosol concentration and direct radiative effect is present during February and March, with maximum values of -2.0 Wm^{-2} . With the exception of 2001, 850 hPa atmospheric humidity and vertical velocity (consistent subsidence at all levels) do not change substantially as a function of time; however, 700 hPa humidity is somewhat greater between August and October (Fig. 3d). This change corresponds with an increase in COT during the same period, though CTP actually increases indicating the presence of lower level, but thicker clouds (Fig. 4d). Anthropogenic indirect aerosol effects are only present during these months for all years except 2001. Relative humidity at 850 hPa is abnormally low this region for the latter half of 2001, reducing the likelihood of cloud formation. As a result, indirect effects are less likely to occur, which is consistent with our observations. The dust+anthropogenic AIE is only -0.4 Wm^{-2} , but this value is almost entirely due to the negative anthropogenic values observed from the August–October biomass burning, which are on the order of -3.0 Wm^{-2} (Fig. 5d). Otherwise, both dust and anthropogenic indirect effects are negligible. AF differs from other regions in that large

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indirect effects are not associated with at least some upward vertical motion. In fact, the only favorable atmospheric condition for an increase in COT after August is the increase in 700 hPa humidity. However, since monthly mean CTP is near 875 hPa during this period, the increase at 700 hPa should not be a significant factor. The question remains as to why indirect effects are observed despite the lack of favorable atmospheric conditions.

On 30 September 2004 MISR observed cloud-heights only up to 2 km, while biomass burning aerosols are likely present even higher in the atmosphere as the TOMS-AI value for this day is 1.5 (Fig. 11, Table 2). For this day, the anthropogenic AIE was -3.3 Wm^{-2} though consistent subsidence is also present. For indirect effects to be occurring then, significant aerosol concentrations should also be present near the surface. Unfortunately, CALIPSO data (or similar lidar information) for this example does not exist to determine if this is indeed the case. Experiments during the SAFARI campaign noted significant aerosol concentrations from biomass burning were trapped in a stable layer between 3 and 5 km a.s.l., allowing for transport well into the central Atlantic while aerosols below the boundary layer tended not to travel as far (Garstang et al., 1996). For this region, it is likely that it is these lower-level aerosols causing indirect effects, not those higher in the atmosphere. Still, further research in this region is required to fully understand the results observed here.

(v) Western Atlantic and Pacific (WA, WP)

Two regions consisting of aerosols produced from anthropogenic pollution sources include the eastern coasts of China/Japan and the United States respectively. Both regions exhibit similar aerosol characteristics with sulfate-based aerosols are the predominant aerosol species (Brenguier et al., 2003). In the western Atlantic, AOT and direct anthropogenic radiative effects are maximized in JJA in association with increased industrial pollution. Cloud cover and cloud thickness on the other hand are maximized during the winter months, in response to low-pressure systems that propagate through this region. However, overall upward vertical motion in JJA is relatively weak and shows

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no low level maximum like the Arabian Sea (Fig. 6b). Since cloud formation and growth are not favored during the period of maximum AOT, the aerosol indirect effect during this time is expected to be weak, but still is non-zero. This is indeed the case, with the correlation between R_c and AOT being near zero during summer months (Fig. 2f).

Anthropogenic AIEs for this period were estimated to be -1.0 Wm^{-2} while dust effects were negligible (Fig. 5f). In addition, some evidence for the first indirect effect does exist with a weak negative correlation during the winter months when synoptic forcing is maximized. This corresponds to a slight increase in the dust aerosol indirect effect, though its significance remains unclear.

East of China, AOT is maximized in the spring months (MAM) with sulfate aerosols accounting for the greatest portion, but with significant contributions from black carbon and dust aerosols as well. The corresponding anthropogenic aerosol direct radiative effect sometimes exceeds -8 Wm^{-2} with a dust effect up to -2.0 Wm^{-2} being observed (Fig. 5g). MODIS cloud fraction is maximized between October and February, with the maximum in COT occurring a month or two later. However, 700 hPa moisture is maximized during the summer months. Aerosol indirect effects were observed during the time of maximized 700 hPa humidity and AOT, but not overall cloud coverage (Figs. 3b, 5b). Upward vertical motion is also maximized at this time, but remains weak, even compared to WA (Fig. 6b). Given this, it would appear that aerosol indirect effects are most likely to occur in deeper, more optically thick clouds. Interestingly, indirect effects were observed during a period when R_c and AOT (and LWP–AOT) are positively correlated (Fig. 2g). Yuan et al. (2008) also found a positive correlation during the summer 2002 period. It possible that the positive correlation between LWP and AOT is offsetting the decrease in R_c due to an overall increase in N .

