3D aerosol climatology over East Asia derived from CALIOP observations

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HIGHLIGHTS

• A 3D aerosol climatology is generated from the CALIOP data over the East Asia.
• Spatio-temporal variation of aerosol optical properties has regional features.
• Errors on climate modeling for the current aerosol climatology are evident.

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ABSTRACT

The seasonal mean extinction coefficient profile (ECP), single scattering albedo (SSA), and scattering phase function (SPF) derived from the CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) version 3 Level 2 5-km aerosol profile product (2011–2014) were compiled into a three-dimensional (3D) aerosol climatology for East Asia. The SSA and SPF were calculated as the weighted averages of the scattering properties of the CALIOP aerosol subtypes. The weights were set to the occurrence frequencies of the subtypes. The single scattering properties of each subtype were extrapolated from the volume-based size distribution and complex refractive indexes based on Mie calculations. For the high-loading episodes (aerosol optical depth \( \geq 0.6 \)), the exponential ECP structures were most frequently observed over the farmland and desert areas, along with the uplifted ECP structures over the marine and coastal areas. Besides the desert areas, high-loading episodes also occurred over areas with frequent agricultural and industry activities. Unlike the conventional half-3D aerosol climatology (vertically constant SSA and SPF), this newly generated climatology specified SSA and SPF in the full-3D space (full-3D aerosol climatology). Errors on the shortwave radiative heating rate (SW RHR) due to the half-3D aerosol climatology approximation were quantified. The SW RHR errors were around ±1 K/day, implying that the half-3D aerosol climatology should be used with caution in climate modeling. This study is among the first to generate a full-3D aerosol climatology from the CALIOP data. This full-3D aerosol climatology is potentially useful for aerosol remote sensing and climate modeling.

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1. Introduction

Aerosols are an important modulator of weather and climate. As important cloud condensation nuclei, water-soluble aerosol particles play an important role in cloud formation and precipitation processes. Menon et al. (2008) demonstrated that precipitation decreased by a factor of four when biofuel and transport-based emissions were doubled. Influences of aerosol particles on climate are usually classified into direct and indirect effects (Pöschl, 2005). Direct effects are caused by the absorption and scattering of aerosol particles (Hansen et al., 2005; Rosenfeld et al., 2014). The indirect effect represents the effects of aerosol-cloud interactions. On the one hand, clouds formed with pollutant aerosol particles are accompanied by a cooling tendency (Ackerman et al., 2004). On the other hand, small ice particles condensed on aerosol particles in the deep convective clouds could be uplifted to the anvil, which favors the absorption of thermal radiation and leads to a warming...
Knowledge of aerosol optical properties, including the extinction coefficient profile (ECP), aerosol optical depth (AOD), single scattering albedo (SSA), and scattering phase function (SPF), is crucial to quantify the direct effects of aerosol on climate (Tiwari et al., 2015). In this study, the aerosol climatology denotes the spatio-temporal statistics of aerosol optical properties. In current research, the aerosol climatology is usually derived from the ground-based measurements by sun-photometers (Holben et al., 2001; Kambezidis and Kaskaoutis, 2008; Xia et al., 2016). The ground-based measurements are of high quality, providing meaningful insights into the spatio-temporal variation of aerosol loading and particle sizes. However, they are spatially sparse. Three main approaches have been adopted to prescribe the aerosol climatology worldwide: (i) Measurements made by satellite passive sensors may be used, sometimes combined with ground-based data (Kaskaoutis et al., 2007; Stefan et al., 2013). In this case, aerosol climatology is in two-dimensional (2D) space. Namely, AOD, SSA, and SPF are provided in the horizontal direction and usually at ground level. In this case, SSA and SPF are assumed to be constant vertically. (ii) A numerical model may be adopted, sometimes in combination with ground-based observations or data from satellite passive sensors. The Global Aerosol Climatology Project (GACP) program is a typical example of a 2D aerosol climatology (Mishchenko et al., 2002). The 3D aerosol climatology is exemplified by the EARLINET program (Schneider et al., 2000) and the Max-Planck-Institute Aerosol Climatology version 1 (MAC-v1) (Stefan et al., 2013), (iii) The Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) data have also been used to generate 3D aerosol climatology (Kosmopoulos et al., 2014; Tiwari et al., 2015). Previous research on the 3D aerosol climatology has provided significant insights into the spatio-temporal variation of ECP. However, the vertical distribution of SSA and SPF remains unknown, and the two parameters are usually assumed to be constant vertically. Strictly speaking, current aerosol climatology only specifies the extinction coefficient in the 3D space whereas specifies SSA and SPF specify in the 2D space. Therefore, it is described as a half-3D climatology in this study. For comparison, the 3D aerosol climatology in this study is described as a full-3D aerosol climatology. Accordingly, two main questions are raised: 1) How are the SSA and SPF distributed in the full-3D space? 2) What are the errors on the climate modeling, taking radiative heating rates (RHRs) as an example, due to the use of the half-3D aerosol climatology approximation?

Besides the extinction coefficient, CALIOP provides vertical scene classification product which classifies aerosols into six subtypes. In this study, the 3D distributions of SSA and SPF were derived from the CALIOP aerosol subtypes and their corresponding occurrence frequencies. For each subtype, SSA and SPF were extrapolated from the physical and optical characteristics provided by Omar et al. (2009). The 3D distribution of extinction coefficient, SSA, and SPF, was compiled into a 3D aerosol climatology over East Asia. Finally, errors on the RHRs due to the half-3D aerosol climatology approximation were quantified. The remainder of this paper is organized as follows. Section 2 presents the data and methods for the compilation of the full-3D aerosol climatology. Section 3 elaborates the experimental design and the methodology to calculate the RHRs. Section 4 presents the full-3D aerosol climatology. Section 5 addresses errors on the RHRs due to the half-3D aerosol climatology approximation. Section 6 discusses the uncertainties of the RHR calculation. Section 7 summarizes the general conclusions.

