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On factors controlling marine boundary layer aerosol optical depth

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Abstract

Sea spray aerosol is one of the largest natural contributors to the global aerosol loading and thus plays an important role in the global radiative budget through both direct and indirect effects. Previous studies have shown either strong or weak relationships between marine boundary layer (MBL) aerosol optical depth (τ) and the near-surface wind speed. However, the marine τ is influenced by a wide range of factors. This study attempts to examine extra contributing factors beyond wind to better characterize MBL τ variations over the global ocean by using 4 year A-train data (2006–2010). The results show that among many factors controlling MBL τ, surface wind speed and MBL depth are the two most important factors. This suggests that not only mechanical production of sea spray particles driven by near-surface wind processes but also vertical redistribution driven by turbulent and shallow convective mixing in the MBL controls MBL τ variations. A new two-parameter parameterization of τ was derived based on satellite measurements. Evaluations with independent data show that the new parameterization improves the prediction of MBL τ. The comparisons between the Fu-Liou radiative transfer model calculations and Aqua Clouds and the Earth's Radiant Energy System observations showed that the new parameterization improves the estimation of aerosol radiative forcing.

1. Introduction

Sea salt is one of the largest natural contributors to the global aerosol loading and thus plays a significant role in global climate [Intergovernmental Panel on Climate Change, 2007]. It dominates submicron and supermicron scatterers in most oceanic regions and the marine boundary layer (MBL) particulate mass concentration in remote oceanic regions [Sievering et al., 2004]. Previous researches showed that sea salt accounts for 50% of the local light scattering over the oceans [Penner et al., 2001] and contributes to 44% of the global aerosol optical depth [O’Dowd and de Leeuw, 2007]. The top-of-atmosphere (TOA) clear-sky global-annual radiative forcing due to sea salt is estimated ranging from −0.6 to −5.03 W/m² according to different models [Winter and Chylek, 1997; Haywood et al., 1999; Jacobson, 2001; Grini et al., 2002; Ayash et al., 2008; Ma et al., 2008]. Sea salt could also act as the dominant source of cloud condensation nuclei over the remote oceanic regions and affects radiative budget by modifying the radiative properties and lifetimes of clouds, the so-called indirect radiative effect [Albrecht, 1989; Twomey, 1974, 1977]. However, sea-salt aerosol is still one of the most poorly constrained aerosols in models according to the recent global model comparison by the Aerosol Comparisons between Observations and Models project [Textor et al., 2006; Kinne et al., 2006].

Recently, understanding the effect of surface wind speed on the marine aerosol optical depth (τ) has gained increasing interests [Mulcahy et al., 2008; Glantz et al., 2009; Huang et al., 2010; Lehahn et al., 2010; Kiliyanpilakkil and Meskhi, 2011; Smirnov et al., 2012; Anderson et al., 2012]. The surface wind speed is the major production mechanism of natural marine aerosols [Blanchard and Woodcock, 1957; Smith et al., 1993; Clarke et al., 2003; O’Dowd and de Leeuw, 2007]. Usually, the surface wind speed at 10 m (U10) is taken as a reference. Most of these particles are produced by bubble bursting within foamy whitecaps. It is estimated that whitecap formation onset occurs at wind speeds of ~4 m/s [O’Dowd and de Leeuw, 2007]. When wind speeds are above ~9 m/s, sea salt can be produced by the direct tearing of wave crests. The generation of sea spray aerosols [Nilsson et al., 2001], as well as the consequent particle concentration [Hoppel et al., 1990; O’Dowd et al., 1997; Glantz et al., 2004] and size distribution [Hoppel et al., 1990; Gong et al., 1997] are all strongly dependent on the U10.

Quantifying the dependence of τ on wind speed can help us better understand the production, evolution, and removal processes of marine aerosols and improve other researches including the atmospheric correction in ocean color studies and the biogeochemical cycles [Zibordi et al., 2009; Janet et al., 2011; Harmel and Chami, 2011].
Although lots of studies have been performed and different $\tau$-wind relationships have been developed [see Smirnov et al., 2012, Table 3], reliable relationships to quantify the dependence of $\tau$ on wind speed are still lacking as highlighted by large differences among published relationships. Smirnov et al. [2003], Lehahn et al. [2010], and Huang et al. [2010] presented linear relationships, whereas others gave power law dependencies [e.g., Glantz et al., 2009; Mulcahy et al., 2008] or even a logarithmic relationship [e.g., Kiliyanpilakkil and Meskhidze, 2011]. Here and after, K2011 denotes Kiliyanpilakkil and Meskhidze [2011], H2010 denotes Huang et al. [2010], L2010 denotes Lehahn et al. [2010], G2009 denotes Glantz et al. [2009], M2008 denotes Mulcahy et al. [2008], and SM2003 denotes Smirnov et al. [2003]. For parameterizations based on ground-based $\tau$ observations, the $\tau$ dependence on moderate wind speed (about 4–10 m/s) are quite similar and are weaker than the dependencies derived from satellite observations [Lehahn et al., 2010; Smirnov et al., 2012]. The largest difference among different parameterizations occurs under low-wind conditions (<4 m/s). Lehahn et al. [2010] found that the averaged background $\tau$ under low-wind conditions shows weak dependency on the surface wind speed, but with large seasonal and spatial variations. Therefore, there should be other factors controlling the behavior of marine $\tau$ and resulting in the observed variations. Furthermore, the previous studies were performed based on data collected with different instruments over different locations in different time periods, and different meteorological data from model, ground-based observations, or satellite-based observations were used. Thus, it is hard to comprehensively compare with each other and to apply these parameterizations over the global ocean [Huang et al., 2010].

