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Comparison of key absorption and optical properties between pure and transported anthropogenic dust over East and Central Asia

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Abstract. Asian dust particulate is one of the primary aerosol constituents in the Earth-atmosphere system that exerts profound influences on environmental quality, human health, marine biogeochemical cycle and Earth's climate. To date, the absorptive capacity of dust aerosol generated from Asian desert region is still an open question. In this article, we compile columnar key absorption and optical properties of mineral dust over East and Central Asia areas by utilizing the multi-year quality assured datasets observed at 13 sites of the Aerosol Robotic Network (AERONET). We identify two types of Asian dust according to threshold criteria from previously published literature. (I) The particles with high aerosol optical depth at 440 nm ($AOD_{440}\geq0.4$) and low Ångström wavelength exponent at 440-870 nm ($\alpha<0.2$) are defined as Pure Dust (PDU) that decrease disturbance of other non-dust aerosols and keep high accuracy of pure Asian dust. (II) The particles with $AOD_{440}\geq0.4$ and $0.2<\alpha<0.6$ are designated as Transported Anthropogenic Dust (TDU), which are mainly dominated by dust aerosol and might mix with other anthropogenic aerosol types. Our results reveal that the primary components of high AOD days are predominant by dust over East and Central Asia regions even if their variations rely on different sources, distance from the source, emission mechanisms, and meteorological characteristics. The overall mean and standard deviation of single-scattering albedo, asymmetry factor, real part and imaginary part of complex refractive index at 550 nm for Asian PDU are 0.935±0.014, 0.742±0.008, 1.526±0.029, 0.00226±0.00056, respectively, while corresponding values are 0.921±0.021, 0.723±0.009, 1.521±0.025, and 0.00364±0.0014 for Asian TDU. Aerosol shortwave direct radiative effects at the top of the atmosphere (TOA), at the surface (SFC), and in the atmospheric layer (ATM) for Asian PDU ($\alpha<0.2$) and TDU ($0.2<\alpha<0.6$) computed in this study, are a factor of 2 smaller than the results of OPAC Mineral accumulated (Mineral acc.) and transported (Mineral tran.) modes. Therefore, we are convinced that our results hold promise of updating and improving accuracies of Asian dust characteristics in present-day remote sensing applications and regional or global climate models.

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1. Introduction

Airborne dust particle (also called mineral dust) is recognized as one of the most important aerosol species in the tropospheric atmosphere, which accounts for about 30% of the total aerosol loading and extinction aerosol optical depth on a global scale (Perlwitz et al., 2001; Kinne et al., 2006; Chin et al., 2009; Huang et al., 2014). High concentrations of dust aerosols hanging over desert source regions and invasive downstream areas would seriously exacerbate air quality, degrade visibility, affect transportation safety, and do adverse effects on public health during the prevalent seasons of dust storms (Chan et al., 2008; Morman and Plumlee, 2013; Wang et al., 2016). When mineral dusts are deposited onto the Earth's surface, they play a key role in biogeochemical cycles of terrestrial ecosystem or ocean (Okin et al., 2004; Jickells et al., 2005; Shao et al., 2011), as well as alter snow and ice albedo (Aoki et al., 2006; Huang et al., 2011; Wang et al., 2014). Last but not least, dust particles can modulate the Earth's energy budget and drive the climate change directly by scattering and absorption of solar/terrestrial radiation (Charlson et al., 1992; Wang et al., 2010b; Huang et al., 2014), and indirectly by acting as effective cloud condensation nuclei or ice nuclei, influencing the cloud microphysics and precipitation processes (Ramanathan et al., 2001; Rosenfeld et al., 2001; DeMott et al., 2003; Huang et al., 2005, 2006, 2010a; Wang et al., 2010c; Creamean et al., 2013). Numerous studies (Sokolik and Toon, 1999; Lafon et al., 2004, 2006) have confirmed that dust particle is one kind of light absorbing substances, and its mass absorption efficiencies at 325 nm (0.06~0.12 m^2/g) are about 6 times larger than at 660 nm (0.01~0.02 m^2/g), owing to the greater absorbing potential of iron oxides at short wavelengths (Alfaro et al., 2004). However, the way of iron oxides mixed with quartz or clay is complicated and strongly impacts the resulting absorption (Claquin et al., 1998, 1999; Sokolik and Toon, 1999). And these mineralogical studies indicate that a lack of consideration of these mixing mechanisms is a significant limitation of the previous dust absorption computations. Although the absorptive ability of dust is two orders of magnitude lower than for black carbon (Yang et al., 2009), the atmospheric mass loading of the

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former is the same magnitude larger than that of the latter, leading to the total absorption in solar spectrum comparable to black carbon. Chin et al. (2009) evaluated that dust may account for about 53% of global averaged aerosol absorption optical depth at 550 nm, which undoubtedly changes the aforementioned dust-cloud-precipitation interaction and exerts a significant effect on hydrological cycle of the Earth-atmosphere system.

East and Central Asia territories are the major source regions of dust aerosols on Earth, which produce a large amount of dust particles every year that become entrained into the upper atmosphere by cold fronts (Zhang et al., 1997; Huang et al., 2009, 2010a, 2014). They can travel over thousands of kilometers, even across the Pacific Ocean and reach the western coast of North America about one week with the prevailing westerly wind (Husar et al., 2001; Uno et al., 2009, 2011), and then modify the climate and environment over extensive area of Asia-Pacific rim. Thus far, there have been a great deal of fruitful field campaigns for exploring Asian dust (e.g., U.S.S.R.-U.S., ACE-Asia, ADEC, PACDEX, EAST-AIRC), however, most focus on intensive observation period (Golitsyn and Gillette, 1993; Huebert et al., 2003; Nakajima et al., 2003; Mikami et al., 2006; Huang et al., 2008a; Li et al., 2011) and lack of long-term and quantitative knowledge of dust optical, microphysical characteristics (especially absorption properties) and chemical compositions over these regions. Hence, the absorptive capacity of Asian dust aerosol is still an outstanding issue. The variations of dust optical features in model calculations are closely related to the uncertainties in particle size distribution and prescribing a value for complex refractive index. Whereas the key parameters of Asian dust aerosols in present-day climate models are still prescribed to the predetermined properties of Saharan mineral dust.

Wang et al. (2004) inferred the refractive index of pure minerals at Qira in Taklimakan Desert during April 12-14, 2002 via combination of theory calculation and composition analysis of aerosol samples, and showed that the value of imaginary part is 0.00411 at 500 nm, which is consistent with the Central Asian dust of 0.004±0.001 (Tadzhikistan Desert; Sokolik and Golitsyn, 1993). Uchiyama et al. (2005)

determined the single-scattering albedo (SSA) of Aeolian dust from sky radiometer and in situ measurements, and concluded that unpolluted Aeolian dust (source from Taklimakan Desert) has low absorption (with SSA_{500} of 0.93~0.97). Kim et al. (2004) analyzed multiyear sky radiation measurements over East Asian sites of Skyradiometer Network (Nakajima et al., 1996; Takamura et al., 2004) and showed the SSA_{500} of dust particles are around 0.9 in arid Dunhuang of northwest China and Mandalgovi Gobi desert in Mongolia. Bi et al. (2014) also reported the similar SSA_{550} (0.91~0.97) of dust aerosol at Dunhuang during spring of 2012. Xu et al. (2004) gained SSA_{530} of 0.95±0.05 in Yulin, China, from a Radiance Research nephelometer and a Particle Soot Absorption Photometer (PSAP) and suggested that both desert dust and local pollution sources contributed to the aerosol loading in Yulin during April 2001. Whereas Ge et al. (2010) examined dust aerosol optical properties at Zhangye (a semiarid area of northwest China) from multifilter rotating shadowband radiometer (MFRSR) during spring of 2008 and found that although there are low aerosol optical depth values (AOD_{670} ranging from 0.07~0.25), dust particles have strong absorption (with SSA_{500} of 0.75±0.02) due to mixing with local anthropogenic pollutants. This result is close to the New Delhi over India (0.74~0.84 for SSA_{500}; Pandithurai et al., 2008). Lafon et al. (2006) revealed that due to containing of less calcite and higher fraction of iron oxide-clay aggregates, mineral dusts in Niger (Banizoumbou, 13°31′N, 2°38′E) have much lower SSA in the visible wavelengths than that of Chinese (Ulan Buh, 39°26′N, 105°40′E) and Tunisian (Maouna, 33°01′N, 10°40′E) desert locations. Therefore, complete clarification of the climate-relevant impacts of Asian dust aerosols requires extensive and long-term measurements of the optical, microphysical and chemical properties, along with their spatial and temporal distributions.