2. CALIOP data and processing method

The CALIOP instrument is onboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) mission, a member of the A-Train constellation of satellites observing aerosol optical properties worldwide (Yang et al., 2012). The CALIOP version 3 Level 2 5-km aerosol profile product from 2011 to 2014 was used in this study. The extinction coefficient is retrieved from the attenuated backscatter by an iterative process termed the Hybrid Extinction Retrieval Algorithm (HERA) (Young and Vaughan, 2009). The accuracy of the HERA depends on the scene classification procedure which determines the lidar ratio, an essential input parameter of the HERA. The aerosol subtype is identified by the selective iterated boundary locator and the scene classification algorithm (Winker et al., 2009). Aerosol subtypes identified by the CALIOP scene classification procedure include clean marine, dust, polluted continental, clean continental, polluted dust and smoke. The extinction coefficients and scene classification data are gridded at a vertical resolution of 30 m below 8.2 km and 60 m between 8.2 km and 20.2 km (Winker et al., 2009).

The aerosol climatology was generated over East Asia (20°N–50°N, 70°E–140°E). East Asia was divided into seven subregions, including the Taklimakan Basin (A), the Mongolian Plateau (B), northeastern China (C), the Tibetan Plateau (D), the North China Plain (E), the western Pacific (F), and the south of the middle and lower reaches of the Yangtze River (G) (Fig. 1). East Asia was divided into these smaller subregions for two reasons: (i) Each subregion represents a specific ecosystem, topography, population and industry density. Therefore, aerosol optical properties for a certain subregion are expected to be statistically representative. (ii) For a certain pixel along the CALIOP footprint, CALIOP data may be invalid as a result of cloud contamination and other factors, and the corresponding ECP may not be readily available. However, valid ECPs could be easily achieved for the domain average.

2.1. AOD and ECP

Within each subregion, an arbitrary CALIOP observation was selected as an independent aerosol episode. The per-layer mean extinction coefficient was calculated by averaging all the quality-assured extinction coefficients (the retrieval uncertainties ≤ 30%) of the selected layer on the altitude-orbit cross section of the CALIOP observation. Calculating the per-layer mean extinction coefficient throughout all layers yields a domain-averaged ECP. For each aerosol episode, AOD was calculated by integrating the domain-averaged ECP, where the integral start altitude from the surface was set to the domain-averaged surface elevation of the selected subregion. The surface elevation was obtained from the GTOPO30 digital elevation map (http://www.temis.nl/data/...
gtopo30.html) and was incorporated into the CALIOP data. The mean AODs were calculated by averaging AODs of all the valid aerosol episodes for different subregions, seasons, and AOD bins with the 532 nm AOD within 0–0.2, 0.2–0.4, 0.4–0.6, and 0.6–1.5. Aerosol episodes with extremely high aerosol loading (AOD at 532 nm > 1.5) were considered invalid to avoid large errors on the retrieved extinction coefficients. Similarly, the mean ECPs were calculated by averaging the domain-averaged ECPs of all the valid aerosol episodes for different subregions, seasons, and AOD bins. Because valid extinction coefficients above the tropopause height, which was also derived from the CALIOP data, were rather scarce, the extinction coefficients of the mean ECPs above the tropopause height were assumed to degrade with altitude with an assumed exponential function with a scale height of 5 km. The AOD in the stratosphere was directly derived from the stratospheric column optical depth provided by the CALIOP data. In the troposphere, gaps in the mean ECPs were filled using linear interpolation. With these methods, the mean ECPs and AODs were generated at both 532 nm and 1064 nm for different subregions, seasons, and AOD bins.

2.2. Occurrence frequencies of the CALIOP aerosol subtypes

For an arbitrary valid aerosol episode within one of the seven subregions, the per-layer occurrence frequency ($\eta_i$) of a certain aerosol subtype was calculated using Equation (1):

$$\eta_i = N_i / N \quad i = 1, \ldots, 6,$$

where $N_i$ denotes the number of pixels identified as the $i$th aerosol subtype and $N$ denotes the total number of the pixels identified as aerosols. Calculating $\eta_i$ throughout all layers yields a vertical structure of occurrence frequency. For a certain aerosol subtype, the mean vertical structure of the occurrence frequency was calculated by averaging the vertical structures of the occurrence frequency of all valid aerosol episodes for different subregions, seasons, and AOD bins. With this method, the mean vertical structures of the occurrence frequency were calculated for all six CALIOP aerosol subtypes. The per-layer occurrence frequencies of the six aerosol subtypes were scaled to guarantee that their sum was equal to one.

2.3. Vertical structures of SSA and SPF

For each of the six aerosol subtypes, SSA and the SPF were extrapolated from the volume-based size distribution and refractive indices provided by Omar et al. (2009). Details are provided in the appendix. For a certain subregion, season, and AOD bin, the per-layer mean SSA was calculated as the weighted average of the SSAs of the six aerosol subtypes, with the weights being the mean occurrence frequencies of the selected altitude. Performing such calculation throughout all layers yielded the mean SSA profiles for different subregions, seasons and AOD bins. Similarly, the per-layer mean SPF was calculated using Equation (2):

$$P(z, \cos \Theta) = \sum_{i=1}^{6} \eta_i(z) P_i(z, \cos \Theta),$$

where $P(z, \cos \Theta)$ denotes the mean SPF at altitude $z$ and scattering angle $\Theta$, $\eta_i(z)$ denotes the mean occurrence frequency of the $i$th aerosol subtype at altitude $z$, and $P_i(z, \cos \Theta)$ denotes the SPF of the $i$th aerosol subtype. According to Section 2.2, $\eta_i(z)$ conforms to Equation (3):

$$\sum_{i=1}^{6} \eta_i(z) = 1$$

For precision, SPF was specified by the Legendre function expansion coefficients as shown in Equation (4) rather than the asymmetry parameter. The latter is only a robust approximation of the original SPF by the Henyey–Greenstein scheme (Boucher, 1998).

$$P(z, \cos \Theta) = \sum_{k=1}^{N} \omega_k(z) P_k(\cos \Theta)$$

where $p_k(\cos \Theta)$ denotes the $k$th moment of the Legendre function, the corresponding Legendre function expansion coefficient is denoted by $\omega_k(z)$; and $N$ denotes the number of phase function moments and was set to 201 in this study. This conversion was performed by the weighted least squares method (Zhou et al., 2016). By simple deduction,

$$\omega_k(z) = \sum_{i=1}^{6} \eta_i(z) \omega_{i,k}$$

where $\omega_{i,k}$ denotes the $k$th moment of the Legendre function expansion coefficient of $P_i(\cos \Theta)$. Therefore, the vertical structures of SPF for different subregions, seasons, and AOD bins were specified by the weighted averages of the Legendre function expansion coefficients of the six CALIOP aerosol subtypes.