While it is difficult to comprehensively quantify the $\tau$-wind relationships using passive remote sensing and ground-based measurements [Kiliyanpilakkil and Meskhidze, 2011], new A-train satellite [NASA, 2003] measurements provide an opportunity to solve the problem. With near-simultaneous observations/estimations of a wide variety of parameters including $U_{10}$, sea surface temperature (SST), aerosols, and clouds, these A-train measurements allow us to explore the effects of different factors on MBL $\tau$ and to establish a reliable relationship over the global ocean.

This study explores additional contributing factors, including marine boundary layer height ($\Delta H$), SST, and lower troposphere stability (LTS, defined as the potential temperature difference between 700 hPa and the surface) [Slingo, 1987; Klein and Hartmann, 1993; Klein, 1997; Wood and Hartmann, 2006; Remer et al., 2009], to improve the $\tau$-wind relationships over the global ocean. The data sources and analysis method are introduced in section 2. The results are presented in section 3, and their implications on aerosol direct radiative forcing estimations are discussed in section 4. The key findings are summarized in section 5.

2. Data and Methodology

Multiple satellite and operational meteorology data sets, including CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations), CloudSat, ECMWF (European Centre for Medium-Range Weather Forecasts), and AMSR-E (Advanced Microwave Scanning Radiometer–EOS), are collocated to carry out the study. With more reliable cloud masks from combining CALIPSO and CloudSat measurements and vertical aerosol properties from CALIPSO lidar measurements, pure marine aerosol layers under clear sky can be reliably identified. Then, MBL $\tau$ at 532 nm are retrieved and evaluated, and its dependency on the external factors is examined.

2.1. Data

Multiple satellite remote sensing and operational meteorology data sets over remote oceans during the period of June 2006 to December 2010 are used in this study, including the following:

1. CALIPSO level 1B data. CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) is a polarization-sensitive lidar capable of measuring backscatter intensity at wavelengths of 532 nm and 1064 nm [Winker et al., 2009], with a 333 m along-track footprint. CALIOP level 1B data provide calibrated and geolocated 532 nm and 1064 nm total attenuated backscatter and 532 nm perpendicular polarization component [Hostetler et al., 2006], which are used for MBL aerosol studies.

2. CloudSat 2B-GEOPROF product. CloudSat carries a 94 GHz cloud profiling radar (CPR). The CloudSat antenna pattern provides an instantaneous footprint of approximately 1.3 km (at mean sea level). It has 125 vertical bins with a bin size of about 240 m. The 2B-GEOPROF product contains the cloud mask information indicating where hydrometeors occur [Mace, 2007].
3. ECMWF-AUX (auxiliary) data. The ECMWF-AUX data set is an intermediate product that contains the set of ancillary ECMWF (AN-ECMWF) state variable data interpolated to each CloudSat CPR bin (http://www.cloudsat.cira.colostate.edu/ICD/ECMWF-AUX/ECMWF-AUX_PDICD_3.0.pdf). AN-ECMWF has ~40 km horizontal resolution and 60 vertical levels (http://www.cloudsat.cira.colostate.edu/ICD/AN-ECMWF/AN-ECMWF_doc_v7.pdf). The temporal and spatial resolutions of ECMWF-AUX are better than those of other available products such as National Centers for Environmental Prediction Reanalysis data. The ECMWF-AUX contains temperature and pressure profiles from the ECMWF operational analysis interpolated in time and space to the CloudSat track [Partain, 2004]. The temperature and pressure profiles will be used to calculate the molecular backscattering and LTS.

4. AMSR-E level 3 daily ocean products version 5. The daily AMSR-E ocean products are produced by Remote Sensing Systems (http://www.remss.com/). The products contain several important geophysical parameters retrieved from observations collected by the AMSR-E instrument, including SST and \( U_{10} \). The orbital data is mapped to 0.25° grid box and is divided into two maps based on ascending and descending passes. Preliminary validations of AMSR-E SST and \( U_{10} \) [Wentz et al., 2003] showed that the root-mean-square (RMS) difference in SST retrievals is 0.76 K, and the RMS difference in wind speed retrievals is 0.92 m/s with a bias of 0.57 m/s.