(vi) Oceanic regions

In more pristine environments such as in the Southern Indian Ocean and the far South Atlantic Ocean, little variability exists in the overall atmospheric conditions (Fig. 3g,h). Relative humidity is maximized between 850 hPa and the surface and vertical veloci-

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ties remain low (Fig. 6). Overall, cloud fraction and COT also do not vary by more than $\pm 10\%$ and ± 0.7 respectively, indicating that the primary cloud mode is maritime stratus. The likelihood for deep cloud formation is low. Both regions do show seasonal variations in AOT, which correspond to small changes in the dust and anthropogenic aerosol concentrations. The contributions from dust and anthropogenic aerosols account for $\sim 40\%$ of the total AOT, but recall that AOT in these ocean regions are the lowest for the eight regions studied here (~ 0.13). Dust and anthropogenic direct aerosol radiative effects vary according to the changes in their concentration, but only reach a maximum of -1.5 Wm^{-2} (Fig. 5g). Variations in aerosol indirect effects are less clear. In the South Indian Ocean, both anthropogenic and dust effects are consistently negative, but almost always less than -1.0 Wm^{-2} . In the South Atlantic, anthropogenic and dust aerosol indirect effects are positive in DJF and small for other seasons. Thus, direct and indirect effects are offsetting each other, though the magnitude of the direct effect remains larger (Fig. 5h). Note that the direct and indirect effects of maritime aerosols are not shown. For these regions maritime AOT accounts for more than half of the total AOT, and as a result has direct and indirect effects on the order of the dust+anthropogenic values listed here. Since, sea salt aerosols make excellent CCN, it is likely that most of the indirect effects that do occurring in this regions are due to these aerosols, not the low concentrations of anthropogenic and dust aerosols present.

5 Conclusions

It is clear from this long term analysis that atmospheric conditions have comparable importance to aerosol concentrations when determining the presence of aerosol indirect effects. We have shown that aerosol indirect effects are unlikely to occur in regions of subsidence and/or low atmospheric humidity even if large concentrations of aerosols are present. While clouds may exist in these regions, they generally exist in thin layers, where indirect effects are less well understood and likely relatively small. In some regions such as the Bay of Bengal, significant cloud cover is present at times, but the

cloud layers do not correspond with the levels of maximum aerosol concentration, thus limiting possible indirect effects. On the other hand, direct aerosol effects are clearly evident in all regions and correspond well to total aerosol concentrations. Some differences in the effectiveness of the direct effects are noted and are likely a result of differing aerosol types being present. The direct aerosol effect can also cool SSTs, which in turn would affect atmospheric stability and the probability of cloud formation. Clearly, this is a complex relationship that cannot be completely explained by any single research effort.

This research represents one of the first to discriminate dust from anthropogenic aerosol indirect effects. Where dust aerosol concentrations are large (AS, EA), a significant positive correlation exists between vertical velocity and AIE. It can be surmised that when dust aerosols are uplifted by buoyant parcels in a sufficiently humid environment, that act as CCN to the resulting clouds. Wang et al. (2004) noted that convective clouds (those associated with greater updrafts) are more likely to be sensitive to aerosols in the atmosphere, with our results providing additional evidence for this conclusion. Updrafts may also allow mixing of sulfates and sea salt primarily located in the boundary layer upward to into the free atmosphere mixing with the dust, further increasing CCN (and helping the dust become good CCN). In AS and EA, the dust effect was greater than the anthropogenic effect. This observation leads credence to the hypothesis that a portion of AIE is being caused by something other than fine mode anthropogenic aerosols, which have historically been the focus of study. While uncertainties in the accuracy of the classification are high, the differences in anthropogenic vs. dust AIE are significantly large in some cases ($>1.0 \text{ Wm}^{-2}$) to outside the range of what could be considered noise. Mixing of dust aerosols with sea salt (also coarse mode) and sulfates (which can coat dust) with the aid of sustained uplift is postulated as one possible explanation for the observations documented here, but many more likely exist. In other regions, such as AF, WA, and WP dust aerosol concentrations are low allowing anthropogenic indirect effects to be dominant. In these regions, the correlation between atmospheric conditions and indirect effect is less evident, but still

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observable.