The SSA and the Legendre function expansion coefficients above the tropopause height were relaxed to the values at the tropopause height. Gaps of the SSA and Legendre function expansion coefficients in the troposphere were filled using linear interpolation.

3. Experimental design and the RHR calculation

To answer the two questions posed in Section 1, this study covered two main topics: (i) A full-3D aerosol climatology was generated by the data and methods introduced in Section 2. (ii) Errors on the RHRs calculation due to the half-3D aerosol climatology approximation were quantified. For the half-3D aerosol climatology, SSA and the Legendre function expansion coefficients were determined using the steps listed below:

1. The Top-Of-Atmosphere (TOA) radiances at 532 nm and 1064 nm were calculated using the SBDART radiative model (Ricchiazzi et al., 1998). The input parameters of the SBDART model include ECP, SSA profile, the Legendre function expansion coefficients as a function of altitude, and other parameters including temperature profile, water vapor density profile, ozone density profile, and surface albedo.

2. The two most predominant aerosol subtypes were determined by selecting the two maximum values of $\zeta$ formulated by Equation (6):

$$\zeta(l) = \sum_{j=1}^{M} \eta(j, l) k_{ext}(j).$$

where $\eta(j, l)$ denotes the occurrence frequency of the $j$th aerosol subtype at the $l$th layer, $M$ denotes the number of the model atmosphere layers and is set to 33 in this study, and $k_{ext}(j)$ denotes the extinction coefficient at the $j$th layer. As indicated, $\zeta$ not only accounts for the occurrence frequency of each aerosol subtype, but also the per-layer contribution of extinction coefficient to the TOA radiance.

3. Using a similar method to Step 1, the TOA radiances at 532 nm and 1064 nm were calculated for the vertically constant SSA
SSA and the Legendre function expansion coefficients \((\hat{c}_i, \hat{\sigma}_i)\). For a certain wavelength, SSA and \(\hat{c}_i\) were calculated by Equations (7) and (8).

\[
SSA_i = \chi_i SSA_1 + (1 - \chi_i) SSA_2 \tag{7}
\]

\[
\hat{\sigma}_i = \chi_i \omega_{1,k} + (1 - \chi_i) \omega_{2,k} \tag{8}
\]

where the subscripts 1 and 2 denote the two most predominant aerosol subtypes determined by Step 2, and \(\chi_i\) denotes the mixing ratio of the two most predominant aerosol subtypes and varies from 0.0 to 1.0 with an increment of 0.05.

4. SSA and the Legendre function expansion coefficients of the half-3D aerosol climatology were determined as the values corresponding to the minimum residual \(\varepsilon\) computed by Equation (9):

\[
\varepsilon(i) = \sqrt{\frac{1}{2} \left[ \left( \frac{I_{\text{half-3D}}(i)}{C_0} - \frac{I_{\text{full-3D}}(i)}{C_0} \right)^2 + \left( \frac{D_{\text{half-3D}}(i)}{C_0} - \frac{D_{\text{full-3D}}(i)}{C_0} \right)^2 \right]},
\]

where \(I_{\text{half-3D}}\) and \(D_{\text{half-3D}}\) denote the TOA radiances of the full-3D aerosol climatology at 532 nm and 1064 nm, respectively, calculated by Step 1; and \(I_{\text{full-3D}}\) and \(D_{\text{full-3D}}\) denote the TOA radiances for the SSA and \(\hat{\sigma}_i\) with the mixing ratio of \(\chi_i\), and were calculated by Step 3. To constrain the accuracy, only the vertically constant SSA and Legendre function expansion coefficients corresponding to the minimum residual smaller than the threshold value of 0.001 W/m\(^2\)/sr were considered valid optical properties of the half-3D aerosol climatology.

The RHRs were calculated for the full-3D aerosol climatology and the corresponding half-3D aerosol climatology determined by Steps 1–4 described above. During the calculation, AODs were set to the mean AODs for different subregions, seasons and AOD bins. The model atmosphere was gridded at a 1-km vertical resolution below 25 km, far coarser than the vertical resolution of the CALIOP data. For a certain SBDART model layer, aerosol optical properties were calculated by averaging the optical properties of several CALIOP layers inside the region which centered on the model layer and extended 0.5 km up and down from this point. By performing the calculation throughout all the SBDART model layers, the CALIPO-derived aerosol climatology was mapped onto the SBDART model layers. The aerosol climatology was specified at 532 nm and 1064 nm. At other wavelengths, the single scattering properties were calculated by the interpolation or extrapolation in logarithmic space (Ricchiazzi et al., 1998). To minimize the errors of RHR calculation due to the interpolation and extrapolation, only the shortwave (SW) (0.2–4.0 \(\mu\)m) RHRs are discussed in this study.

The atmospheric states, including the water vapor density, the temperature, and the pressure as a function of altitude, were derived from the CALIOP data below 29 km and were set to those of the US-62 standard atmosphere above 29 km. In addition, the ozone density was also set to that of the US-62 standard atmosphere. For precision, the atmospheric states are not presented here. The surface albedo was derived from the U.S. geological survey land use data (Grossman-Clarke et al., 2005) and is presented in Fig. 2.

