A Review of Aerosol Optical Properties and Radiative Effects

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ABSTRACT

Atmospheric aerosols influence the earth's radiative balance directly through scattering and absorbing solar radiation, and indirectly through affecting cloud properties. An understanding of aerosol optical properties is fundamental to studies of aerosol effects on climate. Although many such studies have been undertaken, large uncertainties in describing aerosol optical characteristics remain, especially regarding the absorption properties of different aerosols. Aerosol radiative effects are considered as either positive or negative perturbations to the radiation balance, and they include direct, indirect (albedo effect and cloud lifetime effect), and semi-direct effects. The total direct effect of anthropogenic aerosols is negative (cooling), although some components may contribute a positive effect (warming). Both the albedo effect and cloud lifetime effect cool the atmosphere by increasing cloud optical depth and cloud cover, respectively. Absorbing aerosols, such as carbonaceous aerosols and dust, exert a positive forcing at the top of atmosphere and a negative forcing at the surface, and they can directly warm the atmosphere. Internally mixed black carbon aerosols produce a stronger warming effect than externally mixed black carbon particles do. The semi-direct effect of absorbing aerosols could amplify this warming effect. Based on observational (ground- and satellite-based) and simulation studies, this paper reviews current progress in research regarding the optical properties and radiative effects of aerosols and also discusses several important issues to be addressed in future studies.

Key words: aerosol, optical property, radiative effect

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1. Introduction

Aerosols, which are an important component of the earth atmosphere (Ramanathan et al., 2001a; Solomon et al., 2007), comprise a mixture of mainly sulfate, soil dust, carbonaceous material, and sea salt. Atmospheric aerosols are dispersed worldwide. The Indo-Asian haze that was documented by the Indian Ocean Experiment (INDOEX) spread widely across most of the North Indian Ocean, South and Southeast Asia (Müller et al., 2003; Ramanathan et al., 2001b). Biomass burning and dust aerosols from North Africa (Sahara Desert and Sahel regions) are distributed over most of the subtropics (Swap et al., 1992; Chiapello et al., 1997; Goudie and Middleton, 2001). Asian dust (Zhang et al., 1998; Wang et al., 2000; Nakajima et al., 2003; Zhang et al., 2003; Arimoto et al., 2006; Mikami et al., 2006; Hsu et al., 2013; Xu and Ma, 2013) and anthropogenic aerosols can travel across the Pacific following the jet stream into the North American continent (Uno et al., 2001; Gong et al., 2006; Zhao et al., 2006; Jiang et al., 2007). Recently, some studies have also reported the properties of Arctic aerosols (e.g., Breider et al., 2014) and their relationship with Arctic

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warming. The impact of these aerosols is becoming an important environmental and climatic problem (Penner et al., 1992; Jaffe et al., 1999), given of their global distribution.

Aerosols affect the earth's climate system by altering the radiative properties of the atmosphere. Atmospheric aerosols influence the earth's radiation budget directly through scattering and absorbing solar radiation (Charlson et al., 1991, 1992; Miller and Tegen, 1998; Hayasaka et al., 2007; Liu et al., 2011) and indirectly through affecting cloud properties (e.g., Twomey, 1974; Twomey et al., 1984; Ackerman et al., 2000; Huang et al., 2006a, c, 2009, 2010). Studies of the optical properties of aerosols are crucial to fully understand their radiative effects.

Different aerosols scatter or absorb sunlight to varying degrees, depending on their optical properties. Although cooling (negative forcing) is a consequence of the total direct effect of aerosols, different aerosols have different effects. Absorbing aerosols such as dust (e.g., Overpeck et al., 1996) and black carbon (BC) (e.g., Gu et al., 2010) are able to heat the atmosphere by absorbing solar and thermal radiation (Jacobson, 2002; Menon et al., 2002a; Andreae and Gelencsér, 2006). In contrast, non-absorbing aerosols such as sulfate scatter solar radiation (Charlson et al., 1991, 1992; Kiehl and Briegleb, 1993; Pósfai et al., 1999) and generate relatively weaker atmospheric heating effect than the absorbing aerosols.