5. Moderate Resolution Imaging Spectroradiometer (MODIS)/Aqua 10 km aerosol L2 subset along CloudSat v2 (MAC04S1). This is the subset of the wide-swath MODIS/Aqua data within 100 km across the CloudSat track with 10 km horizontal resolution at nadir (http://disc.gsfc.nasa.gov/). A number of validations of MODIS \( \tau \) product have been done since MODIS/Terra was launched. Over ocean, the uncertainties in MODIS \( \tau \) are shown to be about ±(0.03 + 0.05) comparing to Aerosol Robotic Network observations [Remer et al., 2005].

6. Aqua CERES (Clouds and the Earth’s Radiant Energy System) single scanner footprint (SSF) products. The SSF product contains 1 h of instantaneous CERES data for a single-scanner instrument [Geier et al., 2001]. The SSF combines instantaneous CERES data with scene information from a higher-resolution imager such as Visible/Infrared Scanner on Tropical Rainfall Measuring Mission or MODIS. Scene identification and cloud properties are defined at the higher imager resolution, and these data are averaged over the larger CERES footprint. For each CERES footprint, the SSF contains the number of cloud layers and for each layer, the cloud amount, height, temperature, pressure, optical depth, emissivity, ice and liquid water path, and water particle size. The SSF also contains the CERES-filtered radiances. The shortwave (SW) and longwave radiances at spacecraft altitude are converted to top-of-atmosphere fluxes based on the imager-defined scene.

2.2. Cloud Identification and Data Collocation

In this study, only cloud-free CALIPSO backscatter data are used to retrieve aerosol information. CloudSat cloud mask and CALIPSO backscatter data were collocated and combined to detect clouds including optically thin clouds with cloud top higher than 8 km. CloudSat cloud mask together with attenuated lidar scattering ratio from CALIPSO provide more accurate cloud masks [Wang et al., 2008; Adhikari et al., 2010].

Then, related data sets were collocated into AMSR-E 25 km footprint, and cloud-free CALIPSO backscatter data are then averaged. Only ocean data within 50°N and 50°S and 200 km away from continents were used in the following analyses.

2.3. Aerosol Layer Identification

A new threshold algorithm is developed to identify the aerosol layer using the collocated cloud-free CALIPSO backscatter data. The threshold \( \beta_{\text{thr}} \) is chosen as [Vaughan et al., 2005]

\[
\beta_{\text{thr}}(z) = \beta_{m}(z) + 2 \text{MBV}
\]

Here \( z \) is the height; \( \beta_{m} \) is the molecular backscattering coefficients, estimated with temperature and pressure profiles from the ECMWF-AUX product; and MBV represents the measured backscatter variation, estimated as the standard deviation of measured attenuated backscatter coefficients from 30 to 40 km.

Considering the poor signal-to-noise ratio in 532 nm channels and weak molecular attenuation in 1064 nm channel, 532 nm (\( \beta_{532} \)) and 1064 nm (\( \beta_{1064} \)) attenuated backscatters were combined to determine the aerosol layer. For each aerosol layer detected by the threshold method, the aerosol layer should exist in both channels. The aerosol layer identification scheme is outlined as follows:
1. Compute the volume molecular backscattering coefficients \( \beta_m(z) \), two-way transmittances \( T_m^2(z) \), and MBV at 532 nm and 1064 nm, and then molecular attenuations were corrected for the signals of both wavelengths [Vaughan et al., 2005], as

\[
\beta(z) = \beta_{\text{obs}}(z)/T_m^2(z) 
\]

Here \( \beta_{\text{obs}}(z) \) is the measured signal, \( \beta(z) \) is the corrected signal, \( T_m^2(z) = e^{-}\int_0^z S_m \beta_m(z') dz' } \), and \( S_m = 8\pi/3 \) is the molecular lidar ratio.

2. Develop aerosol masks for each channel. To reduce aerosol attenuation incurred within and below the aerosol layers, the estimated aerosol backscattering coefficients \( \beta_e \) was used to identify layers. The backscattering coefficient \( \beta_e \) was computed with the forward iteration method [Young and Vaughan, 2009] by assuming layer top at 8 km and aerosol lidar ratio \( S_a \) of 25 (532 nm) and 40 (1064 nm).

For retrievals at certain level, \( \beta_e \) will be set to zero when

\[
\beta_e < \beta_m(z) + 2 \text{MBV}/T_m^2(z) 
\]

Here \( T_m^2(z) = e^{-}\int_0^z S_m \beta_m(z') dz' } \) is the estimated transmittances of aerosols with \( \beta_e \). Then, aerosol mask at each channel will set to be 1 where \( \beta_e > 0 \).

3. Refine the aerosol mask by combining aerosol masks at 532 nm and 1064 nm. First, for each wavelength, the aerosol layer with range bins less than 3 is removed. Then we combine aerosol mask profiles in two channels and set the new aerosol mask to be 1 if masks in both channels equal to 1 at a certain height. Finally, a three-range bins moving smooth is applied to \( \beta_{532} \) profiles in order to get more accurate aerosol layer top. Then and the aerosol layer top is extended to the highest points, where \( \beta_{532} \) is larger than \( \beta_{532} \) given in equation (1), and color ratio (1064 nm/532 nm) is larger than 0.06.