In this paper, we investigate optical characteristics of Asian dust from multi-year AErosol RObotic NETwork (AERONET) measurements at 13 sites in and around arid or semi-arid regions of East and Central Asian desert sources. The key quantities include single-scattering albedo (SSA), asymmetry factor (ASY), real part (Re) and imaginary part (Ri) of complex refractive index, volume size distribution (dV/dlnr), which are needed for climate simulating and remote sensing applications. We mainly

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compare the vital absorption and optical properties between pure and transported anthropogenic dust over East and Central Asia. This article is arranged as follows. Section 2 introduces the site description and measurement. The identification method and detailed Asian dust optical features are described in Section 3. Discussion of spectral absorption behaviors of different dust aerosol types are given in Section 4 and followed by the Summary in Section 5.

2. Site Description and Measurement

2.1. Site Description

In this article, we select 13 AERONET sites located in arid or semi-arid Asian regions (see Fig. 1), which are recognized as the primarily active centre of dust storms. These drylands are very sensitive to climate change and human activities and would accelerate drought expansion by the end of twenty-first century (Huang et al., 2016). Eight sites over East Asian region are labeled with red colors, and five sites over Central Asian area are labeled with blue colors. The major Great deserts or Gobi deserts along with plateaus are marked with black font (e.g., Great Gobi desert in Mongolia, Taklimakan Desert, Thar Desert, Karakum Desert, Tibetan Plateau, Loess Plateau, and Iranian Plateau). In order to quantitatively explore detailed spectral absorptive characteristics of dust aerosols over East and Central Asia, we choose four East Asian sites (SACOL, Dalanzadgad, Beijing, and Yulin) and four Central Asian sites (Dushanbe, Karachi, Kandahar, and IASBS). They consist of: SACOL located over Loess Plateau of northwest China (Huang et al., 2008b; Guan et al., 2009; Huang et al., 2010b; Wang et al., 2010a), Dalanzadgad in the Great Gobi of southern Mongolia (Eck et al., 2005), Beijing in the downwind of Inner Mongolia (Xia et al., 2007), Yulin on the southwestern fringe of the Mu Us desert in northwest China (Xu et al., 2004; Che et al., 2009, 2015), Dushanbe in Tadzhikistan situated at the transport corridor of Central Asian desert dust (i.e. Karakum Desert; Golitsyn and Gillette, 1993), Karachi located in the southern margin of Thar Desert in Pakistan and about 20 km from the east coast of Arabian Sea (Alam et al., 2011), Kandahar in the arid area of southern Afghanistan, IASBS on the Iranian Plateau of northwest Iran.

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2.2. Sun Photometer Measurements

AERONET is an internationally federated global ground-based aerosol monitoring network utilizing Cimel sun photometer, which comprises more than 500 sites all over the world (Holben et al., 1998). The Cimel Electronique sun photometer (CE-318) takes measurements of sun direct irradiances at multiple discrete channels within the spectral range of 340-1640 nm, which can be calculated aerosol optical depth (AOD) and columnar water vapor content (WVC) in centimeter. Furthermore, the instrument can perform angular distribution of sky radiances at 440, 675, 870, and 1020 nm (nominal wavelengths), which can be simultaneously retrieved aerosol volume size distribution, complex refractive index, single-scattering albedo, and asymmetry factor under cloudless condition (Dubovik and King, 2000; Dubovik et al., 2002a, 2006). The total accuracy in AOD for a newly calibrated field instrument is about 0.010-0.021 (Eck et al., 1999). The retrieval errors of SSA, ASY, Ri, and Re are anticipated to be 0.03-0.05, 0.04, 30%-50%, and 0.025-0.04, respectively, relying on aerosol types and loading (Dubovik et al., 2000). It should be borne in mind that these uncertainties are dependent on AOD_{440} ≥0.4 and for solar zenith angle >50° (Level 2.0 product), and the retrieval errors will become much greater when AOD_{440} <0.4. The datasets of selected 13 AERONET sites in this study come from the Level2.0 product, which are pre- and post-field calibrated, automatically cloud screened, and quality-assured (Smirnov et al., 2000). In addition, a mixture of randomly oriented polydisperse spheroid particle shape assumption with a fixed aspect ratio distribution is applied to retrieve key optical properties of Asian dust (Dubovik, et al., 2002a, 2006).

3. Asian Dust Optical properties

A great amount of publications have verified that mineral dust aerosols are commonly predominant by large particles with coarse mode (radii>0.6 μm), which are the essential feature differentiating the dust from fine-mode dominated biomass burning and urban-industrial aerosols (Dubovik et al., 2002b; Eck et al., 2005; Bi et al., 2011, 2014; Kim et al., 2011). In other word, the values of Ångström exponent at

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440-870 nm (α) for dust aerosols usually range between -0.1 to 0.6. As pointed out by Smirnov et al. (2002) and Dubovik et al. (2002b), sea salt aerosol is also dominant by coarse mode and has small Ångström exponent (~0.3-0.7) but with low AOD_{440} (~0.15-0.2) compared to dust aerosol. Moreover, the selected desert locations in this study are mostly not affected by sea salt. By virtue of these differences, we can distinguish Asian dust aerosols from other fine-mode dominated non-dust particles. The criteria of two thresholds are put forward. (I) The particles with high aerosol optical depth at 440 nm ($AOD_{440} \geq 0.4$) and low Ångström wavelength exponent at 440-870 nm ($\alpha<0.2$) are defined as Pure Dust (PDU) that keep high accuracy of pure Asian dust and eliminate most fine mode aerosols. (II) The particles with $AOD_{440} \geq 0.4$ and $0.2<\alpha<0.6$ are designated as Transported Anthropogenic Dust (TDU), which are mainly dominated by dust and might mix with other anthropogenic aerosol types during transportation. The definition of anthropogenic dust in this study is different from earlier literatures (Tegen and Fung, 1995; Prospero et al., 2002; Huang et al., 2015), which define that anthropogenic dust is primarily produced by various human activities on disturbed soils (e.g., agricultural practices, industrial activity, transportation, desertification and deforestation). It is still a huge challenge to discriminate between natural and anthropogenic components of dust aerosols by using current technology, AERONET products or in-situ measurements. Recently, Ginoux et al. (2012) first estimated that anthropogenic sources globally account for 25% based on Moderate Resolution Imaging Spectroradiometer (MODIS) Deep Blue dust optical depth in conjunction with other land use data sets. Huang et al. (2015) proposed a new algorithm for distinguishing anthropogenic dust from natural dust by using Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) and planetary boundary layer (PBL) height retrievals along with MODIS land cover data set. They revealed that anthropogenic dust produced by human activities mainly comes from semi-arid and semi-humid regions and is generally mixed with other types of aerosols within the PBL that is more spherical than natural dust. Thereby, we assume that anthropogenic dust aerosol originated from Asian arid or semi-arid areas has got smaller size distribution (thus larger Ångström exponent) than that of pure natural dust.

