Assessment of aerosol optical property and radiative effect for the layer decoupling cases over the northern South China Sea during the 7-SEAS/Dongsha Experiment

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Abstract The aerosol radiative effect can be modulated by the vertical distribution and optical properties of aerosols, particularly when aerosol layers are decoupled. Direct aerosol radiative effects over the northern South China Sea (SCS) were assessed by incorporating an observed data set of aerosol optical properties obtained from the Seven South East Asian Studies (7-SEAS)/Dongsha Experiment into a radiative transfer model. Aerosol optical properties for a two-layer structure of aerosol transport were estimated. In the radiative transfer calculations, aerosol variability (i.e., diversity of source region, aerosol type, and vertical distribution) for the complex aerosol environment was also carefully quantified. The column-integrated aerosol optical depth (AOD) at 500 nm was 0.1–0.3 for near-surface aerosols and increased 1–5 times in presence of upper layer biomass-burning aerosols. A case study showed the strong aerosol absorption (single-scattering albedo (ω) ≈ 0.92 at 440 nm wavelength) exhibited by the upper layer when associated with predominantly biomass-burning aerosols, and the ω (>0.95) of near-surface aerosols was greater than that of the upper layer aerosols because of the presence of mixed type aerosols. The presence of upper level aerosol transport could enhance the radiative efficiency at the surface (i.e., cooling) and lower atmosphere (i.e., heating) by up to −13.7 and +9.6 W m⁻² per AOD, respectively. Such enhancement could potentially modify atmospheric stability, can influence atmospheric circulation, as well as the hydrological cycle over the tropical and low-latitude marginal northern SCS.

1. Introduction

Aerosols generated from both natural and anthropogenic sources are distributed throughout the atmosphere over a wide spatial scale because of air mass transport. The transport of aerosols from continental source regions to the oceans is a major reason for the spatiotemporal heterogeneity of aerosol characteristics over the oceans, which could appreciably influence regional climate [Ramanathan et al., 2001a]. Aerosols affect the Earth’s radiation budget directly by scattering and absorbing incoming solar and outgoing terrestrial radiation [Charlson et al., 1992] and indirectly by modifying the formation and microphysical properties of clouds [Penner et al., 2001]. Radiative forcing is a process that modifies the radiation budget of the climate system [Coakley et al., 1983; Ramanathan et al., 1989]. An accurate understanding of aerosol radiative forcing is crucial for accurately estimating the aerosol climate effect on a regional and global scale [Kedia et al., 2010].

The quantification of the radiative impact of atmospheric aerosols over a region requires information about their physical, chemical, and optical properties [Penner et al., 1994; Yoon et al., 2005] and spatiotemporal variability. On a regional scale, particularly over aerosol-prevalent regions such as South China Sea (SCS), aerosol radiative forcing can be greater than the radiative forcing of greenhouse gases by an order of magnitude and can appreciably affect a regional climate and hydrological cycle [Ramanathan et al., 2001a]. The net aerosol radiative forcing at the top of the atmosphere (TOA) over oceans has been estimated to be −5.5 to −6.7 W m⁻² [Haywood, 1999; Chou et al., 2002; Christopher and Zhang, 2004] with an uncertainty of approximately −1.7 W m⁻² [Christopher and Zhang, 2004]. A higher uncertainty can be expected in the presence of elevated aerosol layers comprised biomass burning and dust [Das et al., 2013]. Information on aerosol vertical distributions is thus crucial for estimating radiative effects. Still, the aerosol vertical distribution is uneven...
Two-layer structures of atmospheric aerosol transport have been observed at altitudes of 2–4 km over DSI, with the upper level transport attributed to biomass-burning activities in Indochina [Wang et al., 2013]. The direct and semidirect radiative effects of upper level smoke-haze particles in a column and in different vertical layers depend mainly on the loading of the smoke-haze (i.e., AOD associated with smoke haze) and single-scattering albedo (ω) of the smoke haze [Wang and Christopher, 2006]. The single-scattering properties of aerosols resulting from the presence of the smoke haze over the Southeast Asian Maritime Continent have not been well studied [Davison et al., 2004; Reid et al., 2013; Ge et al., 2014]; moreover, their radiative effects have not been fully characterized. Hence, this study seeks to estimate the aerosol optical properties in the two-layer transport structure by using simultaneously measured AODs, mass concentrations, and chemical composition; these data were obtained from those collected during the 7-SEAS/Dongsha Experiment.