Observations from this research indicate that for aerosol indirect effects to occur, the right combination of uplift, moisture concentration, and aerosols must be present at the same atmospheric level. For most regions, all of these criteria are rarely met at any one time. When they are, such as in the Arabian Sea, the effect is very seasonally dependent. When considering aerosol indirect effects in the future, it is vital the surrounding atmospheric conditions be taken into account. It is clear from this research in particular that it is much more complex than a simple $AOT - R_c$ correlation coefficient. The overarching conclusion of this research is that aerosol indirect effects are not simply a function of aerosol concentration and speciation, but are also highly dependent on the surrounding atmospheric conditions. While this is not necessarily a new observation, the long term data sets used here provide much improved documentation of this inter-relationship than previously available and represents a starting point for further research.

Acknowledgements. This research is supported by NASA's Radiation sciences, Interdisciplinary sciences, an EOS grant, and ACPMAP programs. The CERES SSF data that contains the merged MODIS and CERES and the MISR data was obtained through the NASA Langley Distributed Active Archive Systems.

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Table 1. Six year mean aerosol and cloud properties, and aerosol radiative effects for each region of study.

Region	Code	AOT	τ_{an}	τ_{du}	DRE _{an} [Wm ⁻²]	DRE _{du} [Wm ⁻²]	COT	CF [%]	Rc [μ m]	CTP [hPa]	IE _{an} [Wm ⁻²]	IE _{du}
Arabian Sea	AS	0.33	0.16	0.11	-3.00	-1.04	1.90	57.84	14.23	852.20	-0.1	-0.23
Bay of Bengal	BB	0.27	0.16	0.05	-3.20	-0.39	1.98	63.85	15.05	824.30	0.03	-0.01
S. Indian Ocean	IO	0.13	0.04	0.02	-0.94	-0.28	2.97	68.84	16.13	838.00	-0.51	-0.25
East North Atlantic	EA	0.39	0.13	0.20	-1.36	-1.55	2.08	58.32	14.16	843.80	0.05	0.03
West North Atlantic	WA	0.20	0.11	0.03	-3.25	-0.29	3.88	63.73	12.54	830.43	-0.56	-0.11
African Biomass	AF	0.32	0.22	0.05	-2.77	-0.37	3.27	66.04	11.77	853.30	-0.37	-0.03
South Atlantic	SA	0.13	0.04	0.03	-0.57	-0.22	3.70	68.68	14.54	829.60	-0.14	-0.09
Western Pacific	WP	0.27	0.15	0.05	-3.40	-0.45	4.33	65.51	12.58	831.30	-0.71	-0.16

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Table 2. MODIS AOT and CTP, TOMS-AI, NCEP vertical velocity at 850 hPa (Pa s^{-1}), and derived dust and anthropogenic AIE (Wm^{-2}) for the days corresponding to the examples shown in Figs. 7, 9–11.

Region	Date	AOT	AI	CTP [hPa]	ω_{850} [Pa s^{-1}]	IE_{du} [Wm^{-2}]	IE_{an} [Wm^{-2}]
AS	8 Aug. 2003	0.58	1.8	800	-0.18	-2.1	-0.1
BB	23 Jul. 2003	0.22	0.4	721	-0.13	0.9	0.4
EA	23 Jul. 2004	0.59	1.8	816	-0.06	-0.4	-0.4
AF	30 Sept. 2004	0.61	1.5	860	0	0	-3.3

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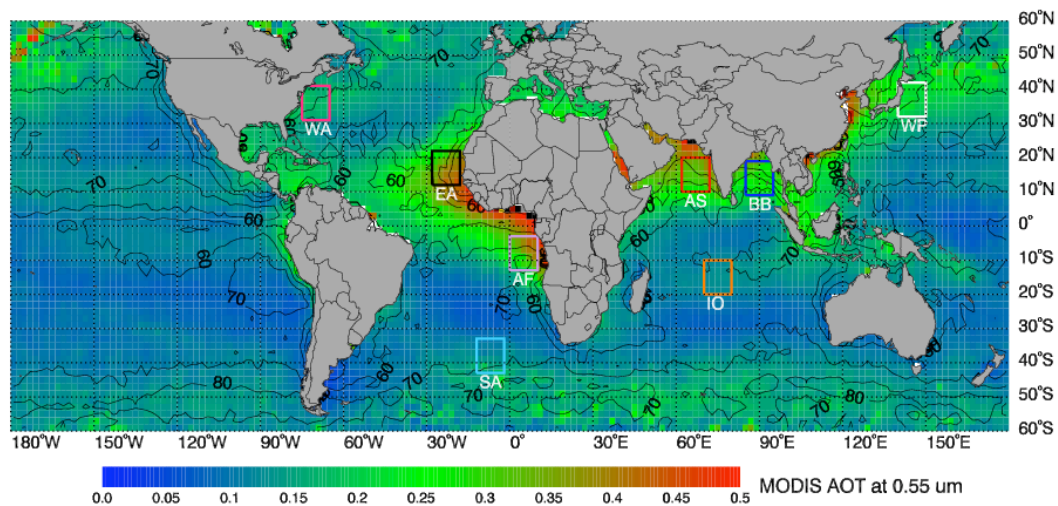