Before insight into dust aerosol optical characteristics, we first analyze the occurrence frequency of Asian dust over the study region that significantly affects the intensity and distribution of mineral dust loading. Figure 2 depicts the total number days of each month for Pure Dust (α<0.2) and transported Anthropogenic Dust (0.2<α<0.6) at selected four East Asian sites and four Central Asian sites. The dust events at four East Asian sites primarily concentrate on springtime and corresponding peak days for PDU and TDU both appear in April. This is greatly attributed to the intrusion of dust particles during spring when dust storms are prevalent over these regions (Wang et al., 2008). For SACOL and Beijing sites, both the PDU and TDU days also occur in whole year except for autumn when is the rainy season, which is linked to long-range transport of dust particulates from desert source areas and locally anthropogenic dust (e.g., agricultural cultivation, overgrazing, desertification, industrial and constructed dust in urbanization). Shen et al. (2016) have demonstrated that urban fugitive dust generated by road transport and urban construction contributes to more than 70% of particulate matter ($PM_{2.5}$) in northern China. The dust episodes in Dushanbe of Tadzhikistan mostly happen from July to October, which are the peak seasons of dust storms (Golitsyn and Gillette, 1993). For Karachi site in Pakistan, the dust activities take place in spring and summer seasons. This is because the region is not affected by the summer monsoon, leaving the land surface sufficiently dry, and hence susceptible to wind erosion by strong winds and meso-scale thunderstorm events typical of this time of year (Alizadeh Choobari et al., 2014). In addition, the transport of summer dust plumes from the Arabian Peninsula can partially contribute dust particles to Karachi site. Note that the occurred months of PDU days are nearly different from TDU days at Dalanzadgad, Kandahar, and IASBS sites, suggesting that dust aerosols over these areas are rarely affected by anthropogenic pollutants. For Kandahar site in Afghanistan, the limited sampling days to some extent may affect the statistical results. Generally, the aforementioned occurrence frequency of dust storms over diverse sites are principally dependent on different climatic regime and synoptic pattern, for instance, geographical location,

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atmospheric circulation, wet season and dry season.

Table 1 summarizes the site information, sampling period, overall average optical properties at 550 nm (e.g., SSA, ASY, Re, Ri, and Ångström exponent at 440-870 nm) for Asian PDU (α<0.2), and total number of PDU and TDU (0.2<α<0.6) days. Note that dust optical feature at a common 550 nm wavelength is utilized here, which can be derived from logarithmic interpolation between 440 and 675 nm. It is worth pointing out that the absorption and optical properties of dust aerosols at two Dunhuang sites exhibit consistent features despite of different sampling periods, which indicate that the chemical composition of dust aerosol at Dunhuang area remains relatively stable.

The SSA or Ri of complex refractive index can characterize the absorptive intensity of dust aerosols, and determine the sign (cooling or heating, depending on the planetary albedo) of the radiative forcing (Hansen et al., 1997). Both two quantities are mainly relied on the ferric oxide content in mineral dust (Sokolik and Toon, 1999). Figure 3 illustrates the overall average spectral behavior of key optical properties for PDU (α<0.2) and TDU (0.2<α<0.6) at selected four East Asian sites. The SSA, ASY, Re and Ri of complex refractive index as a function of wavelength (440, 675, 870, and 1020 nm) are presented. For all cases, the spectral behaviors of aerosol optical parameters exhibit similar features, which can be representative of typical patterns of Asian dust. The SSA values systematically increase with wavelength at 440-675 nm and keep almost neutral or slight increase for the wavelengths greater than 675 nm, which is consistent with the previous results of dust aerosols (Dubovik et al., 2002b; Eck et al., 2005; Bi et al., 2011). In contrast, an opposite pattern is displayed by imaginary part of refractive index, namely, Ri values dramatically decrease from 440 nm to 675 nm, and preserve invariant from 675 nm to 1020 nm. These variations indicate that Asian dust aerosols have got much stronger absorptive ability at shorter wavelength. Alfaro et al. (2004) implied that the absorption capacity of soil dust increase linearly with iron oxide content, and estimated SSA at 325 nm (~0.80) is much lower than at 660 nm (~0.95). Sokolik and Toon (1999) revealed that ferric iron oxides (e.g., hematite and goethite) are often internally mixed with clay minerals and result in significant dust absorption in the UV/visible wavelengths. Hence, the spectral variations of SSA and Ri with wavelengths are attributable to the domination of coarse-mode dust particles that have larger light absorption in the blue spectral band as mentioned above. It is worth noting that spectral ASY values remarkably reduce from 440 nm to 675 nm, and are almost constant at 675-1020 nm range. This suggests that Asian dust aerosols have much stronger scattering at 440 nm than other longer visible wavebands, due to the contribution of coarse mode particles. By contrast, the spectral behavior of Re is not obvious for PDU and TDU at all sites, and the mean Re values at 440 nm vary between 1.50 and 1.56. Although there are 18 years continuous AERONET datasets at Dalanzadgad site, the effective days of PDU and TDU are only 8 and 6 days, respectively, almost appearing in springtime period. There are no identifiable differences for dust absorption properties between PDU and TDU cases for Dalanzadgad, which indicates again that the site is hardly influenced by anthropogenic pollutants. The spectral discrepancies of optical characteristics between PDU and TDU at other three sites show much more apparent than Dalanzadgad, which is ascribed to these regions are not only affected by dust aerosols, but also including local anthropogenic emissions, for instance, urban-industry, coal fuel combustion, biomass burning, mobile source emissions, and agricultural dust (Xu et al., 2004; Xia et al., 2007; Che et al., 2015; Bi et al., 2011; Wang et al., 2015).

Figure 4 is the same as Figure 3, but for selected four Central Asian sites. The wavelength dependencies of PDU and TDU cases at Central Asian sites are consonant with that of East Asian sites, despite of somewhat different variations of magnitude and amplitude. This is expected, because the East Asian desert sites are very close to the Central Asian desert locations and remain similar chemical compositions of dust aerosols (Wang et al., 2004). The spectral behaviors of dust optical properties for PDU at Kandahar and IASBS sites are nearly the same as TDU cases, which agrees well with the consistent variability of occurrence of dust storms. The wavelength dependency of dust characteristics for PDU at Dushanbe and Karachi presents large differences with TDU case, which is also likely due to the influence of local anthropogenic pollutions. Furthermore, the standard deviation of PDU is far less than that of TDU at all wavelengths, suggesting that the robustness of PDU recognition method.

Particle size distribution is another critical agent for deciding the optical and radiative properties of dust aerosol. Nakajima et al. (1996) and Dubovik and King (2000) uncovered that based on the spherical Mie theory, the retrieval errors of volume size distribution do not exceed 10% for intermediate particle size ($0.1 \le r \le 7$ μm) and may greatly increase to 35-100% at the edges of size range ($r<0.1$ μm or $r>7$ μm). As mentioned above, a polydisperse, randomly oriented spheroid method is utilized in this study, which is demonstrated to remove the artificially increased size distribution of fine particle mode with $AOD_{440} \ge 0.4$ and for solar zenith angle >50°. Additionally, the large errors at the edges do not significantly affect the derivation of the main features of the particle size distribution (concentration, median and effective radii, etc.), because typical dust aerosol size distributions have low values at the edges of retrieval size interval (Dubovik et al., 2002a). Figure 5 delineates the overall average columnar aerosol volume size distributions (dV/dlnr, 0.05 μm≤r≤15 μm) for Pure Dust ($\alpha<0.2$) and Transported Anthropogenic Dust ($0.2<\alpha<0.6$) at selected 13 AERONET sites. Corresponding AOD_{440} and effective radius of coarse mode (r_{coarse}) in μm are also shown. It is apparent that the dV/dlnr exhibits a typical bimodal structure and is dominant by coarse mode for PDU and TDU at all sites. The dV/dlnr peak of coarse mode particle varies dramatically and appears at a radius r_{Vc}~2.24 μm for all PDU and TDU cases, while the corresponding peak of fine mode particle arises at a radius r_{Vf} ~0.09-0.12 μm. The dV/dlnr peak and effective radius (r_{coarse}) of coarse mode particles strikingly increase with the increase of AOD ascribed to the intrusion of dust particles. For instance, the AOD_{440}, dV/dlnr peak values of coarse mode, and r_{coarse} for PDU at Minqin site are 0.48, 0.31 $\mu m^3/\mu m^2$, and 1.74 μm, respectively, and corresponding values are 1.13, 0.77 $\mu m^3/\mu m^2$, and 1.93 μm at Lahore site, as shown in Fig. 5(a). The average volume median radii of fine-mode and coarse-mode particles for PDU are 0.159 μm and 2.157 μm, respectively, and 0.140 μm and 2.267 μm for TDU (see Table. 2). The mean volume concentration ratio of coarse mode to fine

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mode particles (C_{vc}/C_{vf}) for Pure Dust is about 18 (varying between 11~31) over East and Central Asia, which is close to the average of ~20 at Dunhuang_LZU during the spring of 2012 (Bi et al., 2014), and much less than that over Saharan pure desert domain (~50) (Dubovik et al., 2002b). The dV/dlnr peak of coarse mode for TDU is clearly smaller than that for PDU, and corresponding mean C_{vc}/C_{vf} value is 9 (~5-11). We attribute the high fractions of coarse-mode particles to high AOD and low Ångström exponent values.