Two-layer transport structure by using simultaneously measured AODs, mass concentrations, and chemical properties have been intensively studied during the Seven Southeast Asian Studies (7-SEAS)/Dongsha Experiment in 2010 [e.g., Wang et al., 2011, 2013; Chuang et al., 2013; Atwood et al., 2013; Bell et al., 2013; Tsay et al., 2013; Lin et al., 2013]. Asian dust transported to DSI is characterized by low-level transport and is well mixed with anthropogenic and maritime aerosols [Wang et al., 2011, 2013; Chuang et al., 2013]. Soil particles (mineral-dust) likely transported from the desert regions of northern China and Mongolia (e.g., Taklimakan and Gobi Deserts) have been observed [Bell et al., 2013]. Atwood et al. [2013] suggested that the aerosol optical depth (AOD) was largely uncorrelated with surface mass concentrations over DSI. This paper discusses the aerosol optical properties for the near-surface and free-tropospheric layers and their contribution to direct aerosol radiative effects.

Measurements and Data

Measurements were performed at DSI during the spring of 2010. Complementary data on AOD from the Aerosol Robotic Network (AERONET; http://aeronet.gsfc.nasa.gov/), recorded using a Cimel Sun-sky radiometer, were obtained; the data consisted of simultaneous measurements in spectral channels centered at 340, 380, 440, 500, 675, 870, 940, and 1020 nm [Holben et al., 1998]. Data on retrieved fine-mode AOD, coarse-mode AOD, ω, and asymmetry parameter (g) were also obtained from AERONET inversion database. Quality assured and cloud screened AERONET Level 2 data [Holben et al., 2006] were used in the present study. Aerosol profile information was obtained from an ultraviolet EZ Lidar system (wavelength: 355 nm; Leosphere Co.). Retrievals of vertical distribution of aerosol extinction and integrated column AOD from the EZ Lidar instrument are detailed in Lollì et al. [2011].

Aerosols were sampled daily at DSI during two periods, from 19–23 March and 10–19 April 2010. Details of the sampling procedure, analytical methods, and resolved chemical species of particulate matter 10 (PM_{10})...
(aerodynamic diameter \( \leq 10 \mu m \)) aerosols are presented in Chuang et al. [2013]. Profiles of the atmospheric state were obtained from the enhanced soundings (including the observed temperature, geopotential height, and humidity data at pressure levels) launched at the naval weather station on DSI.

3. Methodology

3.1. Source Region Identification: Back Trajectory Calculations

Seven-day back trajectory calculations were performed using the National Oceanic and Atmospheric Administration’s Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model [Version 4] [Draxler and Hess, 1998], for qualitatively assessing the transport of aerosols from their source regions to the receptor location. These trajectories started at 00:00 UTC from heights of 10, 100, 500, 1000, and 3000 m aboveground level, which were chosen to provide a good indication for the air mass origin within the MBL and above over DSI. The data were sorted into six different air mass pathways (see section 4.1). A total of 75 back trajectories studied \( N \) at five aforementioned heights during the 15 days (19–23 March and 10–19 April 2010) of measurement period and their relative frequency distribution were calculated.

3.2. Aerosol Optical Properties: Optical Properties of Aerosols and Clouds Model

AOD, \( \omega \), and \( g \) for boundary layer (BL) aerosols (i.e., BL-AOD or \( \omega^{BL} \), \( g^{BL} \)) were calculated from the Optical Properties of Aerosols and Clouds (OPAC) model [Hess et al., 1998]. In the calculation, we sought the most appropriate solutions with considering aerosol chemical compositions from surface measurements. The OPAC model can easily incorporate user-defined aerosol mixtures, aerosol vertical profile, wavelength, and relative humidity (RH). The model provides the wavelength-dependent AOD, \( \omega \), and \( g \) at eight different RH (0%, 50%, 70%, 80%, 90%, 95%, 98%, and 99%). The measured mean RH was \( \approx 80\% \) during the study period. The most suitable aerosols, as determined by aerosol source region, transport path, and previously reported data [e.g., Atwood et al., 2013; Campbell et al., 2013] over the northern SCS, were water-soluble, black carbon (BC), sea salt, and mineral dust aerosols. In the present study, water-soluble aerosols include organic carbon, \( Na^+ \), \( NH_4^+ \), \( K^+ \), \( Mg^{2+} \), \( Ca^{2+} \), \( NO_3^- \), and \( SO_4^{2-} \) aerosols. BC is defined as soot. The sea salt (accumulation mode) is obtained by multiplying the Cl\(^-\) concentration by a value of 1.8 [Watson, 2002], and the mineral dust (accumulation mode) is assumed to account for the remaining part of PM\(_{10}\) aerosol.