4. 3D aerosol climatology

4.1. AOD and ECP

The mean AODs and ECPs for different subregions, seasons, and AOD bins are presented in Figs. 3 and 4, respectively. Because the general ECP patterns were similar at 532 nm and 1064 nm, only ECPs at 532 nm are presented here. The 4-year and seasonal mean AODs for the seven subregions are summarized in Tables 1 and 2.

As indicated, the spatio-temporal variation of ECP and AOD has regional features. For Subregion A, Sun et al. (2015) identified high-loading episodes (AOD \(> 0.6\)) with sand-dust events. As presented by the embedded pie charts in Fig. 4, the occurrence frequency of sand-dust weather shows a spring and summer maximum and autumn and winter minimum, which conforms well to previous research (Xia et al., 2004; Wang et al., 2008; Sun et al., 2015). For Subregions B, C, D, E, and G, the 4-year mean AODs at 532 nm were in good agreement with the values at 500 nm provided by Wang et al. (2008). The high-loading episodes were most frequently observed in Subregions E and G throughout the year, presumably associated with high densities of population and agricultural and industry activities (Cheng et al., 2006). Subregion F had a maximum occurrence frequency of high-loading episodes in spring and winter. A potential cause is the transportation of sand-dust particles in spring (Huang et al., 2008) and the anthropogenic pollutants in winter. In real cases, aerosol loading is affected by many factors including the land use category, local topography, densities of population, etc (Che et al., 2011). The seasonal variation of these factors was different for the seven subregions and even within a certain subregion (Wang et al., 2008). Therefore, no generalized seasonal variation of AOD was attempted over East Asia in this study.

For the high-loading episodes, the seasonal mean ECPs in Subregions A, C, and E share similar characteristics, i.e., the extinction coefficient reaches to its maximum at the near-ground level and degrades with altitude in most cases. Such an ECP structure conforms well to the exponential ECP promoted by Qiu et al. (2005). For Subregion A, the extinction coefficient peaked at the near-ground level, implying that sand-dust particles are originated and entrained in the lower troposphere. For Subregion E, the extinction coefficient peaked at about 7 km in summer, to which a potential contributor is the fugitive materials from the Taklimakan Basin and the Mongolian Plateau (Huang et al., 2008). For Subregions B, D, F,
and G, ECPs show an uplifted structure, i.e., the extinction coefficient peaked at certain altitude above the ground, especially in summer and autumn. The uplifted ECP structure may be caused by the uplifting airflows as a result of turbulence or the topography (Ge et al., 2014; Jia et al., 2015). For the moderate-loading episodes \((0.2 < \text{AOD} < 0.6)\), the ECP structure was rather complicated. Two observations by the Semi-Arid Climate and Environment Observatory of Lanzhou University (located in Subregion B) revealed that the seasonal variation of ECP was affected by human activities (coal combustion in winter) and weather conditions (Cao et al., 2013). For the low-loading episodes \(\text{AOD} \leq 0.2\), aerosols are usually distributed at higher altitude than those of the moderate- and high-loading episodes, which was also validated by the higher scale height (Fig. 5) calculated as the threshold value of height below which AOD equals 63% of the whole-layer AOD (Turner et al., 2001; Sun et al., 2015). A recent study by Petäjä et al. (2016) demonstrates...
that the increase of planetary boundary layer (PBL) pollutants concentration decreases the PBL in urban regions, favoring the further concentration of pollutants below the PBL. Consequently, the scale heights are decreased. The PBL polluted episodes are usually associated with large AOD (Kaskaoutis et al., 2011). Such findings partly explain the lower scale heights observed for the moderate- and high-loading episodes than the low-loading episodes in some cases (see Fig. 5).

4.2. Occurrence frequencies of the CALIOP aerosol subtypes

The mean vertical structures of occurrence frequencies of aerosol subtypes for different subregions, seasons, and AOD bins, the mean vertical structures of SSA and the Legendre function expansion coefficients were calculated as shown in Section 2.3. Because the general SSA patterns were similar at 532 nm and 1064 nm, only the results for SSA at 532 nm are presented (Fig. 7).

Based on the mean vertical structures of occurrence frequencies of aerosol subtypes for different subregions, seasons, and AOD bins, the mean vertical structures of SSA and the Legendre function expansion coefficients were calculated as shown in Section 2.3. Because the general SSA patterns were similar at 532 nm and 1064 nm, only the results for SSA at 532 nm are presented (Fig. 7). As indicated, the vertical variation of SSA was non-negligible. In addition, the vertical variation of the Legendre function expansion coefficients was non-negligible. In order to be concise, the results for the Legendre function expansion coefficients are not presented.

The mean ECPs, vertical structures of SSA and Legendre function expansion coefficients for different subregions, seasons, and AOD bins were compiled into a full-3D aerosol climatology, which was used in the following SW RHR calculation.

5. Effects on the SW RHR calculation

The SW RHRs for the full-3D climatology is presented in Fig. 8. Because the valid extinction coefficients above the tropopause height were scarce, only the SW RHRs in the troposphere are presented. As indicated, the radiative effects due to aerosols tended to warm the troposphere, with the SW RHR ranging from 0 to 6 K/day in most cases. The vertical distribution of the SW RHR was closely related to the corresponding ECP structure. For example, the SW RHR reached its maximum near the underlying surface, and degraded with height below the middle tropopause height for the high-loading aerosol episode in autumn in northeastern China (C3 in Fig. 8), where the mean ECP was fitted to an exponentially decreasing function (C3 in Fig. 4). However, the SW RHR peaked at around 5 km for the large aerosol episode in autumn in the western Pacific (F3 in Fig. 8), which corresponds well with the uplifted ECP pattern (F3 in Fig. 4). This correspondence was mainly caused by the aerosol scattering effects and is in good agreement with the findings of Huang et al. (2009), who performed RHR calculations over the Taklimakan Basin.
The SW RHRs were also calculated for the half-3D aerosol climatology. Fig. 9 presents SW RHR errors due to the half-3D aerosol climatology approximation. The errors were calculated by subtracting the SW RHRs for the full-3D climatology from those for...
the half-3D climatology. During the subtraction process, the effects of water vapor and other gases were eliminated. As indicated, the SW RHRs deviated from the true values by the half-3D aerosol climatology approximation, with the differences ranging from −1.0 K/day to 1.0 K/day in most cases. For the high-loading episode in the spring of the Taklimakan Basin (A1 in Fig. 9) and the moderate-loading episode in the spring of the Mongolian Plateau (C1 in Fig. 9), the SW RHR error due to the half-3D approximation even reached −2.5 K/day and −3.0 K/day, respectively. In addition, an intercomparison between Figs. 7 and 9 reveals that the SW RHR error was mainly determined by the corresponding SSA profile. This phenomenon is exemplified in Fig. 10, which corresponds to the high-loading episode in the summer of the Taklimakan Basin (A2 in Fig. 7). Fig. 10 indicates that SSA was underestimated below 6 km for the half-3D aerosol climatology, and vice versa. This SSA pattern was in good agreement with the underestimation (≥6 km) and overestimation (≤6 km) of the SW RHR shown in Fig. 9 (A2); namely, SW RHR was negatively related to SSA.