In this paper, we postulate that Asian dust particles only possess scattering and absorption characteristics. And the absorption AOD value (AAOD) at a specific wavelength can be obtained from SSA and AOD, namely, $AAOD_{\lambda}=(1-SSA_{\lambda})\times AOD_{\lambda}$, where λ is the wavelength. Thereby, the corresponding absorption Ångström exponent at 440-870 nm (AAE) is calculated from spectral AAOD values by using a log-linear fitting algorithm. Figure 6 outlines the total average Ångström exponent (α) and absorption Ångström exponent at 440-870 nm, volume concentration of coarse mode in $\mu m^3/\mu m^2$, and volume median radius of coarse mode in μm for TDU ($0.2<\alpha<0.6$) and PDU ($\alpha<0.2$) at selected AERONET sites. There are very big differences of all quantities between PDU and TDU cases, except for some sites (e.g., Dunhuang and Minqin). The primary reason is that we only acquire limited datasets of dust days during spring time at Dunhuang and Minqin sites, which are hardly affected by other anthropogenic pollutants. The AE values of TDU show remarkable changes among each site, ranging from 0.24 to 0.44, whereas corresponding values of PDU keep comparatively slight variations for selected 13 sites (~0.04-0.15). Furthermore, all the AAE values of PDU are greater than 1.5, ranging between 1.65 and 2.36, and the AAE of TDU vary from 1.2 to 2.3. We can conclude that the Asian pure dust aerosols have got AE values smaller than 0.2 and corresponding AAE larger than 1.50, which is another typical feature distinguishing with other non-dust aerosols. Yang et al. (2009) attributed the high AAE values of dust aerosol in China to the presence of ferric oxides. It is evident that volume concentrations of coarse mode for PDU are significantly higher than TDU case, which is expected for the more coarse-mode particles in PDU. While the volume median radius of coarse mode for TDU is greater

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than PDU case, although there are some smaller values for TDU at Dalanzadgad and Yulin sites. This is owing to dust particles at these sites usually mix with other anthropogenic aerosol species and substantially enhance their median radii.

Figure 7 characterizes the overall mean optical properties (e.g., SSA, ASY, Re, and Ri) at 440 nm for selected 13 sites. In general, the absorption capacity of PDU is less than that for TDU. That is, higher SSA and smaller Ri values for PDU, except for Dalanzadgad site. A reasonable interpretation is that threshold criterion method for PDU in this study has effectively eliminated the fine mode aerosols, which are mostly the much stronger absorbing aerosols (e.g., soot and biomass burning aerosol) over East and Central Asia but weaker absorbing pollution aerosols (i.e., sulfate and nitrate) over Dalanzadgad. Wu et al. (2012, 2014) have documented that sulfate and nitrate in background atmosphere most likely originated directly from surface soil at the north and south edges of Taklimakan desert and comprised steadily about 4% of dust particulate matters, which could partially explain our results. Additionally, the overall mean ASY and Re of PDU are greater than that of TDU, which again verifies that the Asian pure dust has got much stronger forward scattering ability than the mixture of Asian dust. Note that the standard deviation of SSA and Ri for PDU is a factor of two to four lower than those from TDU. And the total average values of SSA, ASY, Re, and Ri at 550 nm wavelength for Asian PDU are 0.935±0.014, 0.742±0.008, 1.526±0.029, and 0.00226±0.00056, respectively, while corresponding values are 0.921±0.021, 0.723±0.009, 1.521±0.025, 0.00364±0.0014 for TDU. Yang et al. (2009) took advantage of various in situ aerosol optical and chemical measurements at Xianghe, China during the EAST-AIRC campaign, and deduced a refractive index of 1.53-0.0023i at 550 nm of dust aerosol, which is close to the result of PDU in this study. Nevertheless, the TDU case should be much closer to actual airborne dust aerosol in the real world. When the elevated dusts over desert source regions are transported eastward, they generally mix with other chemical species and react heterogeneously with anthropogenic pollutants, and thus may significantly modify their chemical composition and microphysical properties (Arimoto et al., 2004). Recently, Kim et al. (2011) presented that the annual mean SSA, ASY, Re, and Ri of complex refractive index for nearly pure Saharan dust are 0.944±0.005, 0.752±0.014, 1.498±0.032, and 0.0024±0.0034 at 550 nm, respectively, which are close to our results of pure Asian dust but exist some differences of quantitative values and spectral behaviors.

Average spectral optical properties (at 440, 675, 870, and 1020 nm) for PDU and TDU over East and Central Asian regions are tabulated in Table 2. To our knowledge, this is the first built on Asian dust optical characteristics utilizing multiyear and multi-site AERONET measurements, which will hopefully improve uncertainties of Asian dust shortwave radiative forcing in current regional and global climate models.

4. Discussion

Figure 8 describes the mean spectral behaviors of Re, RI, and SSA for Asian Pure Dust (α<0.2) in this study along with published dust results over various geographical locations (Carlson and Caverly, 1977 or C77; Patterson et al., 1977 or P77; WMO, 1983; Hess et al., 1998 or OPAC; Dubovik et al., 2002b or Persian Gulf; Alfaro et al., 2004 or Ulan Buh Desert; Wang et al., 2004 or ADEC; Todd et al., 2007 or T07). It is well known that a lot of present-day dust models commonly take advantage of the Optical Properties of Aerosols and Clouds (OPAC, Hess et al., 1998) or World Meteorological Organization (WMO, 1983) databases. Curves C77 and P77 show the complex refractive index of Saharan dust in Cape Verde Islands, Barbados West Indies, Tenerife Canary Islands obtained from laboratory analysis by Carlson and Caverly (1977) and Patterson et al. (1977), respectively. Curve P77 gives one of the most widely used datasets of Ri value in the range 300-700 nm. Curve Persian Gulf(98-00) displays the refractive index and SSA of dust over Bahrain-Persian Gulf Desert during period of 1998-2000 derived from Dubovik et al. (2002b). Curve T07 shows the optical properties of mineral dust over Bodélé Depression of northern Chad during 2005 retrieved from Cimel sun photometer by Todd et al. (2007). And the curves ADEC and Ulan Buh exhibit the dust absorptive properties over aforementioned Taklimakan Desert and Ulan Buh Desert of northwest China by Wang et al. (2004) and Alfaro et al. (2004). Figure 8(a) presents that the spectral behaviors

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of Re have relatively slight variations with values ranging from 1.50-1.56 apart from T07 that shows lower Re values of 1.44-1.47. Todd et al. (2007) utilized Scanning Electron Microscope (SEM) analysis of airborne dust material and confirmed that the mineral dust is dominated by fragmented fossil diatoms from the dry lake bed of the Bodélé Depression, which is to some extent different from the typical desert soil. As shown in Figure 8(b), wavelength dependences of Ri exhibit comparably greater differences in UV wavebands. In mid-visible and near infrared, our results are slightly larger than Persian Gulf (98-00) and T07 that are retrieved from Cimel sun photometer, but still comparable. It is very distinct that the absorbing ability of Asian pure dust (α<0.2) in the whole spectrum range is about a factor of 4 smaller than current dust models (WMO, 1983; Hess et al., 1998), and is a factor of 2 to 3 lower than the results from in situ measurements combined with laboratory analysis or model calculations (Carlson and Caverly, 1977; Patterson et al., 1977; Wang et al., 2004). Meanwhile, the wavelength dependences of SSA agree well with Persian Gulf (98-00) and Ulan Buh Desert, but are much higher than OPAC. The discrepancy increases dramatically with decreasing wavelength. Such big differences of dust absorption capacity for diverse dust models (OPAC and WMO) and researches will certainly lead to different radiative impacts on regional or global climate change.