The number densities of aerosol components were estimated from the respective measured mass concentration and were provided as input to the OPAC model. The relative fraction of each component measured at the surface can represent the aerosol composition throughout the MBL. For simplification, we assumed that the aerosols within the MBL were externally mixed (no physical or chemical interaction between the components) and spherical in nature. Campbell et al. [2013] also observed that the near-surface aerosols, within 0.5 and 1.5 km, were mostly spherical over the Southeast Asia and the Maritime Continent by using linear depolarization of Cloud-Aerosol Lidar with Orthogonal Polarization backscatter signals. The distribution of aerosols with height in the OPAC model is described by means of exponential profiles given by

\[
N(h) = N(0)e^{-(h/z)},
\]

where \( h \) is the altitude above the ground in kilometers, \( N(0) \) is the number concentration at the surface layer, and \( z \) is the scale height in kilometers [Hess et al., 1998]. For the aerosol vertical profile, we used the same exponential function as in OPAC, but with appropriate MBL height. The mean MBL heights were obtained as the top of a surface-based inversion [e.g., Bradley et al., 1993; Seidel et al., 2010] from enhanced soundings daytime values.

The AOD of each component was derived, and its relative contribution to the BL-AOD was estimated. The AOD of the background atmosphere (BK-AOD; i.e., AOD of the free-troposphere (between the top of the MBL and a height of 12 km) and the midstratosphere (height range of 12–35 km)) was also estimated. It is worth mentioning here that BK-AOD represents the aerosol extinction without the upper layer biomass-burning plume. The AOD of the upper layer aerosols (BB-AOD), mainly attributed to biomass-burning activities in Indochina (see section 4.1), was estimated by subtracting all of the AODs (BL-AOD and BK-AOD) from the total AOD (obtained from AERONET).
Further, $\omega$ and $g$ of upper layer biomass-burning aerosols (i.e., $\omega^{\text{Upper}}$ and $g^{\text{Upper}}$) were estimated from the weighting function as

$$\omega^{\text{Total}} = \omega^{\text{Upper}} \times \frac{\tau^{\text{Upper}}}{\tau^{\text{Total}}} + \omega^{\text{BL}} \times \frac{\tau^{\text{BL}}}{\tau^{\text{Total}}}$$

(2)

and

$$g^{\text{Total}} = g^{\text{Upper}} \times \frac{\omega^{\text{Upper}}}{\omega^{\text{Total}}} \times \frac{\tau^{\text{Upper}}}{\tau^{\text{Total}}} + g^{\text{BL}} \times \omega^{\text{BL}} \times \frac{\tau^{\text{BL}}}{\tau^{\text{Total}}}$$

(3)

where $\tau^{\text{Total}}$, $\omega^{\text{Total}}$, and $g^{\text{Total}}$ are the AOD, $\omega$, and $g$ corresponding to the total column integrated aerosols, obtained from AERONET over DSI. $\tau^{\text{Upper}}$, and $\tau^{\text{BL}}$ are the upper layer AOD and BL-AOD, respectively.

### 3.3. Direct Aerosol Radiative Effect Estimation

The direct aerosol radiative effect was estimated using the methodology of Wang et al. [2010]. AOD and extinction, $\omega$, and $g$ are the key parameters used in the radiative transfer calculations, obtained from data sets constrained by our measurements. Detailed aerosol and meteorological profile were also used. The direct aerosol radiative effect (DARE; $W/m^2$) was defined as the change in the net radiation under clear-sky conditions, and it is given by

$$\text{DARE}(p) = F_{\text{wa}}(p) - F_{\text{ns}}(p).$$

(4)

where $F_{\text{wa}}(p)$ and $F_{\text{ns}}(p)$ represent the net downward flux in the presence and absence of aerosols, respectively, at pressure level $p$. The value for DARE was estimated for both TOA as well as at the surface (SFC). DARE$_{\text{TOA}}$ and DARE$_{\text{SFC}}$ denote the DARE at the TOA and the surface, respectively. DARE$_{\text{ATM}}$ (semidirect forcing), which is the solar heating of the atmosphere due to the presence of aerosols, was computed as the difference between DARE$_{\text{TOA}}$ and DARE$_{\text{SFC}}$. Finally, the semidirect aerosol heating rate of an atmospheric layer between $p$ and $p + \Delta p$, $\Delta Q(p)$, is estimated by defining it as being proportional to DARE$(p) - \text{DARE}(p + \Delta p)$.