The derived aerosol layer depth is a good proxy for the \( \Delta H \). Luo et al. [2014] evaluated the threshold method with the ground-based lidar and radiosonde measurements. The results showed that lidar derived \( \Delta H \) over ocean agrees well with \( \Delta H \) derived from sounding profiles. The bias and RMS difference of lidar-derived \( \Delta H \) is \(-0.12 \pm 0.24 \) km relative to the sounding observations. CALIPSO-derived \( \Delta H \) was further evaluated with marine stratiform cloud top height over the global ocean with bias and RMS difference of \(-0.08 \pm 0.37 \) km.

### 2.4. Pure Marine Aerosol Identification

Several criteria were used to identify the clean marine aerosol cases. First, it should be a single layer with layer top within 0.3 km to 3 km and layer bottom below 0.1 km. Second, a threshold of volume depolarization ratio larger than 0.06 were used to identify and exclude dust aerosols [Liu et al., 2008]. Third, a threshold of integrated attenuated backscatter at 532 nm smaller than 0.01 sr\(^{-1}\) was applied to the remaining profiles to avoid possible clouds or continental aerosol cases [Kiliyanpilkikil and Meskhidze, 2011]. With these criteria, 934,981 pure marine aerosol cases were identified for analyses.

### 2.5. The \( \tau \) Retrieval and Lidar Ratio Selection

The \( \tau \) at 532 nm were retrieved by the forward iteration method. The lidar ratio \( (S_o) \) for marine aerosol is selected by comparing with MODIS 550 nm \( \tau \) (\( \tau_M \)) products. \( S_o \) values for clean marine aerosols were reported to be around 20–30 [Vaughan et al., 2009]. The \( S_o \) value of 20 for clean marine aerosols is used in CALIPSO level 2 data processing. However, CALIOP marine \( \tau \) at 532 nm is biased low as compared to other data sets, which indicate that \( S_o \) of 20 may be too small [Kiliyanpilkikil and Meskhidze, 2011; Oo and Holz, 2011; Sayer et al., 2012]. Therefore, a 1 year (2007) marine \( \tau \) was retrieved using different \( S_o \) ranging from 20 to 30 with an interval of 1. Then the retrieved \( \tau \) with different \( S_o \) was collocated into MAC04S1 footprint. Because MODIS can only observe the volume aerosol optical depth, identified sea-salt-type and single-boundary aerosol layer cases are used to constrain the lidar ratio. Figure 1a shows the mean differences between CALIPSO and MODIS retrievals over oceans, which clearly indicates that CALIPSO retrieved \( \tau \) with the selected \( S_o \) of 25 (\( \tau_C \)) produces the smallest mean difference. Figure 1b shows the distribution of \( \tau_C \) with \( \tau_M \). The \( \tau_C \) agrees well with \( \tau_M \) within the whole data range. The global averaged distributions of \( \tau_C \) are similar to that of \( \tau_M \) (not shown here), with the geographical differences within \( \pm 0.05 \) over the global ocean.
3. Results

The marine $\tau$ is influenced by a wide range of factors. $U_{10}$ controls the emissions of sea salt. Humidity affects the size and the optical properties of sea-salt particles. The sea-salt aerosol is mainly concentrated in the boundary layer, and thus, its vertical distribution is strongly influenced by boundary layer processes. Therefore, the potential controlling factors of $\tau$ at 532 nm considered in this study are mainly $U_{10}$, SST, $\Delta H$, LTS, and relative humidity (RH).

3.1. Surface Wind Speed

As shown in Figure 2, $\tau$s are most sensitive to the changes in $U_{10}$ (Figure 2a). In Figure 3, the regressive analysis shows linear relationship for $U_{10}$ within 4 m/s and 15 m/s, which gives

$$\tau_U = 0.005 + 0.0106 \times U_{10}, \text{ when } U_{10} > 4 \text{ m/s}$$

Here $\tau_U$ denotes the $\tau$-wind relationships at 532 nm only considering $U_{10}$; the unit of $U_{10}$ is m/s.

The slope of 0.0106 is larger than most ground-base measurement results, which are within the range of 0.002 to 0.007, but is quite close to MODIS-derived results [Lehahn et al., 2010; O’Dowd et al., 2010; Grandey et al., 2011]. Lehahn et al. [2010] suggested that the larger slopes based on MODIS observations may be

Figure 2. The distribution of marine $\tau$ as a function of (a) $U_{10}$, (b) $\Delta H$, (c) SST, and (d) LTS.
resulted from the possible overestimation in aerosol optical depth due to whitecaps under high-wind conditions. However, there is no such problem in CALIPSO retrievals. There are several limitations in using ground-based data to study the $\tau$-wind relationships, for example, spatial and temporal coverage, spatial separations between $\tau$ observed from coastal surface stations and $U_{10}$ from models or satellites, uncertainties in the interpolation of $U_{10}$ from model simulations, and difficulties in identifying pure marine aerosols near the land. Furthermore, some ground-base studies used daily-averaged values of $\tau$ and $U_{10}$. Therefore, these limitations may result in weaker $\tau$-wind dependencies and could introduce large uncertainties when applied them to remote oceans.