Figure 9 draws the aerosol shortwave direct radiative effects (ARF) at the top of atmosphere (TOA), at the surface (SFC), and in the atmospheric layer (ATM) for Asian Pure Dust (α<0.2) and Transported Anthropogenic Dust (0.2<α<0.6) acquired in this study, and corresponding ARF values for OPAC Mineral accumulated (Mineral acc.) and transported (Mineral tran.) modes are also presented for comparison. We make use of the Santa Barbara Discrete-ordinate Atmospheric Radiative Transfer model (SBDART, Ricchiazzi et al., 1998) to calculate the ARF, which has been proved to be a reliable software code and widely used for simulating plane-parallel radiative fluxes in the Earth's atmosphere (Halthore et al., 2005; Bi et al., 2013). The main input parameters of spectral AOD, surface albedo, WVC, and columnar ozone amount are prescribed to same values, and the spectral SSA, ASY, Re, and Ri values are obtained from aforementioned various dust models. It is evident that Earth's energy budget is modulated and redistributed by different absorbing properties of mineral dusts. The results indicate that the cooling rate at SFC (negative radiative forcing) gradually increases with PDU (α<0.2), TDU (0.2<α<0.6), OPAC Mineral accumulated and transported dust modes. By contrast, the cooling intensity at TOA gradually decreases with diverse dust cases, and even becomes positive radiative forcing for OPAC transported dust mode, with ARF varying from -15.6, -13.8, -6.9, and +0.24 Wm^{-2}, respectively. Therefore, the heating intensity in the atmospheric layer sharply increases from +22.7, +29.5, +46.6, and +58.3 Wm^{-2}. The heating rate in ATM for OPAC Mineral (acc. and tran.) modes is about two-fold greater than Asian dust cases (PDU and TDU). Such large diabatic heating rates might warm the dust layer, suppress the development of convection under the lower atmosphere, thus exert profound impacts on the atmospheric dynamical and thermodynamic structures and cloud formation together with the strength and occurrence frequency of precipitation (Rosenfeld et al., 2001; Huang et al., 2010a; Creamean et al., 2013). Hence, accurate and reliable absorbing characteristics of Asian dust should be considered in present-day regional climate models.

5. Summary

In this study, we have proposed two threshold criteria to discriminate two types of Asian dust: Pure Dust (PDU, α<0.2) and Transported Anthropogenic Dust (TDU, 0.2<α<0.6). PUD can represent nearly "pure" dust in desert source regions and decrease disturbance of other non-dust aerosols, which would also exclude some fine mode of dust particles. The spectral behaviors of TDU exhibit similar variations with PDU, but show much stronger absorption and weaker scattering than PDU cases. There are two markedly identifiable characteristics for Asian PDU. (I) spectral SSA values systematically increase with wavelength from 440 nm to 675 nm and remain almost neutral or slight increase for the wavelength greater than 675 nm, whereas an opposite pattern is shown for imaginary part of refractive index. (II) Asian pure dust aerosols have got AE values smaller than 0.2 and AAE larger than 1.50. Compared with current common dust models (e.g., OPAC and WMO), Asian dust aerosol has relatively weak absorption for wavelengths greater than 550 nm (SSA~0.96-0.99), but presents a moderate absorption in the blue spectral range (SSA_{440}~0.92-0.93). The overall average values of SSA, ASY, Re, and Ri at 550 nm wavelength for Asian PDU are 0.935±0.014, 0.742±0.008, 1.526±0.029, and 0.00226±0.00056, respectively, while corresponding values are 0.921±0.021, 0.723±0.009, 1.521±0.025, 0.00364±0.0014 for TDU.

It should be noted that the definition of anthropogenic dust in this paper is ambiguous, and TDU here represents more accurately dominant dust mixing with other anthropogenic aerosols. Because it is very difficult to quantify the anthropogenic contribution due to large uncertainties in defining the anthropogenic fraction of ambient dust burden (Sokolik et al., 2001; Huang et al., 2015). Diverse human activities (e.g., agricultural cultivation, desertification, industrial activity, transportation, and construction in urbanization) in vulnerable environments might modify the land use and Earth's surface cover, and would affect the occurred frequency and intensity of anthropogenic dust. Hence, the optical features of anthropogenic dust aerosols are dependent on the source regions and chemical compositions. However, as concluded by Huang et al. (2015), anthropogenic dust generated by human activities mainly comes from semi-arid and semi-humid regions (Guan et al., 2016) and is generally mixed with other types of aerosols within the PBL. And we primarily investigated dust aerosols in arid or semi-arid regions over East and Central Asia, where are somewhat disturbed by human activities. Therefore, the key optical properties of TDU derived from this study should to some extent contain the anthropogenic fraction. To fully elucidate exact optical properties of anthropogenic dust, we need to explore detailed morphology, mineralogy, and chemical compositions by means of in situ measurements, laboratory analysis, active and passive remote sensing methods (e.g., multi-wavelength lidar, AEROENT, MODIS) as well as model calculations.

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Table 1. Overall average and standard deviation of key optical properties at 550 nm (e.g., single-scattering albedo, asymmetry factor, real part and imaginary part of complex refractive index) for Asian pure Dust (PDU). Ångström wavelength exponent (α) is in the range of 440-870 nm. Minimum and maximum values of the optical properties are in parenthesis for each corresponding column. Measuring period and the total number of PDU (α<0.2) and Transported Anthropogenic Dust (TDU, 0.2<α<0.6) days are in the parenthesis for the first and last column, respectively.

Site (sampled period)	SSA (min, max)	ASY (min, max)	Re (min, max)	Ri ($\times 10^{-3}$)	Ångström (440-870 nm)	PDU/days (TDU)
SACOL (2006-2012)	0.932±0.018 (0.888, 0.971)	0.741±0.012 (0.715, 0.771)	1.534±0.044 (1.438, 1.60)	2.251±0.788 (0.913, 5.51)	0.120±0.049 (0.0, 0.198)	38 (97)
Dalanzadgad (1997-2014)	0.930±0.012 (0.912, 0.949)	0.746±0.010 (0.724, 0.766)	1.512±0.046 (1.447, 1.60)	2.407±0.414 (1.649, 3.19)	0.127±0.079 (-0.06, 0.199)	8 (6)
Beijing (2001-2015)	0.917±0.020 (0.863, 0.963)	0.742±0.012 (0.716, 0.769)	1.557±0.043 (1.401, 1.60)	2.801±0.865 (1.032, 6.20)	0.117±0.067 (-0.048, 0.199)	46 (67)
Yulin (2001-2002)	0.907±0.024 (0.863, 0.952)	0.748±0.010 (0.731, 0.771)	1.559±0.038 (1.476, 1.60)	3.564±1.589 (1.370, 7.92)	0.077±0.068 (-0.024, 0.188)	13 (16)
Dushanbe (2010-2015)	0.941±0.012 (0.916, 0.959)	0.739±0.011 (0.710, 0.765)	1.529±0.041 (1.436, 1.60)	2.011±0.551 (1.022, 3.475)	0.128±0.054 (-0.02, 0.198)	26 (95)
Karachi (2006-2014)	0.945±0.012 (0.916, 0.977)	0.741±0.011 (0.714, 0.767)	1.518±0.030 (1.449, 1.60)	1.938±0.561 (0.758, 3.439)	0.141±0.041 (-0.005, 0.20)	83 (286)
Lahore (2007-2015)	0.930±0.014 (0.901, 0.957)	0.740±0.010 (0.721, 0.765)	1.519±0.038 (1.432, 1.60)	2.253±0.611 (1.207, 3.623)	0.136±0.052 (0.023, 0.198)	26 (248)
IASBS (2010-2013)	0.933±0.017 (0.883, 0.958)	0.725±0.011 (0.704, 0.746)	1.572±0.024 (1.525, 1.60)	2.290±0.845 (1.245, 5.029)	0.098±0.050 (0.021, 0.195)	19 (12)
Kandahar (2008/04-06)	0.925±0.013 (0.903, 0.955)	0.729±0.017 (0.700, 0.768)	1.534±0.035 (1.492, 1.60)	2.855±0.775 (1.445, 4.65)	0.147±0.054 (0.00, 0.199)	10 (4)
Dunhuang (2001/03-05)	0.947±0.015 (0.918, 0.970)	0.745±0.013 (0.723, 0.761)	1.547±0.037 (1.494, 1.60)	1.714±0.697 (1.014, 3.14)	0.039±0.029 (-0.003, 0.091)	6 (0)
Dunhuang_LZU (2012/04-05)	0.958±0.007 (0.951, 0.968)	0.741±0.021 (0.707, 0.771)	1.495±0.042 (1.451, 1.580)	1.589±0.292 (1.092, 1.84)	0.153±0.026 (0.117, 0.184)	5 (4)
Inner_Mongolia (2001/04-05)	0.948±0.012 (0.930, 0.960)	0.751±0.006 (0.743, 0.759)	1.499±0.042 (1.426, 1.54)	1.641±0.457 (1.169, 2.45)	0.069±0.054 (0.011, 0.165)	4 (1)
Minqin (2010/05-06)	0.945±0.002 (0.942, 0.947)	0.756±0.014 (0.740, 0.764)	1.469±0.023 (1.449, 1.494)	2.036±0.220 (1.883, 2.29)	0.119±0.023 (0.103, 0.146)	2 (0)
Overall Mean	**0.935±0.014**	**0.742±0.008**	**1.526±0.029**	**2.258±0.556**	**0.113±0.033**	**PDU**
Overall Mean	**0.921±0.021**	**0.723±0.009**	**1.521±0.025**	**3.643±1.372**	**0.355±0.06**	**TDU**