In this study, the DARE estimations were performed in three scenarios (i.e., BL, multilayer, and a representative day cases) based on input aerosol data. For the BL case, considered as a single layer (i.e., no sublayers within the MBL height), the $\tau^{\text{BL}}$, $\omega^{\text{BL}}$, and $g^{\text{BL}}$ were used to characterize the DARE estimation. For the multilayer case calculation, we followed Wang et al. [2010]. The mean values of $\omega^{\text{Total}}$ and $g^{\text{Total}}$ were applied throughout each layer, and $\tau^{\text{Total}}$ was proportionally distributed within each layer based on the aerosol extinction profile. For a representative day case, we assessed the DARE estimate by using the most complete aerosol information (i.e., $\tau^{\text{BL}}$, $\omega^{\text{BL}}$, $g^{\text{BL}}$, $\omega^{\text{Upper}}$, $\omega^{\text{Upper}}$, and $g^{\text{Upper}}$) in the radiative transfer calculation. Our experiment design allows us to know the sensitivities of each data input to the DARE calculations.

### 4. Results and Discussions

#### 4.1. Back Trajectory Analysis

The seven-day back trajectories on the measurement days were used for examining the air mass transport paths as they influenced the surface and total columnar aerosol loading measured over DSI. Major back trajectory paths (Figure 1a), indicating the influence of air masses originating from different source regions on the aerosols at the study site, were as follows:

1. Path A: These back trajectories originated from elevated regions over northern China and Mongolia and covered the longest distance through the coastal areas of China;
2. Path B: Originating from mainland China, but passed mainly along the Chinese coast and over a distance considerably shorter than Path A;
3. Path C: Originating mainly from the western Pacific;
4. Path D: Originating locally in the vicinity of the northern SCS;
5. Path E: Originating from the Indochina region (comprising Cambodia, Laos, Myanmar, and Thailand);
6. Path F: Originating from the maritime western Pacific and passed through the Philippines and the northern SCS.

Figure 1b shows the relative frequency distribution (in percentage) of the six types of back trajectory paths identified over DSI at lower heights (10, 100, and 500 m) and higher heights (1000 and 3000 m). Back trajectories at the lower heights include Paths A (36%), C (27%), B (20%), D (11%), and F (7%). The back trajectories at 1000 m were...
mostly oceanic; they originated from the western Pacific (Path C, 40%) or the maritime western Pacific and passed through the Philippines and northern SCS (Path F, 47%), indicating that air masses in this layer were likely to contain sea-salt aerosols. At 3000 m, most back trajectories over DSI originated from Indochina (Path E, 80%), and a very small number of trajectories originated from oceanic regions (i.e., Paths C, D, and F; 7% each).

Figure 1. (a) Possible paths based on 7 day backward trajectories over DSI at different heights. (b) Relative frequency (in percentage) of six types of back trajectory paths (N is the number of back trajectories studied) at the different heights indicated. (c) MODIS fire counts over Indochina and Southeast Asia during March and April 2010 (http://firms.modaps.eosdis.nasa.gov/firemap/).

Fire count data (Figure 1c) from Moderate Resolution Imaging Spectroradiometer (MODIS; https://firms.modaps.eosdis.nasa.gov/firemap/) indicate that the primary biomass-burning source region was northern Indochina during the study period. Open burning emissions from Indochina mostly contribute to aerosols in upper atmospheric layers over the SCS [e.g., Tsai et al., 2012; Lin et al., 2013; Yen et al., 2013; Tsay et al., 2013; Wang et al., 2015]. The presence of biomass-burning plume with a higher depolarization ratio was observed by the same lidar used in this study [Wang et al., 2013].

4.2. Aerosol Optical Properties Determined From AERONET Observations

Figure 2a shows the daily average AOD, as the sum of the fine and coarse modes, observed at 500 nm for the period of study. Clearly, the main contribution is from fine-mode aerosols, contributing 76%–99% to the total
AOD. The mean AOD at 500 nm was 0.33 ± 0.22 (0.32 ± 0.22, 0.02 ± 0.03), 0.49 ± 0.25 (0.45 ± 0.24, 0.05 ± 0.04), and 0.41 ± 0.25 (0.38 ± 0.24, 0.03 ± 0.04) for March, April, and the entire study period, respectively. By comparison, the AOD at 500 nm was 0.26 ± 0.06 (Angström exponent (AE) = 0.61 ± 0.28) over Sanya Bay, a tropical marine bay in SCS, during the spring [Wang et al., 2011]. Tang et al. [2014] reported an AOD value lower than 0.3 over East Sea and the SCS. Furthermore, an AOD value of approximately 0.3 was observed over the Arabian Sea around premonsoon (March–May) season [Satheesh et al., 1999]. The AOD at DSI was generally higher and more variable than those at the aforementioned locations, indicating relatively complex conditions during our study period.