When $U_{10} \leq 4$ m/s, the wind-driven aerosol production process is very weak. Meanwhile, uncertainties in retrieved $U_{10}$ become relatively larger when $U_{10}$ is small. Therefore, the relatively large uncertainty in $U_{10}$ introduces relatively a large uncertainty in the $\tau$-wind relationships. The regressive analysis shows a weak linear dependency with a slope of 0.004, which reads as

$$
\tau = 0.0355 + 0.004 \times U_{10}, \text{ when } U_{10} \leq 4 \text{ m/s}
$$

The comparisons of different $\tau$-wind relationships from this study and the other studies are presented in Figure 3. It is clear that there are large differences among these parameterizations (Figure 3a), which may be resulted from different study regions, time periods, or data sources. To examine these possible causes, $\tau$-wind relationships over different previously studied regions were examined with our data set. As shown in Figure 3b, there are no evident differences among the $\tau$-wind relationships over the different regions. The differences among different years are small when further examining the subset of data by year. Thus, the differences presented in Figure 3a mainly result from different data sources. The consistent mean relationships over different regions also indicate that a general parameterization can be applied to the global ocean. Thus, in the rest analysis, regional differences will not be considered. However, Figure 2a shows large $\tau$ variation ranges at a given $U_{10}$. Therefore, it is critical to find out other processes responsible for the variations.

3.2. Other Factors
As shown in Figure 2b, $\tau$ has stronger dependence on $\Delta H$ than on SST (Figure 2c) or LTS (Figure 2d). To further explore the possible impacts of $\Delta H$, SST, and LTS, Figure 4 shows the $\tau$-wind relationships under different ranges of SST, LTS, and $\Delta H$. Figure 4a clearly shows strong dependencies on $\Delta H$ increases when $\Delta H$ is deeper. Consistent with Figure 2, Figures 4b and 4c only show weak dependences of $\tau$ on SST and LTS. Therefore, $\Delta H$ is the main external factor explaining the wide variation range of $\tau$-wind relationships in Figure 2a.

The dependence on $\Delta H$ indicates that the $u(2)$ boundary layer processes are important in controlling marine $\tau$. The evolution of boundary layer is promoted by mechanical turbulence driven by wind shear and thermodynamic...
turbulence driven by surface heating and is depressed by upper temperature inversion [Stull and Eloranta, 1984; Boers et al., 1984; Melfi et al., 1985; Boers and Eloranta, 1986]. Vogelezang and Holtslag [1996] suggested that the boundary layer depth could be determined by the Richardson number (Ri), which is defined as

$$\text{Ri}(z) = \frac{g z (\theta(z) - \theta(s))}{\theta(s) \left[ (u(z) - u(s))^2 + (v(z) - v(s))^2 + b u^* \right]^2}$$

where $b = 100$, $g$ is the gravity acceleration $U_{10}$ (9.8 m/s$^2$), $\theta(z)$ and $\theta(s)$ are the potential temperatures at height of $z$ and surface, $u(z)$ and $u(s)$ are the $u$ winds at height of $z$ and surface, $v(z)$ and $v(s)$ are the $v$ winds at height of $z$ and surface, and $u^*$ is the friction velocity. The boundary layer top is assigned to the height, where the Ri exceeds the critical value of 0.25.

Here $U_{10}$, SST, and LTS are used to represent the terms of wind shear, surface heating, and upper temperature inversion in equation (6) indirectly, and their relationships with CALIPSO-observed $\Delta H$ were shown in Figure 5. $\Delta H$ increases with increasing in $U_{10}$ or decreasing in normalized stability ($g^*\text{LTS/SST}$). When normalized stability is larger, it plays a minor role. The reason for this may be that $U_{10}$ can only represent for the wind shear production near surface, not for the whole boundary layer. This means that $\Delta H$ is controlled by the cooperations of multiple-boundary layer processes and thus is not strongly dependent on any single factor.

Under a given $U_{10}$, which means the same production rate of sea-salt particles, increasing of $\Delta H$ tends to dilute the near surface sea-salt aerosols by redistributing them into a larger volume [Glantz et al., 2009]. However, the enhancing of $\tau$ in our results suggests a positive contribution of more effective turbulent transportation and mixing when $\Delta H$ is deepening. MBL is capped with different magnitudes of temperature inversions near the MBL tops, which inhabit aerosol injection into the free troposphere unless convections penetrating into the

![Figure 4](image-url)  
**Figure 4.** $\tau$ as a function of $U_{10}$ under different bins of (a) $\Delta H$, (b) SST, and (c) LTS.