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Table 2. Spectral optical properties of Pure Dust (α<0.2) and Transported Anthropogenic Dust (0.2<α<0.6) averaged for 13 sites over East and Central Asia areas.

Asian Dust	Pure Dust (α<0.2)	Transported Anthropogenic Dust (0.2<α<0.6)
ω_0(440/675/870/1020)	0.906/0.962/0.971/0.975 ±0.009	0.897/0.943/0.954/0.959 ±0.019
Re(440/675/870/1020)	1.520/1.533/1.517/1.503 ±0.026	1.509/1.533/1.532/1.525 ±0.027
Ri(440/675/870/1020) **$\times 10^{-3}$**	3.413/1.574/1.449/1.449 ±0.450	5.064/2.737/2.510/2.486 ±1.300
ASY(440/675/870/1020)	0.758/0.727/0.724/0.726 ±0.008	0.736/0.711/0.710/0.712 ±0.009
r_{Vf} (μm); σ_f	0.159±0.029	0.140±0.011
r_{Vc} (μm); σ_c	2.157±0.112	2.267±0.214
Cvf ($\mu m^3/\mu m^2$)	0.037±0.011; 0.06×τ(1020)-0.001	0.038±0.011; 0.12×τ(1020)-0.014
Cvc($\mu m^3/\mu m^2$)	0.632±0.167; 0.88×τ(1020)-0.07	0.343±0.084; 0.90×τ(1020)-0.06
Cvc/Cvf	17.9 (11~31)	9.1 (5~11)

Each variable is accompanied by a standard deviation (e.g.., ±0.01). r_{Vf} and r_{Vc} are the volume median radii of fine-mode and coarse-mode particles in μm; Cvf and Cvc denote the volume concentrations of fine-mode and coarse-mode particles in $\mu m^3/\mu m^2$, respectively. The dynamic dependencies of dust optical properties are exhibited as functions of AOD_{1020}, with correlation coefficients greater than 0.93 for all cases.

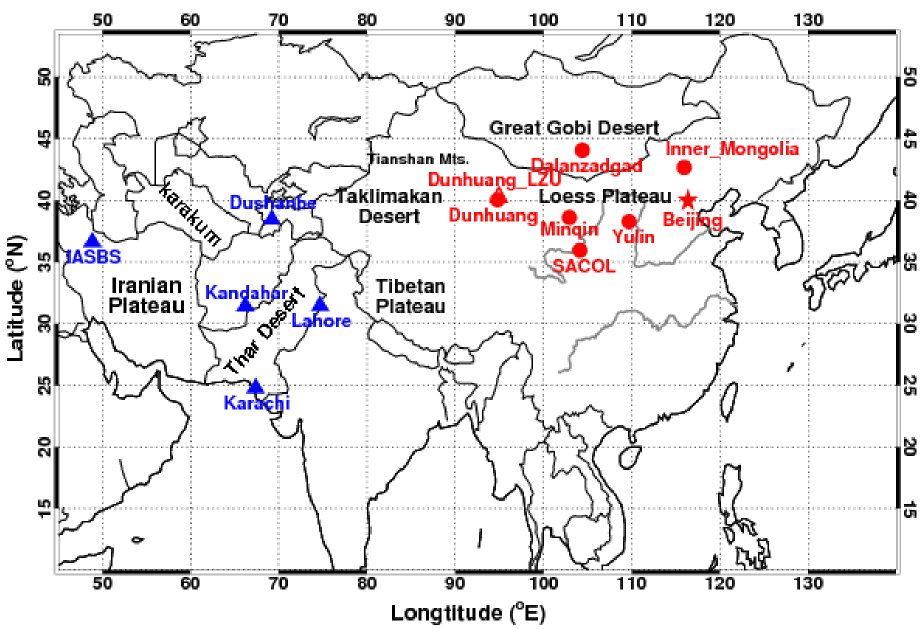

Figure 1. Geographical location of selected 13 AERONET sites in this study. Eight sites over East Asian region are labeled with red colors, and five sites over Central Asian region are labeled with blue colors. The major Great deserts or Gobi deserts along with plateaus are marked with black font.

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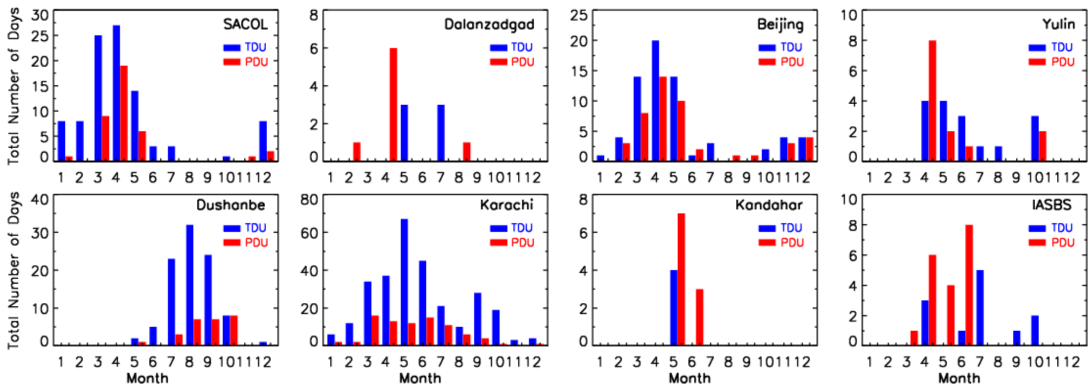

Figure 2. Occurrence frequency of total number days for Pure Dust (α<0.2, PDU with red color) and Transported Anthropogenic Dust (0.2<α<0.6, TDU with blue color) at selected four East Asian sites (top panel) and four Central Asian sites (bottom panel).