Angström exponent (AE) values estimated by spectral AOD at wavelengths of 440 and 870 nm during the study period are shown in Figure 2b. The daily mean AE values ranged between 1.08 and 2.52, with averages of 1.75 ± 0.46, 1.42 ± 0.19, and 1.59 ± 0.38 during March, April, and the entire study period, respectively. By comparison, background maritime aerosol has AE values in the range of 0.77 to 1.1 [Kaufman et al., 2001]. The typical AE value for fine particles such as sulfates or carbonaceous aerosols is between 1 and 2 [Liousse et al., 1995; Leon et al., 2001]. AE is widely used to characterize columnar aerosol size distributions and predominantly fine aerosol size distributions, such as those associated with urban pollution, and biomass burnings generally correspond to a high AE value. By contrast, coarser aerosols such as sea-salt and dust aerosols are associated with a low value. The daily AE estimates thus indicated its predominant contribution to columnar aerosol loading from fine mode aerosols, which is confirmed by the AOD trend. Fine particles observed over DSI primarily originate from continental anthropogenic aerosols that have been transported over a long distance, whereas coarse aerosols are either sea-salt or transported mineral dust aerosols [Wang et al., 2011].

4.3. Aerosol Vertical Profiles

Aerosol profiles from lidar observations, which were available for seven days (i.e., 22 and 23 March and 11, 12, 13, 15, and 19 April), were analyzed to determine the aerosol vertical distribution over DSI. Figure 3a shows that the aerosol over DSI in the spring was distributed in atmospheric layers as high as 5 km and that a layer associated with biomass-burning aerosols was often present at a height range of 2–4 km. Wang et al. [2013] also reported the presence of a distinct aerosol layer of Indochinese haze plume on the basis of an attenuated backscattering ratio and normalized linear depolarization ratio obtained from the same lidar system. In this study, we retrieved additional aerosol extinction profiles and accumulative AOD for the abovementioned days to use in radiative transfer calculations. In addition, the vertical profile of AOD at 355 nm calculated from aerosol extinction coefficient is shown in Figure 3b. In general, upper layer biomass-burning aerosols contribute considerably to the aerosol loading of the entire column. The AOD from the lidar observations for the above-inversion (fractional AOD: 0.11–0.71; relative contribution to the total AOD: 33%–77%) was relatively higher over DSI than that of the below-inversion (0.17–0.31; 23%–67%), except 22 March, when the above-inversion AOD (0.01; 4%) contributed significantly less as compared with the below-inversion (0.19; 96%) to the total AOD.
4.4. Estimated Aerosol Optical Properties

The daily mass concentrations of surface PM$_{10}$ (=total aerosol over the study site) ranged from 13.2 $\mu$g m$^{-3}$ on 15 April to 94.1 $\mu$g m$^{-3}$ on 21 March, with an average concentration of 26.5 ± 19.4 $\mu$g m$^{-3}$ for the study period [Chuang et al., 2013, Figure 5]. The highest PM$_{10}$ mass concentration was associated with a dust storm on 21 March. The daily relative contribution of aerosol components to the total aerosol surface mass concentration (Figure 4a) was 26%–70% from water soluble, 1%–6% from soot, 1%–48% from sea salt, and 11%–66% from mineral dust during the study period. Among the average contributions of the different aerosol components to the near-surface aerosol mass, the average contribution of water soluble (≈55%) was the highest, followed by mineral dust (≈23%), sea-salt (≈20%), and soot (≈4%) aerosols.

The MBL height (Figure 4b) varied from 0.8 to 2 km. The relative contributions of the AOD values of different components to the total BL-AOD (Figure 4c) were 61%–92% from water-soluble, 2%–9% from soot, 1%–34% from sea salt, and 1%–13% from mineral dust aerosols during the study period. Among the average relative contributions of the different components to the BL-AOD over DSI, the average relative contribution of water-soluble aerosols (≈77%) was highest, followed by sea-salt (≈16%), soot (≈4%), and mineral dust (≈3%) aerosols.