Tables 3 and 4 summarize the vertical averaged SW RHR errors and the corresponding standard deviations, respectively. As indicated, two characteristics are apparent: (i) The vertical averaged SW RHR errors due to the half-3D approximation were within ±0.38±0.26 K/day, which implies that the SW RHR of the half-3D climatology approximates well to that of the full-3D climatology on the vertical average. (ii) The standard deviations of the SW RHR errors in the vertical direction had a maximum value of 1.07 K/day. Therefore, larger SW RHR errors due to the half-3D approximation could be expected at a certain altitude. Theoretically, large SW RHR errors hamper the accurate estimation of static stability of the atmosphere determined by the temperature profile. Johnson and Ciesielski (2000) revealed that a 1.5 K/day variation of RHR during the Madden–Julian oscillation over the Western Pacific influences the tropical Walker and Hadley circulations. In addition, Ma and Kuang (2011) indicated that the RHR anomaly (±0.3 K/day) is involved in the modulation of Kelvin waves. Therefore, the half-3D aerosol approximation should be used with caution in climate modeling.

6. Uncertainties of the SW RHR calculation

The uncertainties of the SW RHR calculation have their roots in the uncertainties of the surface albedo, ECP, vertical structures of SSA and SPF (the Legendre function expansion coefficients), and the inherent uncertainties of the SBDART radiative model. Here we only focus on the influences of AOD, SSA, and SPF.

6.1. Uncertainties of AOD

The results in Section 5 are based on the mean AODs (\(\overline{\text{AOD}}\)) for different subregions, seasons, and AOD bins. The uncertainties (\(\text{dAOD}\)) due to the statistical method were calculated as the standard deviations of the AODs of all the valid aerosol episodes for different subregions, seasons, and AOD bins. Fig. 11 presents the uncertainties of AOD due to the statistical method at 532 nm and 1064 nm.

The perturbed AOD (\(AOD'\)) was formulated by Equation (10) with a constraint that \(AOD'\) is larger than zero:

\[
AOD' = AOD + (-1)^{\text{rand}} \text{dAOD}
\]

where \(\text{rand}\) is set to 1 (negative perturbation) or 2 (positive perturbation) randomly. The SW RHR profiles were recalculated for the perturbed AOD. We calculated the SW RHR uncertainties by subtracting the SW RHRs for the full-3D aerosol climatology from those for the perturbed full-3D aerosol climatology (only AOD was perturbed in this case). The SW RHR uncertainties as a function of altitude are presented in Fig. 12. Because the SW RHRs are mainly caused by the scattering effects of aerosol particles, the SW RHR was positively related to the aerosol loadings. Therefore, the results
shown in Fig. 12 are reasonable. We further calculated the SW RHR errors due to the half-3D aerosol climatology corresponding to the perturbed full-3D aerosol climatology. The results indicate that the vertical averaged SW RHR errors due to the half-3D approximation ranged from –0.49 K/day to 0.44 K/day, with the standard deviations in the vertical direction ranging from 0.01 K/day to 1.44 K/day.

6.2. Uncertainties of SSA

The per-layer SSA uncertainty was calculated as the standard deviation of the SSAs of all the valid aerosol episodes for a specific AOD bin, season, and subregion. Fig. 13 presents the SSA uncertainties due to the statistical method as a function of altitude. The perturbed SSA was formulated by Equation (11) with a constraint that SSA is smaller than 1:

\[
\text{SSA}'(z) = \text{SSA}(z) + (-1)^{nd}(\delta \text{SSA}(z))
\]

where \(\text{SSA}(z)\) denotes the domain-averaged SSA at altitude \(z\), \(\delta \text{SSA}(z)\) is the corresponding uncertainty, \(\text{SSA}'(z)\) denotes the perturbed SSA. The SW RHR profiles were recalculated for the perturbed full-3D aerosol climatology (only SSA was perturbed in this case). The results indicate that the vertical averaged SW RHR

<table>
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<tr>
<th>AOD bin</th>
<th>Subregion</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
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<td>0.06</td>
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<td>–0.08</td>
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uncertainties ranged from $0.06$ K/day to $0.25$ K/day, implying that the SW RHR is sensitive to the small perturbation of SSA ($\leq 0.06$). For the perturbed full-3D aerosol climatology, the vertical averaged SW RHR errors due to the half-3D approximation ranged from $0.31$ K/day to $0.39$ K/day, with the standard deviations in the vertical direction ranging from $0.06$ K/day to $0.94$ K/day.