![Figure 5](image-url)  
**Figure 5.** (a) Relationship of $\Delta H$ with $U_{10}$, SST, and LTS. (b) Relationship of mean $\Delta H$ as a function of normalized stability ($g^*\text{LTS/SST}$) under different $U_{10}$. 

free troposphere. Deeper boundary layer indicates relatively stronger shallow convections and vertical mixing, corresponding with relatively larger-size particles in the MBL, which could counteract the deposition of sea salt and result in enhancing of \( \tau \).

Figure 4a also shows that \( \tau \) is more strongly dependent on \( \Delta H \) than on \( U_{10} \) when \( U_{10} \) is smaller than 4 m/s. The \( \tau \) has poor linear dependence on \( U_{10} \) because the wind-driven sea spray process is weak under the low-wind conditions. The impacts of SST and LTS were examined but could not account for the large variation of \( \tau \) under this condition. Therefore, \( \Delta H \) acts as the dominated factor controlling \( \tau \) variations under the low-wind conditions.

SST and LTS can affect \( \tau \) by affecting MBL processes. Generally, higher SST or lower LTS corresponds to larger \( \tau \). Higher SST may be associated with relatively strong surface heating, which leads to more surface water vapor evaporation and stronger convections. Lower LTS means weaker restriction on the development of convective cells and promotes the development of a deep MBL. As shown in Figure 5b, these situations may be associated with higher \( \Delta H \) and thus with higher \( \tau \) at the given \( U_{10} \). However, boundary layer process is also controlled by lots of other factors. As could be seen in Figure 5, \( \Delta H \) is not strongly dependent on any single factors. In our results, \( \Delta H \) shows a stronger influence on \( \tau \) than SST and LTS. Besides of altering boundary layer process, SST could control the sea-salt productions and thus influence the behavior of \( \tau \) [Mårtensson et al., 2003; Jaeglé et al., 2011]. However, Figure 6b shows no consistent relationship between the SST and \( \tau \), similar to the findings of Sayer et al. [2012]. The influence of SST on sea-salt production is weaker than that of \( U_{10} \) [Mårtensson et al., 2003], while \( \tau \) is the result from complex MBL processes, which include not only production but also other processes such as removal and turbulent transportation. Furthermore, increasing of SST tends to decrease the number of small particles and increase concentrations of large ones [Mårtensson et al., 2003]. The large sea-salt particles are easier to be activated as cloud condensation nucleus and removed than smaller ones. Therefore, the influences of SST on \( \tau \) through controlling source are not significant in our data sets. However, the influences of SST and LTS could be partially accounted by \( \Delta H \).
3.3. Humidity Effect

The humidity effect is also important to the MBL $\tau$, as reported by Glantz et al. [2009]. Sea-salt aerosol is hygroscopic and can grow rapidly to larger size at high relative humidity, which increases their scattering coefficients [Jaeglé et al., 2011]. Based on calculations from a simple aerosol model, Glantz et al. [2009] estimated that up to 50% of the enhanced $\tau$ with increasing wind speed seems to be due to the hygroscopic growth of marine aerosols and the remaining part due to increase in sea-salt particle mass concentrations (without hygroscopic growth). However, several other observational studies indicated that the humidity effects on MBL $\tau$ are not so straightforward [Smirnov and Shifrin, 1989; Smirnov et al., 1995; Sayer et al., 2012]. Their results showed that there is no correlation between the RH and $\tau$ as a whole, while an anticorrelation and a positive correlation of $\tau$ and relative humidity could be observed for relative humidity <75% and >75%, respectively. However, these anticorrelations and positive correlations are very weak [Smirnov and Shifrin, 1989; Smirnov et al., 1995]. Sayer et al. [2012] suggested that there is a potential influence of transported dust or cloud contamination resulting poorly sampled cases for lowest and highest humidity in their results. If excluding these poorly sampled cases, no trend could be observed between the RH and $\tau$ over a RH range between 60% and 80% with sufficient sampling. Similarly, no trend between the column water vapor and $\tau$ could be observed in their results.

The influence of humidity in our results was found to be minor too. Several humidity data sets, including the column water vapor from AMSR-E and relative humidity at the surface from Atmospheric Infrared Sounder level 2 and ECMWF-AUX products, were used to examine the humidity impact in different subsets of $U_{10}$ and $\Delta H$. Results indicate that humidity only has very weak influence on $\tau$-wind relationships similar as conclusions from Sayer et al. [2012]. Many possible reasons attribute to the results. First, data sets used in this study only included cloud-free data, which typically have a small range of relative humidity variations and thus have little changes in size distribution parameters. However, large uncertainties in the available humidity products over the oceans make the analysis challenge. Second, large sea-salt particles are quickly removed from the MBL, which may counteract the influence of humidity. Third, due to the high occurrence of the decoupling of MBL, the impacts of high near-surface humidity mainly limit within the lower shallow mixing layer [Bretherton and Wyant, 1997; Wood and Bretherton, 2004; Jones et al., 2011; P. Caldwell, et al., How Often is the Stratocumulus-Topped Boundary Layer Well-mixed? An observational perspective, submitted to Geophysical Research Letters, 2012]. Therefore, the near-surface humidity has weak impact on $\tau$, which is determined by the total sea-salt loading within the whole MBL. Last, the available water vapor in MBL is related to other meteorology parameters [Glantz et al., 2009]. The humidity influence may be included in the other boundary layer processes and parameters. Several studies suggested that the increasing of turbulent exchange in the MBL tends to increase the $\tau$ and decrease the RH [Smirnov and Shifrin, 1989; Smirnov et al., 1995; Glantz et al., 2009; Sayer et al., 2012]. Similar effects on RH are observed in our data sets. The wet cases are usually associated with lower mixing layer height, and drier cases are usually associated with higher mixing layer height, which will redistribute the aerosol loading and overwhelm the influences of RH.