The BK-AOD and BB-AOD at 500 nm were in the ranges of 0.014–0.023 and 0.04–0.5, respectively, during the study period. Furthermore, the relative contributions of different AODs (BL-AOD, BK-AOD, and BB-AOD) to the total AOD (obtained from AERONET) were also estimated. Figure 4d shows the relative contributions of different AODs to the total columnar aerosol loading over DSI. The contribution of the BL-AOD to the total AOD over the island was 20%–88%, followed by a BB-AOD (20%–76%), and a BK-AOD (3%–18%). The contributions of the below-inversion and above-inversion AODs to the total AOD estimated from the lidar observations at 355 nm were 23%–96% and 4%–77%, respectively, during the study period. The estimated relative percentage contributions from the BL-AOD and upper layer AOD (sum of BB-AOD and BK-AOD) to the total AOD matched the AOD estimates (below-inversion and above-inversion AODs) from lidar observations, except 13 April, when the BL-AOD was slightly overestimated by the OPAC model.

We also calculated $\omega$ and $g$ for below and above the inversion layer, because these values are critical for partitioning radiative forcing between the atmosphere and the underlying surface [Chou et al., 2006]. $\omega$ is proportional to the ratio of the scattering coefficient to the extinction coefficient (the sum of the scattering and absorption coefficients). Parameter $g$ can be expressed as the cosine-weighted average of the scattering phase function and contains information on aerosol size and aerosol scattering properties. The mean $\omega_{\text{Total}}$
Total of the total aerosols, obtained from AERONET over DSI at 440 nm, were 0.94 and 0.72, indicating the presence of moderate to strongly absorbing aerosols. The estimated mean $\omega_{BL}$ and $g_{BL}$ of the BL aerosols were 0.95 and 0.73 at 440 nm over DSI. Error may be caused in the radiative transfer calculations by the assumption of a constant columnar average of $\omega$ and $g$ throughout two-layer structure of atmospheric aerosol transport [Wang et al., 2010]. The differences in $\omega$ and $g$ for the near-surface and upper layer aerosols can be considered in terms of different source regions and transport pathways at different height layers over DSI. The different $\omega$ was observed for the near-surface and upper layer aerosols over Niamy [Osborne et al., 2008]; Chung-Li, Taiwan [Wang et al., 2010]; Gwangju, South Korea [Shin et al., 2014]; and Indian Ocean [Höpner et al., 2016]. A representative day (11 April) was thus selected because of the availability of AERONET data and also because of its clear two-layer structure and heavy aerosol loading.

The BB-AOD was the highest on 11 April, which is attributed to the maximum transport of pollutants from the Indochina region. The estimated $\omega_{BL}$ and $g_{BL}$ of the BL aerosols on that day were 0.95 and 0.72 at 440 nm over DSI. The estimated $\omega_{Upper}$ and $g_{Upper}$ of upper layer biomass-burning aerosols on that day were 0.92 and 0.74 at 440 nm, respectively, suggesting the presence of relatively higher absorbing smoke-haze plumes in the upper layer over northern SCS. The mean $\omega_{Upper}$ and $g_{Upper}$ values are in good agreement with values (shown in Table 1) measured from an AERONET site at Mount Lulin (23.51°N, 120.92°E; 2862 m above mean sea level; ≈600 km northeast of DSI), which is also strongly influenced by the biomass-burning aerosols transported from Indochina [e.g., Lin et al., 2013; Chuang et al., 2014; Chuang et al., 2015].

### 4.5. Clear-Sky Shortwave Aerosol Radiative Effects

Scatterplots of the daily mean DARE$_{TOA}$ and DARE$_{SEC}$ versus AOD at 500 nm are shown in Figure 5. Because of limited data availability and sky conditions, estimations were computed only for 7 days (i.e., 22 and 23 March and 11, 12, 13, 15, and 19 April) in the radiative transfer model. Each point (circles) represents radiative calculations on the corresponding day, obtained using the daily-mean spectral AOD, $\omega$, and $g$. Linear regression lines are also shown. The correlation between the DARE and the AOD at 500 nm (with a negative correlation coefficient of greater than 0.94) was found to be strongly linear in both cases (i.e., BL and multilayer cases). The large deviation at some points from the linear regression line is due to the broad ranges of the observed
ω and g values. The slight positive deviation of DARE from the linear regression is due to the large ω and g; conversely, the weak negative deviation of DARE is due to the relatively small ω and g.