Fig. 1. Locations of study regions overlaid on globally averaged AOT, with contours indicating average cloud fraction (%).

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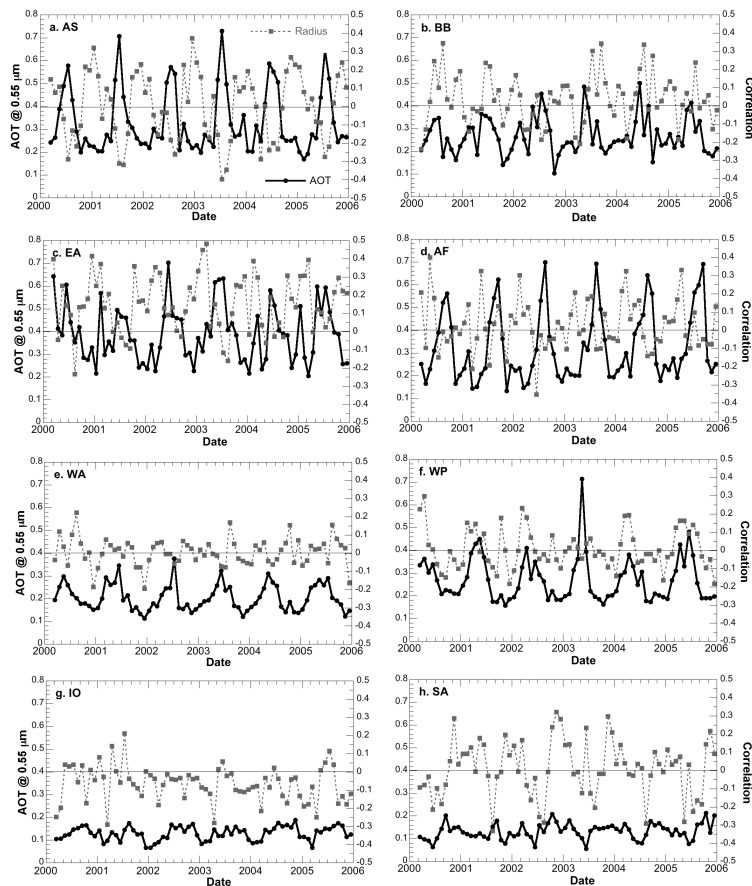


Fig. 2. Monthly mean AOT and correlation between AOT and R_c for each region between 2000 and 2006. Correlations in excess of ± 0.1 are considered statistically significant.

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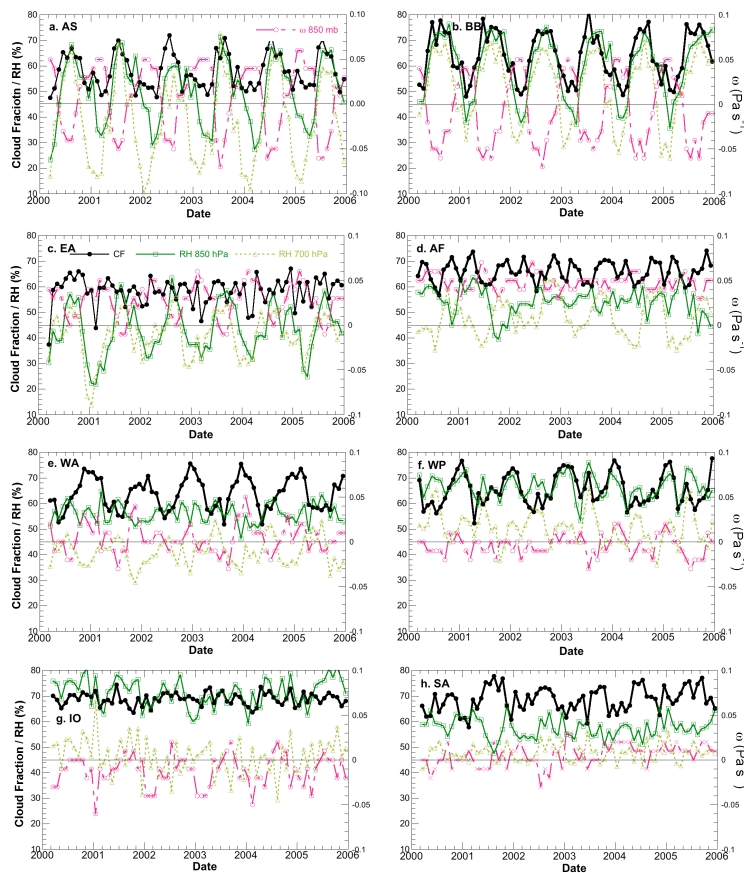