3.4. New Parameterization and Evaluation

As presented above, not only wind-driven sea spray process but also vertical transportation and mixing driven by turbulence in the boundary layer is important in determining marine $\tau$. Based on these, new two-parameter relationships of MBL $\tau$ are developed in terms of $U_{10}$ and $\Delta H$, as shown in Figure 6. Global ocean data is sorted into different ranges of $\Delta H$, and two-parameter regression is performed as follows:

$$\tau_{UH} = a + b \times U_{10} + c \times U_{10}^2$$

(7)

Here $\tau_{UH}$ denotes the $\tau$-wind relationships at 532 nm considering $U_{10}$ and $\Delta H$; the $a$, $b$, and $c$ are the functions of $\Delta H$. Regressions give

$$a = 0.0159 + 0.0132 \times \Delta H + 0.00477 \times \Delta H^2 + 0.000168 \times \Delta H^3$$

(8)

$$b = -0.00516 + 0.0155 \times \Delta H - 0.00616 \times \Delta H^2 + 0.0006 \times \Delta H^3$$

(9)

$$c = 0.00035 - 0.000242 \times \Delta H + 0.000154 \times \Delta H^2 - 0.0000183 \times \Delta H^3$$

(10)

The new parameterization was applied to the collocated global ocean data (as detailed in section 2), and its performance was evaluated with CALIPSO and MODIS observations and compared with other single-parameter
parameterizations in Figure 7. Comparing to CALIPSO observations, Figures 7a and 7b clearly shows that the new parameterization provides smaller mean bias (0.0027) than other parameterizations (with absolute values within 0.02–0.04), except for $\tau_U$ (with absolute value of 0.0033). The new parameterization also provides a smaller RMS difference (0.035) than the other parameterizations (0.04–0.05). Comparing to MODIS observations, seen in Figures 7c and 7d, the new parameterization slightly underestimates $\tau$ (with mean bias of -0.01). The reason of underestimation is that MODIS observes the total column $\tau$. However, the new parameterization could still provide better RMS difference (0.029) than the others (0.03–0.05). The most of all, the new parameterization has the best performance within the full region of $\Delta H$, while single-parameter parameterizations can only predict the $\tau$ well at certain range of $\Delta H$, as shown in Figures 7b and 7d. The new parameterization also increases the correlation between predictions and observations to 0.65 for CALIPSO observations and to 0.66 for MODIS observations, while the others are smaller than 0.5 and 0.6, respectively.

4. Influence of $\tau$-Wind Parameterizations on Aerosol Radiative Forcing

With the data collocation process described in section 2, CALIPSO backscatter data and other data sets were collocated into Aqua CERES SSF products (~25 km). Then, the TOA SW radiations under clear-sky pure marine aerosol conditions were calculated using the Fu-Liou radiative transfer model [Fu and Liou, 1992, 1993]. This model is a delta four-stream radiative transfer scheme with 15 spectral bands from 0.175 to 4.0 $\mu$m in SW and 12 long-wave spectral bands between 2850 and 0 cm$^{-1}$. Due to different footprints of CALIPSO and CERES, the cloud-free condition is selected when the cloud fractions in both data sets are equal to 0. Ocean surface albedos were computed according to Jin et al. [2004], using $\nu_{110}$ from AMSR-E, monthly ocean chlorophyll concentration from MODIS (Aqua), and retrieved CALIPSO 532 nm $\tau$ and solar cosine zenith from CERES. The meteorology parameters are from the ECMWF-AUX products. Fu-Liou model requires $\tau$ at 550 nm and aerosol type as inputs to calculate aerosol optical properties. The calculations selected the aerosol type 1 (maritime aerosol with eight sets of relative humidity dependent properties [d’Almeida et al., 1991]). Aerosol type is then
used to determine the spectral normalized extinction, scattering, and absorption properties of the aerosol by the Mie theory, and the spectral dependence of the \( \tau \) [Fu-Liou Online Code Users Manual]. The \( \tau \) at 550 nm is provided by CALIPSO observations and different parameterizations. Only single aerosol layer cases were used to exclude the influences of elevated aerosol layer.