Table 2 shows the mean values of the radiative estimates over DSI during the study period for the all three scenarios (i.e., BL, multilayer, and a representative day cases). Higher DARE_TOA and DARE_SFC were observed for the layer-decoupling cases than that of near-surface aerosols. The relatively absorbing nature of the upper layer biomass-burning aerosols was accountable for the heating of the lower atmospheric column (free troposphere) and also resulted with considerably more negative DARE_SFC than DARE_TOA. The mean values of DARE_TOA and DARE_SFC over DSI (Table 2) were –4.8 and –7.9 W m\(^{-2}\), respectively, for the BL case, whereas they were –10.1 and –18.6 W m\(^{-2}\) for the multilayer case. By comparison, DARE_TOA and DARE_SFC over Chung-Li (a rural region in northern Taiwan) were estimated as –11.2 and –18.7 W m\(^{-2}\), respectively, for aerosols with two-layer transport [Wang et al., 2010]. Semidirect forcing, DARE_ATM due to absorption of solar radiation by aerosols in the atmosphere over DSI, was +3.2 and +8.5 W m\(^{-2}\).

| Table 1. Spectral Optical Properties of BL and Upper Layer Aerosols at DSI and Compared With Mount Lulin Data |
|---------------------------------------------------------------|-----|-----|-----|-----|-----|
| Wavelengths (nm)                                             | 440 | 500 | 675 | 870 | 1020 |
| BL case (11-Apr-2010)                                         |     |     |     |     |     |
| AOD                                                          | 0.25 | 0.22 | 0.14 | 0.10 | 0.08 |
| ω                                                           | 0.95 | 0.95 | 0.94 | 0.92 | 0.91 |
| g                                                            | 0.72 | 0.71 | 0.69 | 0.66 | 0.65 |
| Upper layer (11-Apr-2010)                                    |     |     |     |     |     |
| AOD                                                          | 0.61 | 0.52 | 0.32 | 0.19 | 0.15 |
| ω                                                           | 0.92 | 0.91 | 0.90 | 0.90 | 0.88 |
| g                                                            | 0.74 | 0.68 | 0.62 | 0.54 | 0.51 |
| Mount Lulin (02-Apr-2013)                                    |     |     |     |     |     |
| AOD                                                          | 0.60 | 0.50 | 0.31 | 0.18 | 0.14 |
| ω                                                           | 0.93 | 0.93 | 0.92 | 0.90 | 0.88 |
| g                                                            | 0.70 | 0.67 | 0.64 | 0.59 | 0.55 |

Figure 5. Scatterplots of DARE at the TOA versus the AOD at 500 nm for (a) the BL case and (b) the multilayer case. (c and d) Same as Figures 5a and 5b but for DARE at the surface. The circles represent radiative calculations made for 7 days (i.e., 22 and 23 March and 11, 12, 13, 15, and 19 April) during 2010.
for BL and multilayer cases, respectively. The radiative effect of Indo-Asian aerosols over the Indian Ocean during the INDOEX period was estimated to cause a large negative forcing at the surface (−20 W m⁻²) and relatively large atmospheric heating [Ramanathan et al., 2001b].

The aerosol radiative efficiency (ARE), defined as the change in DARE per unit AOD at 500 nm, is a useful parameter for quantifying and comparing aerosol radiative effects at different places under different aerosol conditions [Satheesh and Ramanathan, 2000]. ARE_TOA (ARE at the TOA) and ARE_SFC (ARE at the surface) were estimated from the slopes of the linear regression lines shown in Figure 5. The overall mean values of ARE_TOA (ARE_SFC) were −23.8 (−50.6) and −27.5 (−54.7) W m⁻² for BL and multilayer cases, respectively, over DSI.

### 4.6. DARE Enhancement Resulting From Upper Layer Transport

We investigated changes in DARE and the atmospheric heating rate by considering the vertical distribution of aerosol optical properties over DSI on a representative day (11 April). DARE_TOA values on 11 April over DSI were found to be −20.9 and −5.7 W m⁻² for the two-layer and BL cases, respectively. Similarly, DARE_SFC value was −40.9 and −10 W m⁻² for two-layer and BL cases, respectively. The presence of moderately absorbing upper layer biomass-burning aerosols increased the magnitude of DARE_TOA and DARE_SFC by a factor of 4 (ratio between DARE_TOA for two-layer and BL cases was ≈3.6, while ratio between DARE_SFC for two-layer and BL cases was ≈4.1 on 11 April). The more negative DARE_SFC indicates the significant decrease of the solar radiation at the surface that is enhanced by atmospheric absorption. The mean values of ARE_SFC (ARE_ATM) on 11 April were −46.4 (+19.8) and −60.1 (+29.4) W m⁻² for BL case and two-layer aerosol transport, respectively.