Fig. 3. Same as Fig. 2, but for NCEP 850 and 700 hPa relative humidity (%), 850 hPa vertical velocity (Pa s^{-1}), and MODIS cloud fraction. Upward motion is indicated by negative values.

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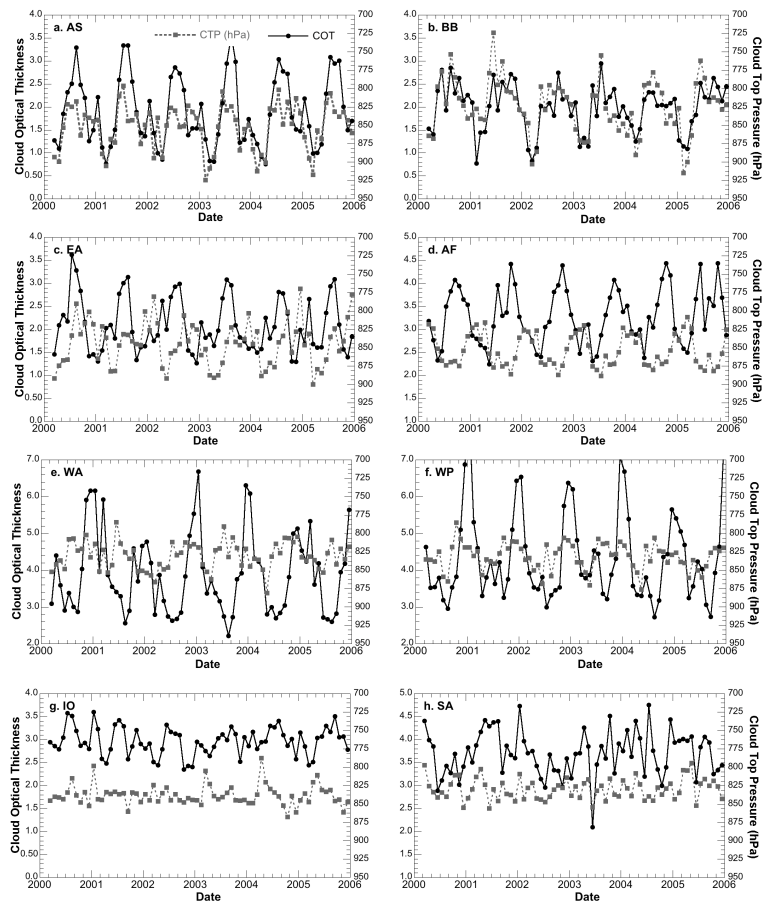


Fig. 4. Same as Fig. 2, but for cloud optical thickness (COT) and cloud top pressure (CTP, hPa).