In addition to the profound direct impact of aerosols on the radiation budget of the earth-atmosphere system, the development of clouds in a polluted environment can also significantly affect the radiation budget, and changes in cloud properties can have an influence on precipitation (e.g., Flossmann et al., 1985; Andreae et al., 2004; Kaufman et al., 2005a; Andreae and Rosenfeld, 2008; Jiang et al., 2011; Gu et al., 2012). Aerosols play a critical role in the process of cloud formation, although the absorbing and non-absorbing aerosols affect clouds differently (Kaufman and Koren, 2006). Whereas absorbing aerosols prevent clouds from forming, non-absorbing aerosols extend cloud lifetimes and are associated with enhanced cloud cover (Coakley et al., 1987; Platnick et al., 2000). Absorbing aerosols suspended near clouds are also believed to contribute to cloud evaporation (Hansen et al., 1997; Huang et al., 2010). However, numerous studies describe additional mechanisms whereby absorbing aerosols may either reduce or increase cloud cover (Chen et al., 2000; Ackerman et al., 2000; Jacobson, 2002; Small et al., 2011). In contrast, non-absorbing aerosols, such as sulfate, are largely derived from anthropogenic activities and are concentrated mainly in the Northern Hemisphere. Their rapid increase in the atmosphere could have contributed significantly to the cross-equatorial sea surface temperature gradient, which substantially alters low-latitude cloud, circulation, and rainfall (Hulme and Kelly, 1993; Williams et al., 2001; Rotstayn and Lohmann, 2002; Ackerley et al., 2011).

The effects of aerosols on radiation and climate through their interaction with clouds are complex and incompletely captured by climate models (e.g., Takemura et al., 2005, 2007; Suzuki et al., 2008). As suggested in the 5th Assessment Report (AR5) of the Intergovernmental Panel on Climate Change (IPCC; Stocker et al., 2013), scientific understanding of aerosol radiative effects is still at mid-low or low levels.

This review summarizes the progress made in studies of aerosol optical properties and their radiative effects. The review has concentrated on the literature regarding aerosol optical properties, and radiative effects of the main types of aerosols (dust, carbonaceous, sulfate, and sea salt). Based on the latest literature on aerosol optical properties and/or their radiative effects, previous studies have been traced and major results are taken into consideration. Section 2 focuses on the aerosol optical properties, and the aerosol radiative effects are discussed in Section 3. Summary and discussion are provided in Section 4.

2. Optical properties of aerosols

The interaction of aerosols with radiation is usually measured by aerosols' optical properties, e.g., the scattering coefficient, absorption coefficient, aerosol optical depth (AOD), single scattering albedo (SSA),

and Angström exponent (AE). The AOD is a parameter used to measure the magnitude of aerosol extinction due to scattering and absorption, integrated in the vertical column. It represents the e-folding length of the decrease in a direct beam when traveling through the aerosol layer. The SSA is a ratio of the scattering coefficient to the extinction coefficient and measures the relative importance of scattering and absorption. The aerosol effects on the radiation budget at the top of the atmosphere (TOA) switch from net cooling to warming at a certain value of the SSA, depending on the local surface albedo (Hansen et al., 1997). The AE represents the wavelength dependence of AOD, with high values of AE indicating small particles and low values representing large particles.

As the uncertainties associated with aerosol radiative effects stem mainly from optical properties with high inhomogeneous horizontal and vertical distributions (Liu et al., 2009; van Donkelaar et al., 2010; Zhang and Tang, 2012), attempts to better understand aerosol optical properties are underway (e.g., Zhou et al., 2011; Gao et al., 2012; Wang Na et al., 2013). The investigation of aerosol optical properties has encompassed a range of complementary approaches, including in-situ monitoring (Nakajima et al., 1996; Dubovik et al., 2002; Anderson et al., 2003; Yan et al., 2010; Yang et al., 2012; Zheng et al., 2013; Che et al., 2014), satellite remote sensing (Huang et al., 2008a, b; Chen et al., 2010; Wang W. C. et al., 2010; Cai et al., 2011; Wang Zhao et al., 2013; Ahn et al., 2014), and numerical modeling (Takemura et al., 2002, 2003; Kinne et al., 2006; Textor et al., 2006; Han et al., 2010).

Table 1 summarizes some major ground-based aerosol observation networks. Ground-based measurements can provide databases to validate satellite retrievals and model simulations. Among the ground-based aerosol networks, Aerosol Robotic Network (AERONET) and Skyradiometer Network (SKYNET) are two major long-term, continuous, and readily accessible public domain databases that have been used in studies of aerosol optical properties. AERONET includes information for 897 sites around the world, covering almost all major tropospheric aerosol regimes (Holben et al., 1998). SKYNET includes information for about 25 sites around the world (Nakajima et al., 1996; Campanelli et al., 2004). Both AERONET and SKYNET observations are collected via passive remote sensing depending on sunlight, and provide information on aerosols at the surface. As some compensation, Raman lidar and micro-pulse lidar, which are used in lidar networks such as Micro-Pulse Lidar Network (MPLNET), have been used to retrieve profiles of aerosol backscattering and extinction during both day and night.