First, measured TOA SW radiations were used to evaluate Fu-Liou model calculations using CALIPSO retrieved \( \tau \) (see Figure 8a). Comparing to Aqua CERES observations, the mean bias of simulated SW radiation is \(-0.7 \text{ W/m}^2\). Ninety percent of the simulations have the difference smaller than 10 W/m\(^2\) and 87% of points with relative error smaller than 10%. These results indicate that Fu-Liou model can provide reliable estimations of TOA SW radiation. The difference between calculations and observations may be resulted from several reasons. The uncertainties in humidity from ECMWF-AUX could introduce uncertainties in aerosol optical properties calculations in the Fu-Liou model. Second, the surface albedo was calculated based on the parameters from satellite retrievals. However, there are uncertainties in these observations. Furthermore, CERES uses MODIS cloud mask, which has several issues in identify broken or optically thin clouds by using passive measurements [Ackerman et al., 2008; Trepte et al., 2010]. Although cloud information from both CALIPSO and CERES were combined to make sure the cloud fractions is 0, however, CERES has a much large footprint than the CALIPSO narrow path (100 m wide). Therefore, identified cloud-free CERES footprint is still likely contaminated by clouds difficult for MODIS to identify.

Then, with the same configurations of the Fu-Liou model, the retrieved \( \tau \) was replaced with parameterized \( \tau \) from different \( \tau \)-wind parameterizations, including \( \tau_U \), \( \tau_B \), and the other six parameterizations shown in Figure 3a. Then the SW fluxes calculated using parameterized \( \tau \) were compared to those calculated using measured \( \tau_C \) and observed by Aqua CERES, so as to evaluate the impact of different \( \tau \)-wind parameterizations on aerosol SW radiative forcing estimations. Figure 8b shows their biases and RMS total errors comparing to the SW fluxes using retrieved \( \tau \). As shown in Figure 8b, the new two-parameter parameterization has the best performance (0.4 ± 2 W/m\(^2\)). The \( \tau_U \) parameterization derived by us has the second best performance (0.6 ± 2.5 W/m\(^2\)). The other parameterizations give poorer performance (with the absolute values of the mean biases larger than 1.6 W/m\(^2\) and RMS total errors larger than 2.5 W/m\(^2\)). The SW fluxes using parameterized \( \tau \) were further compared with the Aqua CERES observations, as shown in Figure 8c. Because of the uncertainties due to different footprints and cloud identifications, etc., the biases and RMS errors of all parameterizations are increased comparing to the results in Figure 8b. However, the new two-parameter parameterization still has the best performance (−0.6 ± 4 W/m\(^2\)). The \( \tau_U \) parameterization has the second best performance (−0.7 ± 4 W/m\(^2\)). The other parameterizations give poorer performance (with the absolute values of the mean biases > 1 W/m\(^2\) and RMS total errors around 5 W/m\(^2\)). Therefore, the new two-parameter parameterization not only gives the best estimation of \( \tau \) but also gives the best estimation of SW radiative forcing.

5. Conclusions

Sea spray aerosol is one of the largest natural contributors to the global aerosol loading and thus plays an important role in the global radiative budget through both direct and indirect effects. Improving our knowledge
about marine $\tau$ is very important for climate forcing assessments and accurate modeling of the climate system. This paper aims at better understanding the connection of marine $\tau$ and external factors with A-train satellite measurements over the global ocean during June 2006 to December 2010.

First, $\tau$-wind relationships were reexamined to reconcile the existing differences among published $\tau$-wind parameterizations. The new global data set is used to study the $\tau$-wind relationships over different geographical regions, and no evident regional difference was found. This indicates that the different data sources used in the past is the main reason for the differences in published $\tau$-wind relationships. The consistent mean relationships over different regions also indicate that a general relationship may be applied to the global ocean.

Second, more potential factors controlling $\tau$ variations were further examined. Results showed that other than wind-driven sea spray process, vertical transportation and mixing driven by the turbulence in marine boundary layer are important in determining marine $\tau$. Especially, when the wind-driven aerosol production process ($U_{10} \leq 4 \text{ m/s}$) is very weak, the marine $\tau$ was found to be strongly dependent on boundary layer height. A new two-parameter (wind and boundary layer height) parameterization of $\tau$ was derived based on the new global observations. Evaluations with independent observations showed that the new parameterization greatly improves $\tau$ predictions over the global ocean. The bias and RMS of predicted $\tau$ by the new parameterization is $-0.002$ and $0.035$ relative to the observations, which is smaller than the other parameterizations. The new parameterization also improves the radiative forcing estimation. The Fu-Liou transfer model with $\tau$ from different $\tau$-wind parameterizations were used to calculate TOA shortwave fluxes, which are further compared with CERES observations and calculations with observed $\tau$. The new parameterization gave the best estimation of aerosol shortwave radiative forcing among different parameterizations.

As a continuation of this study, the influence of other processes, such as hygroscopic growth, cloud-drizzle-aerosol interactions, and entrainment mixing, on the behaviors of marine $\tau$ will be further explored in the near future.

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