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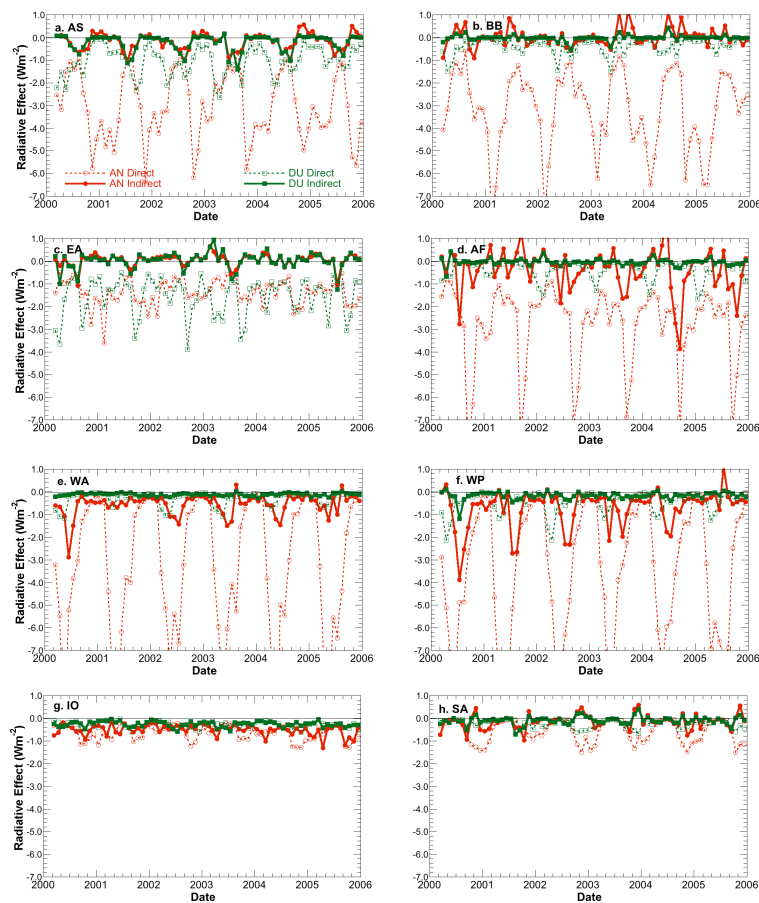


Fig. 5. Same as Fig. 2, but for direct and indirect dust (DU) and anthropogenic (AN) aerosol effects.

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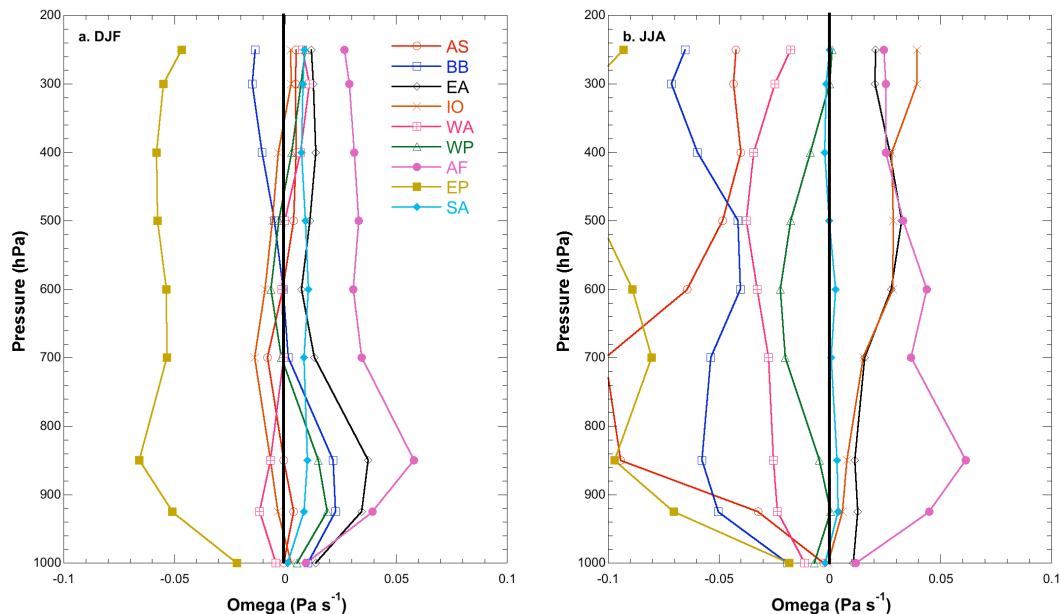


Fig. 6. Seasonally averaged (**(a)** DJF, **(b)** JJA) vertical velocity acquired from NCEP reanalysis data between 1000 and 250 hPa at 12:00 UTC for each region. Upward motion is indicated by negative values.