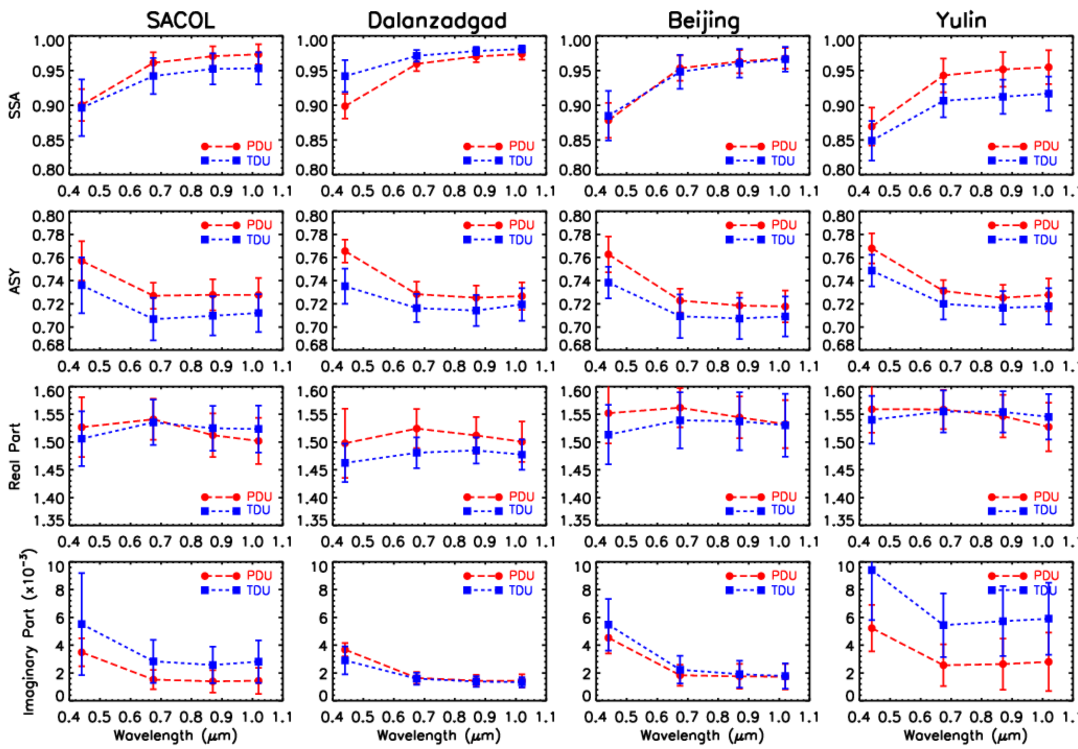

Figure 3. Overall average spectral behavior of key optical properties for Pure Dust (α<0.2, PDU with red circle) and Transported Anthropogenic Dust (0.2<α<0.6, TDU with blue square) at selected four East Asian sites (SACOL, Dalanzadgad, Beijing and Yulin). The error bars indicate plus or minus one standard deviation.

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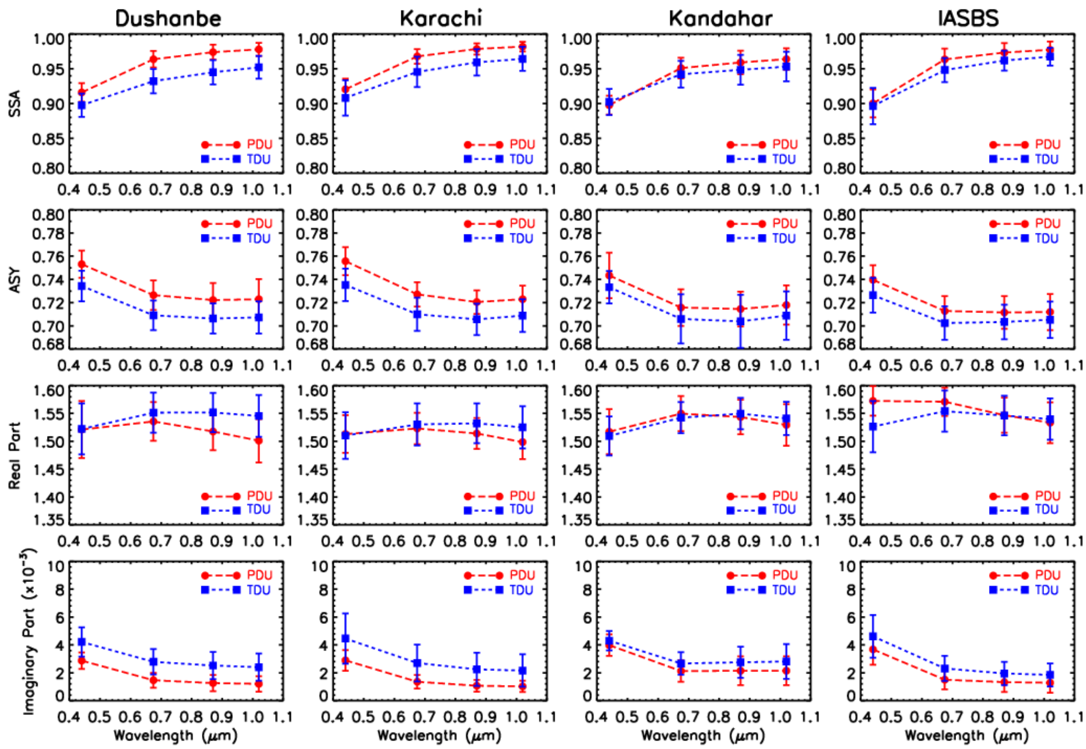

Figure 4. The same as Figure 3, but for selected four Central Asian sites (Dushanbe, Karachi, Kandahar and IASBS).

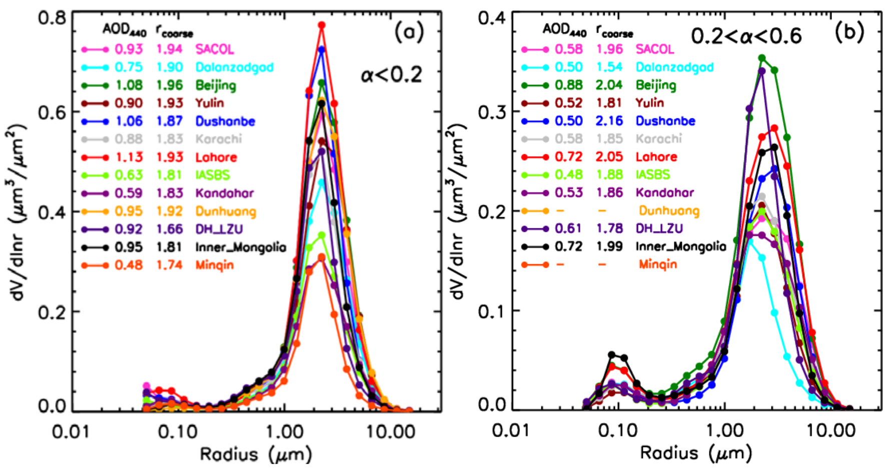

Figure 5. Overall average of aerosol volume size distributions in the entire atmospheric column for (a) Pure Dust (α<0.2) and (b) Transported Anthropogenic Dust (0.2<α<0.6) at selected 13 AERONET sites. Corresponding aerosol optical depth at 440 nm (AOD_{440}) and effective radius of

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coarse mode (r_{coarse}) in µm are also shown.

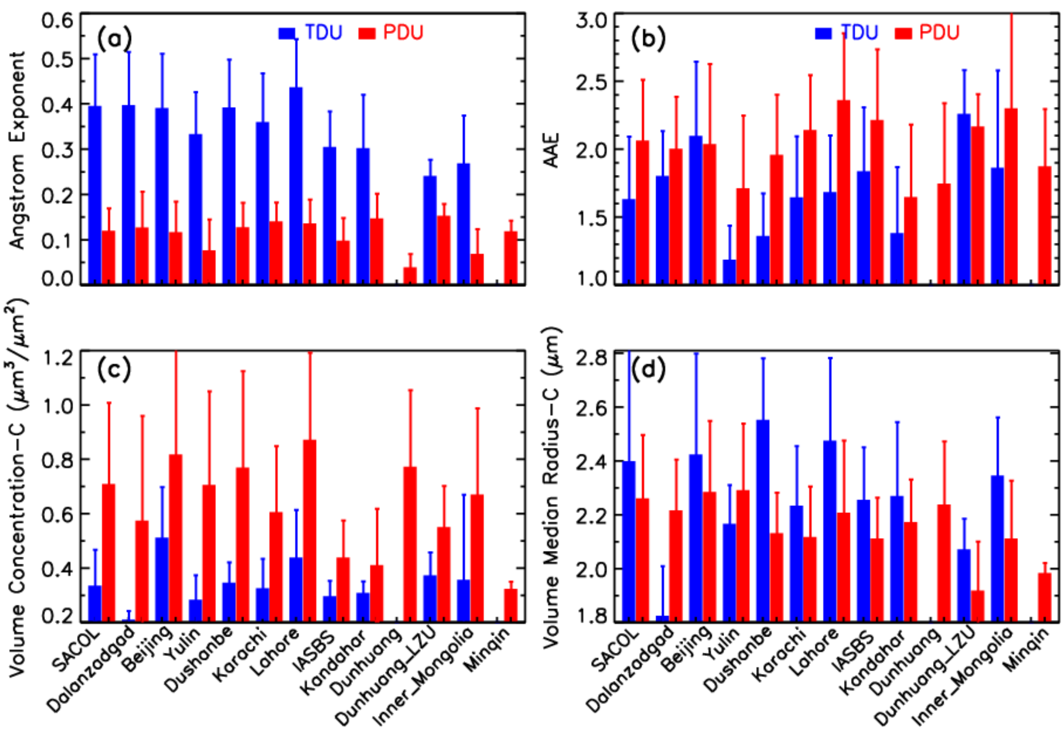

Figure 6. Total average values of (a) Ångström exponent (440-870 nm), (b) absorption Ångström exponent at 440-870 nm (AAE), (c) volume concentration of coarse mode ($\mu m^3/\mu m^2$), and (d) volume median radius of coarse mode in µm for Transported Anthropogenic Dust ($0.2<\alpha<0.6$, blue color) and Pure Dust ($\alpha<0.2$, red color) at 13 selected AERONET sites. The error bars indicate plus or minus one standard deviation.