Absorbing aerosols can have a warming effect in the atmosphere and a cooling effect at the surface. A higher ARE_SFC for two-layer aerosol transport indicated the enhancement of the cooling effect at the surface because of the transport of absorbing biomass-burning aerosols in the upper layer. The presence of upper level aerosol transport enhanced the ARE_SFC and ARE_ATM values by up to −13.7 (difference between the ARE_SFC for two-layer (−60.1 W m⁻²) and BL (−46.4 W m⁻²) cases; see Table 2) and +9.6 (difference between ARE_ATM for two-layer (+29.4 W m⁻²) and BL (+19.8 W m⁻²) cases; see Table 2) W m⁻², respectively. ARE_ATM for two-layer aerosol transport over DSI (+29.4 W m⁻²) was considerably higher than that over Chung-Li (+16.8 W m⁻²) reported by Wang et al. [2010]) by a factor of ≈2, indicating the presence of moderately absorbing biomass-burning aerosols in the upper layer of northern SCS than that over northern Taiwan.

Figure 6 shows the vertical distribution of the mean ΔQ (K d⁻¹) for the BL case and two-layer aerosol transport on 11 April over DSI. The parameter ΔQ was estimated to be as high as 1.3 K d⁻¹ over DSI because of the maximum absorption of upper layer biomass-burning aerosols. The decrease in ΔQ (below 2 km) was due to the presence of absorbing upper layer aerosols. The ΔQ value for BL case indicated that the upper layer absorption of aerosols enhanced the upper layer ΔQ value and reduced the near-surface ΔQ value. Solar absorption by upper layer biomass-burning aerosols in the atmosphere can lead to a substantial reduction in the solar radiation reaching to the ocean surface. Enhanced surface cooling can decrease the rate of evaporation over marginal northern SCS, which may also affect the tropical rainfall. Enhanced atmospheric absorption by upper layer biomass-burning aerosols, as shown in our findings for two-layer aerosol structures over the SCS, can affect atmospheric stability and regional climates.

### 5. Conclusions

In this study, we estimate the direct aerosol radiative effect (DARE) values by modulating the vertical distribution of aerosol optical properties, particularly in the scenario of decoupled aerosol layers over the northern South China Sea (SCS). The study seeks to investigate, in detail, the two-layer structure of aerosol
transport, in terms of aerosol types and optical properties, and their effects on regional climate. In addition, DARE values were estimated by synergizing the observed data set for various aerosol properties (physical, chemical, and optical) obtained during the Seven South East Asian Studies (7-SEAS)/Dongsha Experiment in spring 2010 with a radiative transfer model. The long-range, high-altitude transport of biomass-burning aerosols from the Indochina region was observed at 3000 m and above. Among the contributions of the different aerosol components to the aerosol optical depth (AOD) due to boundary layer (BL) aerosols (i.e., BL-AOD) over DSI, the mean relative contribution of water-soluble (≈77%) was the highest, followed by sea-salt (≈16%), soot (≈4%), and mineral dust aerosols (≈3%). The BL-AOD at 500 nm was 0.1–0.3 but increased 1–5 times in the presence of upper layer biomass-burning aerosols. The relative contribution of BB-AOD (AOD due to upper layer biomass-burning aerosols) to total AOD was 20%–76% (mean: ≈41%).

A case study (distinct two-layer transport with heavy aerosol loading) shows the presence of relatively stronger absorbing biomass-burning aerosols (ω and g for upper layer aerosols at 440 nm; ω upper: =0.92, g upper: =0.74) in upper layer compared with for near-surface aerosols (ω and g for BL aerosols at 440 nm; ω BL: =0.95, g BL: =0.72). DARESEC (DARE at surface), for the representative case, was −10 and −40.9 W m⁻² for the BL and two-layer aerosol transport cases, respectively. Similarly, DARETOA (DARE at TOA) was −5.7 and −20.9 W m⁻² for the BL and two-layer aerosol transport, respectively. The upper layer biomass-burning aerosols decrease the solar radiation by a factor of 4 both at the surface and at the TOA. It could also enhance the radiative efficiency at the surface (i.e., cooling) and lower atmosphere (i.e., heating) by up to −13.7 and +9.6 W m⁻² per unit AOD, respectively. Moreover, the atmospheric heating rate associated with the absorption of upper layer biomass-burning aerosols was estimated to be as high as 1.3 K d⁻¹.

Our research first demonstrates that the effect of aerosols on surface cooling and free troposphere warming can be enhanced by the transport of biomass-burning plumes over the tropical northern SCS, which in turn influence regional atmospheric stability as well as the hydrological cycle. Comprehensive findings from this study provide key information on aerosol types, source origins, optical properties, and radiative impacts for the two-layer aerosol transport of Asian continental outflows over a tropical and low-latitude marginal sea. This study also gives some thoughtful insights to the experts for the conclusion of the 7-SEAS field campaign.

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