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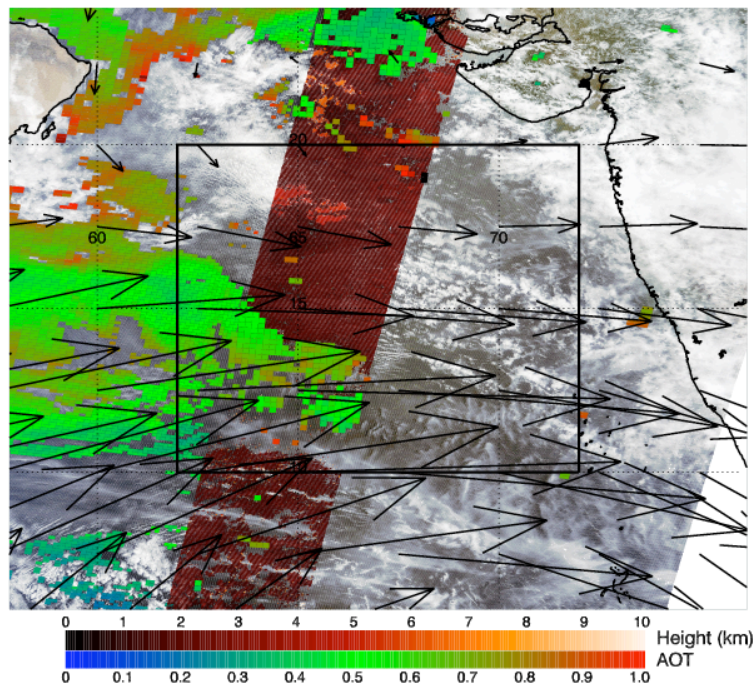


Fig. 7. Terra MODIS three band overlay for 8 August 2003 with AOT at $0.55 \mu\text{m}$ from both MODIS and MISR overlaid. The MISR stereo height product represents the height above sea level the upper most cloud layer is located. Vectors indicate 850 hPa wind speed and direction.

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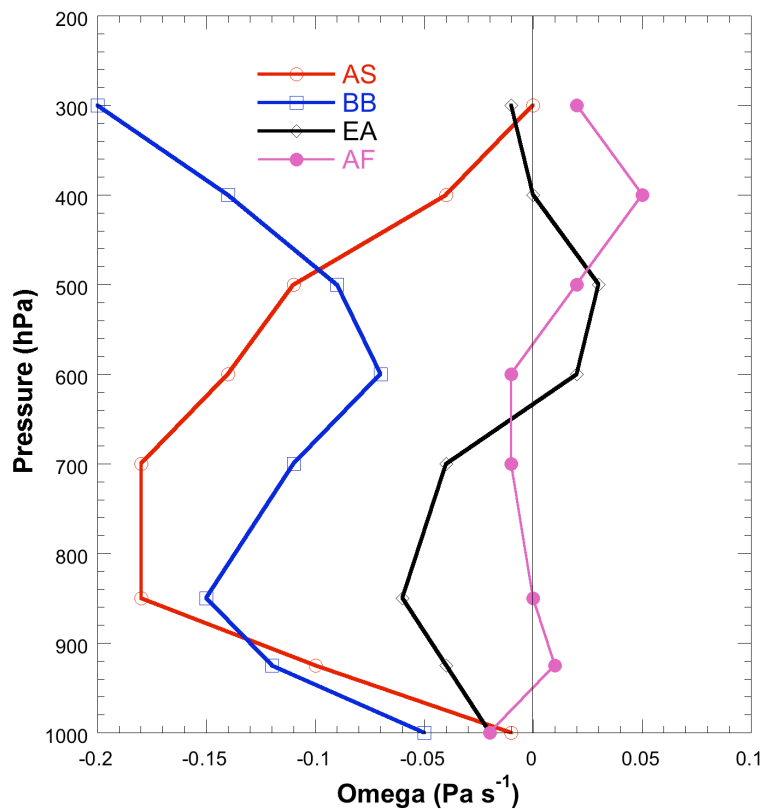


Fig. 8. Same as Fig. 6, but vertical velocities corresponding to date and time of the MODIS-MISR examples for regions AS, BB, EA, and AF respectively.

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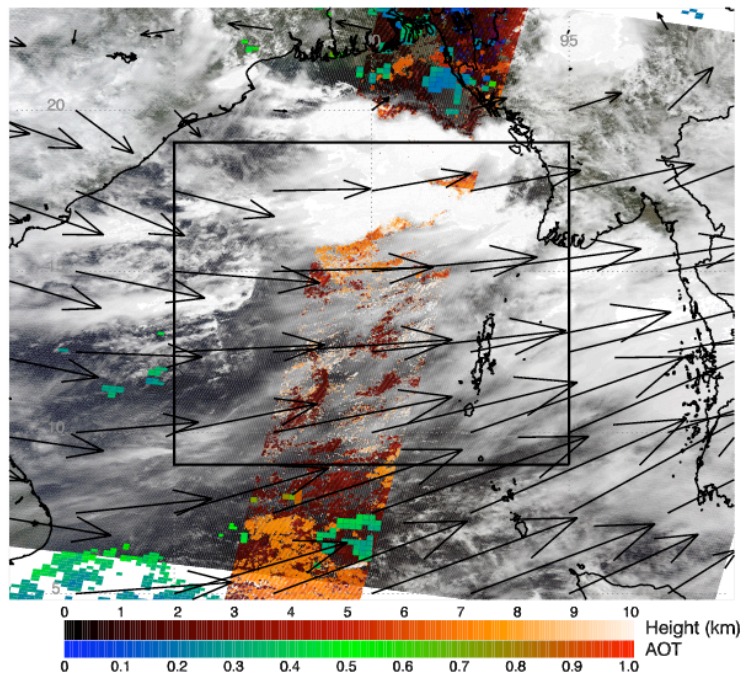