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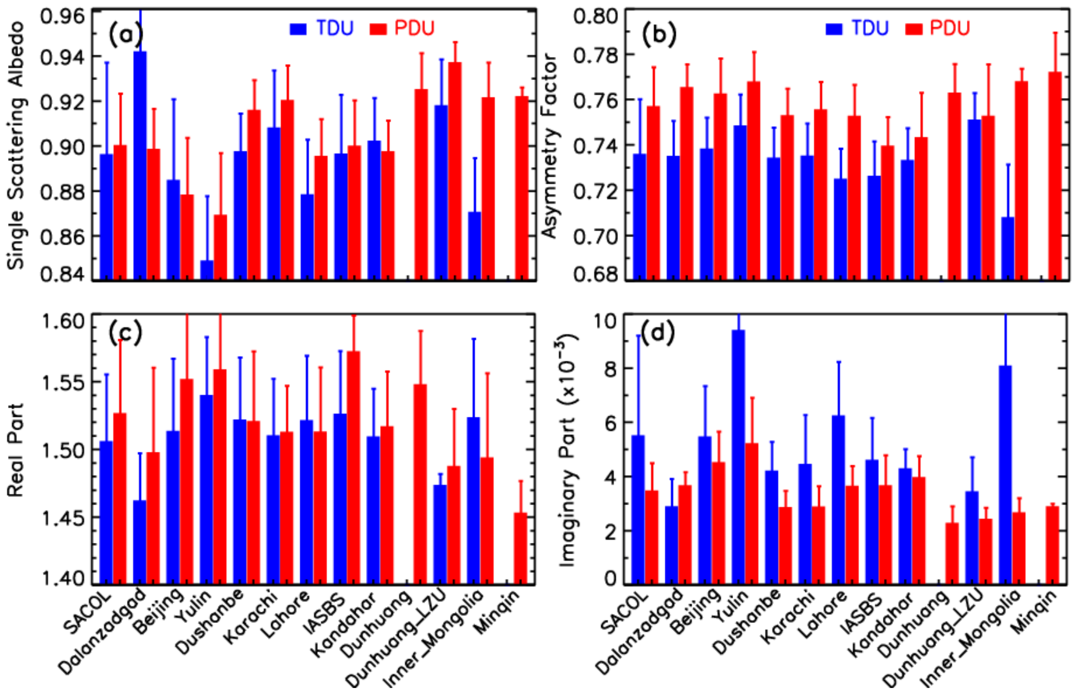

Figure 7. The same as Figure 5, but for (a) sing-scattering albedo, (b) asymmetry factor, (c) real part and (d) imaginary part of complex refractive index at 440 nm.

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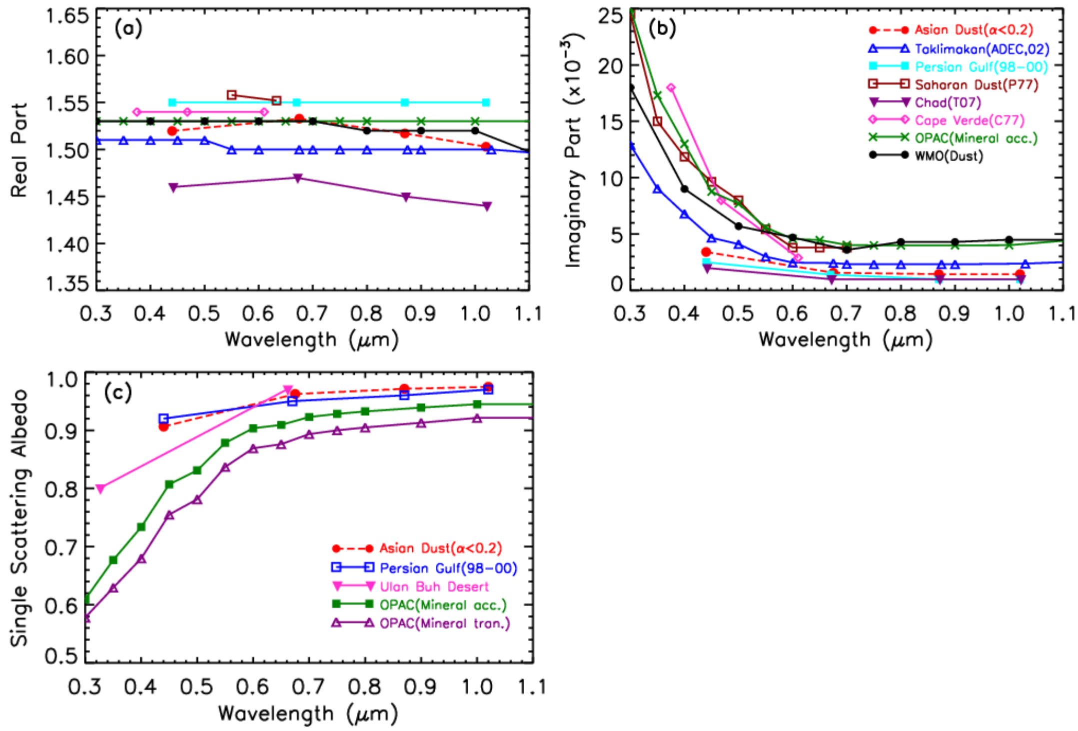

Figure 8. Mean spectral behaviors of (a) real part, (b) imaginary part of complex refractive index, and (c) single-scattering albedo for Asian Pure Dust (α<0.2) calculated for 13 AERONET sites, and results of current common dust models (OPAC, WMO), Bahrain-Persian Gulf of Desert dust (1998-2000), Saharan dust (Chad, Cape Verde Islands), and Chinese Gobi desert (Taklimakan, Ulan Buh Desert) are also shown for comparison.

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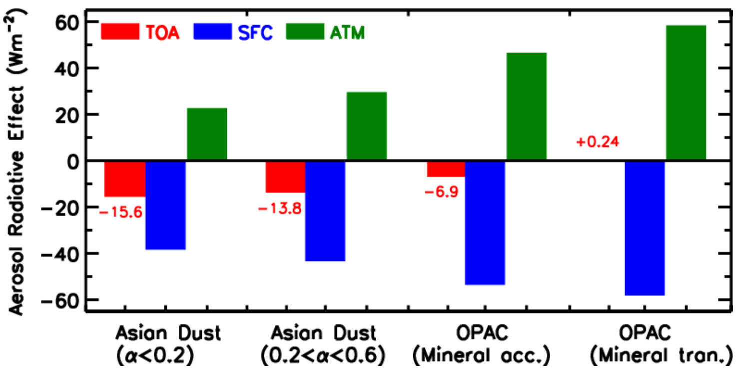

Figure 9. Aerosol shortwave direct radiative effects at the top of the atmosphere (TOA, red color), at the surface (SFC, blue color), and in the atmospheric layer (ATM, green color) for Asian Pure Dust ($\alpha<0.2$) and Transported Anthropogenic Dust ($0.2<\alpha<0.6$) computed in this study, and corresponding values for OPAC Mineral accumulated (Mineral acc.) and transported (Mineral tran.) modes are also presented for comparison.