### 6.3. Uncertainties of SPF

According to Equation (5), the uncertainties of SPF were denoted by the uncertainties of the Legendre function expansion coefficients, which were calculated as the standard deviations of the Legendre function expansion coefficients of all the valid aerosol episodes for a specific Legendre function moment, layer, AOD bin, season, and subregion. In order to be concise, the SPF uncertainties are not presented here, but their influences on the SW RHR calculation were calculated. The SW RHRs were calculated for the perturbed full-3D aerosol climatology (only the Legendre function expansion coefficients were perturbed in this case). The perturbed Legendre function expansion coefficients were calculated by the method similar to those formulated by Equations (10) and (11). The results indicated that the vertical averaged SW RHR uncertainties due to the SPF uncertainties ranged from $-0.06$ K/day to $0.04$ K/day, implying that the SW RHR is insensitive to the SPF perturbation. For the perturbed full-3D aerosol climatology, the vertical averaged SW RHR errors due to the half-3D approximation were within $-0.31$ K/day and $0.28$ K/day, with the standard deviations in the vertical direction ranging from $0.03$ K/day to $1.01$ K/day.

In this section, the uncertainties of the optical properties only account for the uncertainties due to statistical methods. For AOD, uncertainties were also caused by the retrieval uncertainties of the CALIOP extinction coefficients ($\leq 30\%$). For SSA and SPF, the uncertainties were also caused by the aerosol classification procedure. Moreover, the CALIOP aerosol cluster process captures the microphysical and optical properties on the global scale, leaving uncertainties of the calculated SSA and SPF over East Asia (Appendix). Based on the results presented above, these uncertainties are unlikely to affect the general findings in Section 5. Namely, the SW RHR errors due to the half-3D aerosol climatology were non-negligible, and the half-3D aerosol climatology should be used with caution in climate modeling.

### 7. Conclusions

In this study, the mean ECPs, vertical distributions of SSA and the SPF for different subregions, seasons, and AOD bins were derived from the CALIOP version 3 Level 2 5-km aerosol profile product from 2011 to 2014 over East Asia. The SSA and SPF of the full-3D aerosol climatology were calculated as the weighted averages of the scattering properties of the six CALIOP aerosol subtypes. The weights were set to the occurrence frequencies of the aerosol subtypes. The SSA and SPF for each aerosol subtype were derived from the volume-based size distribution and refractive indexes provided by Omar et al. (2009). The CALIOP-derived aerosol optical properties were compiled into a full-3D aerosol climatology over

<table>
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<th>AOD bin</th>
<th>Subregion</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>F</th>
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<td>0.28</td>
<td>0.20</td>
<td>0.36</td>
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East Asia, which was divided into seven subregions including the Taklimakan Basin (A), the Mongolian Plateau (B), northeastern China (C), the Tibetan Plateau (D), the North China Plain (E), the western Pacific (F), and the south of the middle and lower reaches of the Yangtze River (G). To explore the influences of the half-3D aerosol climatology approximation (SSA and SPF were assumed

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**Fig. 12.** Uncertainties of the SW RHRs due to the uncertainties of AOD. The positive AOD perturbation corresponds to the profiles on the right and vice versa (cf. Fig. 4).

**Fig. 13.** Uncertainties of SSA due to the statistical method (cf. Fig. 4).
vertically constant) on the climate modeling, errors on the SW RHRs were quantified. The three main conclusions are summarized below.

1. For the high-loading episodes (AOD ≥ 0.6), the seasonal mean ECPs show exponential structures in most cases over the farmland and desert areas of Northern China (Subregions A, C, and E), and frequently show uplifted structures in the marine and coastal areas (Subregions F and G) and the Mongolian Plateau (Subregion B). In the other cases not mentioned above, ECP structures were rather complicated, presumably originating from the complex meteorological conditions and local topographies (Huang et al., 2009; Liu et al., 2012). Besides Subregions A and B, which represent the main sand-dust aerosol sources, high-loading episodes occurred frequently in Subregions C, E, and G, which are the areas with high densities of agricultural and industrial activities. This implies that anthropogenic pollutants are an important contributor to the high turbidity of the atmosphere.

2. Dust, polluted dust, and smoke aerosols were consistently observed over East Asia. Over the marine area (Subregion F) and coastal areas (Subregions C, E, and G), clean marine aerosols also occurred but were mainly restricted to the lower troposphere. Based on the vertical distribution of the occurrence frequencies of the six aerosol subtypes, we calculated the mean vertical structures of SSA and SPF (Legendre function expansion coefficients) for the seven subregions. The spatio-temporal variation of SSA and SPF was clear and showed distinct regional features.

3. The CALIPSO-derived aerosol optical properties were compiled into a full-3D aerosol climatology. The SW RHRs were calculated for the full-3D aerosol climatology. The results indicate that aerosols tend to warm the troposphere, with the vertical averaged SW RHRs ranging from 1.33 K/day and 3.89 K/day. In addition, the SW RHRs for the half-3D aerosol climatology were computed. The SW RHR errors due to the half-3D aerosol climatology approximation, calculated by subtracting the SW RHRs for the full-3D climatology from those for the half-3D climatology, were around ±1 K/day. Such errors may hamper the accurate modeling of the general circulation of atmosphere (Johoson and Giesielski, 2000) and atmospheric waves (Ma and Kuang, 2011). Therefore, the half-3D aerosol climatology approximation should be used with caution in current climate modeling.

This study is among the first to generate the full-3D aerosol climatology from the CALIOP data. The seven subregions have specific land surfaces and topographies. Therefore, the full-3D aerosol climatology is expected to be statistically representative, and is potentially useful for aerosol remote sensing and climate modeling. However, as a result of the uncertainties of the CALIOP extinction coefficient and the CALIOP aerosol model derived single scattering properties, there were uncertainties in the derived full-3D aerosol climatology. These problems require further investigation in future model evaluation studies.

Acknowledgements

This research was supported by National Natural Science Foundation of China (Grant No. 41575020). The CALIOP data were obtained from the NASA Langley Atmospheric Science Data Center (ASDC). The phase functions of the six CALIOP aerosol subtypes are available from the authors upon request (xuejin.sun@outlook.com, yongbo.zhou@yahoo.com).