Fig. 9. Same as Fig. 7, but for the Bay of Bengal on 23 July 2003.

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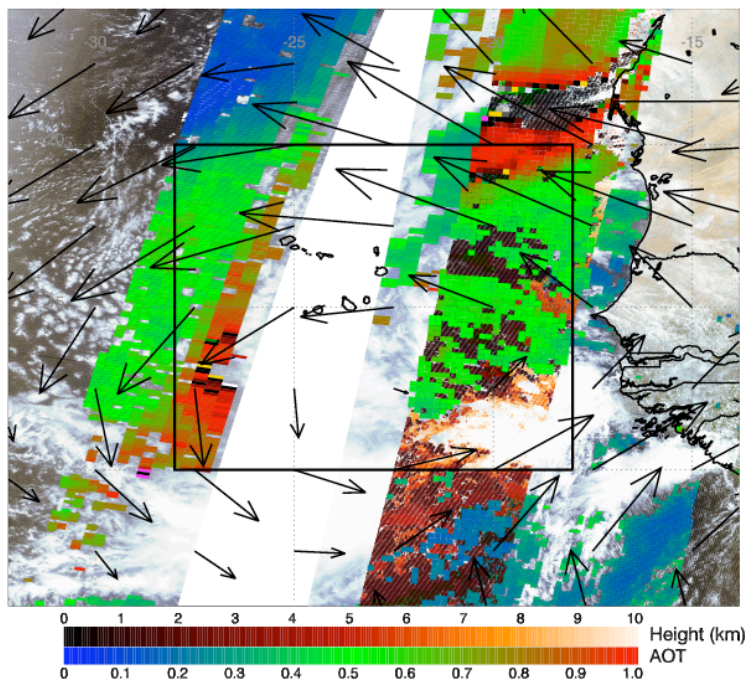


Fig. 10. Same as Fig. 7, but for the North Eastern Atlantic region on 23 July 2004.

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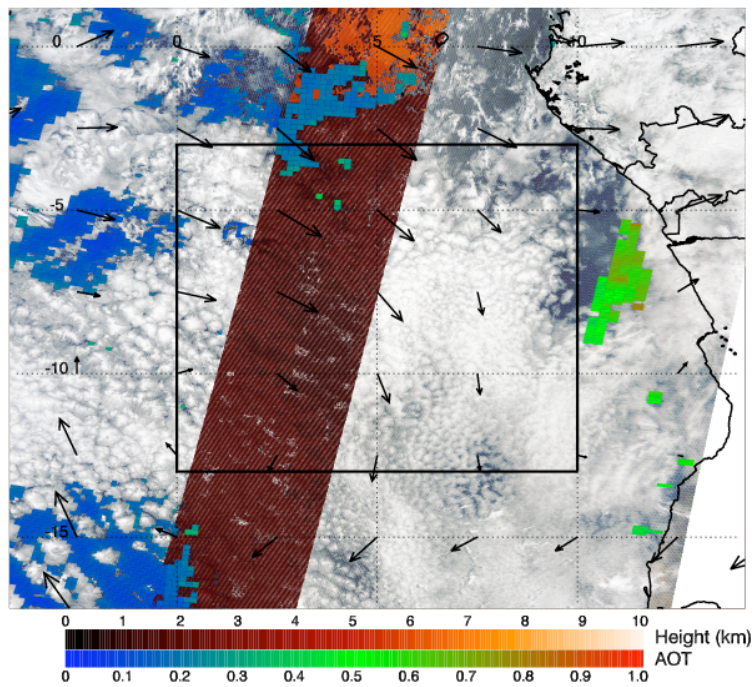


Fig. 11. Same as Fig. 7, but for the South Eastern Atlantic off Africa on 30 September 2004.

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