Appendix

Number-based particle size distribution

Omar et al. (2009) provided the parameters determining the volume-based particle size distributions (PSDs) of the six CALIOP aerosol subtypes, and indicated that the volume-based PSDs fitted well to the bi-lognormal distributions. For the lognormal PSD, the volume-based PSD (the third moment of the PSD) could be converted to the number-based PSD (zeroth moment of the PSD). The number-based PSD was formulated by Equation (A1).

\[ n(r) = \frac{1}{\sqrt{2\pi \ln \mu_0}} \left( \ln r - \ln \mu_0 \right) \exp \left( -\frac{\left( \ln r - \ln \mu_0 \right)^2}{2(\ln \mu_0)^2} \right). \] (A1)

where \( n(r) \) denotes the number distribution for radius \( r \). \( \ln \mu_0 \) and \( \ln \delta_0 \) denote the mean and standard deviation of the particle radius in the logarithmic space. According to Heintzenberg (1994), the standard deviation of the particle radius is the same for the zeroth and the third moment of size distribution, and the mean particle radius of the zeroth and the third moments of the PSD conform to Equation (A2):

\[ \mu_0 = \mu_3 \exp \left( -3 \ln \delta_0^2 \right) \] (A2)

where \( \mu_3 \) are the mean particle radius of the third moment of the PSD.

According to Omar et al. (2009), each CALIOP aerosol subtype is composed of fine and coarse modes. The volume-based mixing ratio is converted to the number-based mixing ratio by Equation (A3) (Heintzenberg, 1994):

\[ V = \frac{4}{3} \pi \left[ \frac{\mu_3 \exp \left( 3(\ln \delta_0^2) \right)}{2} \right]^3 \exp \left\{ -\frac{\left( \ln (\mu_3 \exp \left( 3(\ln \delta_0^2) \right)) - \ln \mu_0^2 \right)}{2(\ln \delta_0^2)^2} \right\}, \] (A3)

where \( V \) and \( N \) denote the total volume and total number of aerosols per unit volume air, respectively.

Based on Equations (A2) and (A3), the volume-based PSD was converted to the number-based PSD. Parameters determining the number-based PSDs are summarized in Table A1. The number-based PSDs are shown in Fig. A1.

<table>
<thead>
<tr>
<th>Table A1</th>
<th>Parameters determining the number-based PSDs of the six CALIOP aerosol subtypes.</th>
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<tr>
<td>( \delta_0 ) (( \mu ))</td>
<td>Coarse mode</td>
</tr>
<tr>
<td>Number-based mixing ratio</td>
<td>Fine mode</td>
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</table>
Mean volume single scattering properties

The mean volume single scattering properties, including the single scattering albedo (SSA), asymmetry parameter (ASY), and scattering phase function ($P(\Theta)$), were calculated by Equations (A4)–(A6).

$$\text{SSA} = \frac{\sigma_{\text{scattered}}}{\sigma_{\text{ext}}},$$  \hspace{1cm} \text{(A4)}$$

$$\text{ASY} = \frac{\int_0^\infty \left[ \beta_{\text{ext},C}(r) \pi r^2 n_C(r) \eta + \beta_{\text{ext},F}(r) \pi r^2 n_F(r)(1 - \eta) \right] \text{d}r}{\int_0^\infty \left[ \beta_{\text{scattered},C}(r) \pi r^2 n_C(r) \eta + \beta_{\text{scattered},F}(r) \pi r^2 n_F(r)(1 - \eta) \right] \text{d}r},$$ \hspace{1cm} \text{(A5)}$$

$$\text{P}(\Theta) = \frac{\int_0^\infty \left[ P_C(\Theta, r) \beta_{\text{scattered},C}(r) \pi r^2 n_C(r) \eta + P_F(\Theta, r) \beta_{\text{scattered},F}(r) \pi r^2 n_F(r)(1 - \eta) \right] \text{d}r}{\int_0^\infty \left[ \beta_{\text{scattered},C}(r) \pi r^2 n_C(r) \eta + \beta_{\text{scattered},F}(r) \pi r^2 n_F(r)(1 - \eta) \right] \text{d}r},$$ \hspace{1cm} \text{(A6)}$$

In Equation (A4), $\sigma_{\text{ext}}$ and $\sigma_{\text{scattered}}$ denote the mean volume extinction cross section and scattering cross section, which are calculated by Equations (A7) and (A8), respectively:

$$\sigma_{\text{ext}} = \frac{\int_0^\infty \left[ \beta_{\text{ext},C}(r) \pi r^2 n_C(r) \eta + \beta_{\text{ext},F}(r) \pi r^2 n_F(r)(1 - \eta) \right] \text{d}r}{\int_0^\infty \left[ n_C(r) \eta + n_F(r)(1 - \eta) \right] \text{d}r},$$ \hspace{1cm} \text{(A7)}$$

$$\sigma_{\text{scattered}} = \frac{\int_0^\infty \left[ \beta_{\text{scattered},C}(r) \pi r^2 n_C(r) \eta + \beta_{\text{scattered},F}(r) \pi r^2 n_F(r)(1 - \eta) \right] \text{d}r}{\int_0^\infty \left[ n_C(r) \eta + n_F(r)(1 - \eta) \right] \text{d}r},$$ \hspace{1cm} \text{(A8)}$$

where $\beta_{\text{ext},C}(r)$ and $\beta_{\text{scattered},C}(r)$ denote the extinction efficiency and scattering efficiency for the coarse aerosol mode, and $\eta$ denotes the coarse fraction by number. Similarly, $\beta_{\text{ext},F}(r)$ and $\beta_{\text{scattered},F}(r)$ denote the extinction efficiency and scattering efficiency for the fine aerosol mode. The fine fraction by number is denoted by $(1 - \eta)$, and $n_C(r)$ and $n_F(r)$ denote the number-size distribution of the coarse and fine aerosol modes.

In Equation (A5), $g_C(r)$ and $g_F(r)$ denote the asymmetry...
parameter of a spherical particle with a radius of \( r \) for the coarse and fine aerosol modes. The two parameters were formulated by Equations (A9) and (A10):

\[
\begin{align*}
g_C(r) &= \frac{\int_1^1 P_C(r, \cos \Theta) \cos \Theta d \cos \Theta}{\int_1^1 P_C(r, \cos \Theta) d \cos \Theta} \\
g_F(r) &= \frac{\int_1^1 P_F(r, \cos \Theta) \cos \Theta d \cos \Theta}{\int_1^1 P_F(r, \cos \Theta) d \cos \Theta}
\end{align*}
\]

(A9) (A10)

where \( P_C(r, \cos \Theta) \) and \( P_F(r, \cos \Theta) \) denote the scattering phase functions of a spherical particle with radius \( r \) for the coarse and fine aerosol modes, respectively, and \( \Theta \) denotes the scattering angle.

For each aerosol subtype, SSA, \( \text{ASY} \), and \( P(\Theta) \) were calculated in two main steps. (i) The extinction efficiency, the scattering efficiency, and the scattering phase function were simulated by the BHMIE code (Bohren and Huffman, 1983) (available at http://www.astro.princeton.edu/~draine/scattering.html) for a series of \( r \) spanning from 0.01 \( \mu \)m to 20.0 \( \mu \)m with an increment of 0.01 \( \mu \)m. (ii) SSA, \( \text{ASY} \), and \( P(\Theta) \) were calculated according to Equations (A4) – (A6). The single scattering properties calculated by this procedure are summarized and presented in Table A2, Table A3, and Fig. A2.

The CALIOP aerosol models developed by Omar et al. (2009) were constructed from global-scale measurements. However, aerosol single scattering properties exhibit distinct regional variation. Considering that the SW RHR is sensitive to SSA, we discuss the representativeness of the SSAs summarized by Table A2 over the East Asia. (a) The SSA at 532 nm of sand-dust aerosols ranged from 0.94 to 0.97 over the East Asia (Noh, 2014), and from 0.96 to 0.99 over the Saharan Desert (Petzold et al., 2011). Therefore, the 532 nm SSA of the East Asia dust was underestimated by 0.02 – 0.05 in this study, although the result corresponds well with that of Hamill and Lopez-Garibay (2011), who calculated SSA of the pure dust at 532 nm based on the CALIOP aerosol models by Omar et al. (2009). (b) The polluted continental cluster involves AERONET observations in the spring of 2001 – 2004 of Beijing (Cattrall et al., 2005). Therefore, the CALIOP aerosol model-derived SSA should be representative of the SSA in this region. Qiu et al. (2004) suggested the yearly mean SSA of 0.86 at 1 \( \mu \)m, which is close to the simulated SSA at 1.064 \( \mu \)m (0.88). Besides, Xia et al. (2016) indicated that SSA in Beijing varies with month, with the value spanning from 0.85 to 0.95 at 0.55 \( \mu \)m. Therefore, SSA of the polluted continental over Beijing was underestimated by around 0.01 – 0.11 at 532 nm and by around 0.02 at 1064 nm. The overestimation of SSA presumably originates from the increasing emissions of the absorbing aerosols in recent years. (c) Aerosols over Dunhuang (40.163°N, 94.802°E), Mt_WLG (36.28°N, 100.90°E), and NAM_CO (30.77°N, 90.96°E) are

| Table A2 |
| SSA of the six CALIOP aerosol subtypes. |
| Wavelength & Dust & Smoke & Clean continental & Polluted continental & Clean marine & Polluted dust |
| 532 nm & 0.9182 & 0.8335 & 0.9037 & 0.9570 & 0.9861 & 0.8502 |
| 1064 nm & 0.9080 & 0.7011 & 0.9539 & 0.8801 & 0.9942 & 0.7842 |

| Table A3 |
| Asymmetry parameters of the six CALIOP aerosol subtypes. |
| Wavelength & Dust & Smoke & Clean continental & Polluted continental & Clean marine & Polluted dust |
| 532 nm & 0.6916 & 0.6507 & 0.8344 & 0.6807 & 0.7869 & 0.6705 |
| 1064 nm & 0.6934 & 0.5949 & 0.7706 & 0.5577 & 0.7865 & 0.6725 |

Fig. A2. Normalized scattering phase functions of the six CALIOP aerosol subtypes.
potentially mixed with smoke/soot (Zheng et al., 2008; Yi et al., 2014), and are very similar to the CALIOP polluted dust. Zheng et al. (2008) demonstrated that the SSA in Dunhuang is within 0.87 and 0.93 at 532 nm and within 0.89 and 0.95 at 1064 nm. For Mt_WLG and NAM_CO, SSAs are very different between different seasons, with the values at 0.441 nm ranging from 0.4 to 1.0 (Yi et al., 2014). Therefore, the CALIOP aerosol model derived SSA is representative of the observed SSA in certain regions of the East Asia, but with large uncertainties. The large uncertainties of SSA presumably originate from the fact that the SSA of the polluted dust is sensitive to the chemical composition of the mixtures of black carbon, organic carbon, and water-soluble aerosols (Fan et al., 2014), which exhibit distinct spatio-temporal variation. (d) Aerosols over Gosan Island (33°N, 126°E), Ron Brown (35°N, 130°E), Twin Otter (33°N, 128°E) and C-130 (35°N, 125°E) are similar to the CALIOP clean marine aerosols, and SSA in these locations is estimated to be around 0.93–0.97 at 532 nm and 0.97–1.00 at 1064 nm (Bergstrom et al., 2004). Therefore, the clean marine SSA over East Asia was overestimated by about 0.01–0.06 at 532 nm while it corresponds well with the model simulated SSA at 1064 nm. Quantitative measurements of SSA of the clean continental and the smoke aerosols over East Asia are scarce. Using the same CALIOP aerosol models by Omar et al. (2009), Hamill and Lopez-Garrabch (2011) produced almost the same SSA as the CALIOP aerosol model-derived SSA at 532 nm for the smoke aerosol (0.83). The representativeness of the single scattering properties of the six CALIOP aerosol subtypes over East Asia requires further validations.

References