Satellite remote sensing is the best, and indeed, the only way to observe aerosols on the global scale due to the short lifetime of aerosols and their complex chemical composition. To effectively determine the properties of aerosols, some sensors such as Advanced Very High Resolution Radiometer (AVHRR) (Heidinger et al., 2002; Zhao et al., 2008) and Total Ozone Meteorological Satellite (TOMS) (Ginoux and Torres, 2003) have been designed to monitor aerosols from space. Satellite remote sensing now includes new and enhanced sensors such as Polarization and Directionality of the Earth's Reflectance (POLDER; Fougnie et al., 1999), Moderate Resolution Imaging Spectroradiometer (MODIS; Salomonson et al., 1989), and Multi-angle Imaging SpectroRadiometer (MISR; Fisher et al., 2014). The launch of satellite-borne lidars such as Geoscience Laser Altimeter System (GLAS; Zwally et al., 2002) and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO; Winker et al., 2006) has further enabled profiling of vertical aerosol distribution. These sensors allow quantitative analysis of aerosol optical properties, especially AOD, and provide additional information regarding aerosol size, SSA, and refractive index. Such advanced sensors will also provide aerosol global distribution information, seasonal and interannual variations in the sources, optical properties, and the direct and indirect effects of aerosols.

Combining ground-based measurements and satellite remote sensing makes it possible to obtain relatively reliable information regarding global aerosol distribution. Data from AERONET and satellite remote sensing (Fig. 1) indicate that high aerosol loads over the continents are found in and downwind of the

regions with specific sources. For example, high AOD values have been recorded in the east of China due to the transport of Asian dust. Such observational data provide a basis for understanding the properties

Table 1. List of main ground-based networks for aerosol observation

Name	Product	Major reference
AERONET	Level 1.0: unscreened and may not have final calibration applied Level 1.5: automatically cloud cleared but may not have final calibration applied Level 2.0: pre- and post-field calibration applied, automatically cloud cleared and manually inspected	(Holben et al., 1998)
SKYNET	Level 1: range of scattering angle: 3°–30°, use of only diffuse radiation calibration (Disk scan method), including Level 1.30 (global irradiance data are not available for cloud screening) and Level 1.50 (global irradiance data are available for cloud screening) Level 2: range of scattering angle: full, use of diffuse radiation (Disk scan method) and direct radiation method (Improved Langley method), including Level 2.00 (global irradiance data are not available for cloud screening) and Level 2.20 (global irradiance data are available for cloud screening)	(Nakajima et al., 1996; Campanelli et al., 2004)
MPLNET	Level 1: normalized relative backscatter signals Level 1.5a: real time aerosol properties Level 1.5b: real time aerosol, cloud, and PBL heights Level 1.5c: real time cloud properties Level 1.5d: polar stratospheric clouds Level 2.0: quality assured aerosol properties Level 2.0a: quality assured aerosol properties Level 2.0b: quality assured aerosol, cloud, and PBL heights Level 2.0c: quality assured aerosol, cloud, and PBL heights	(Campbell et al., 2002; Welton et al., 2000)
EARLINET	Backscatter and coefficient profiles at 355, 532, and 1064 nm	(Matthais et al., 2004)
AD-Net	Attenuated backscatter coefficient at 532 and 1064 nm; volume depolarization ratio, aerosol extinction coefficient, dust extinction coefficient, spherical particle extinction coefficient, and aerosol depolarization ratio at 532 nm	(Shimizu et al., 2004)

EARLINET: European Aerosol Research Lidar Network; AD-Net: Asian Dust Network.

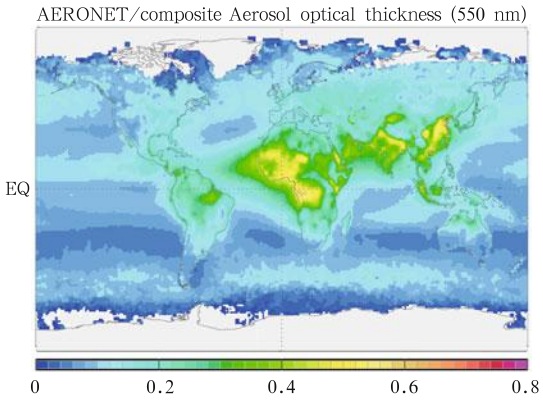

Fig. 1. Global distribution of AOD. The image was obtained by merging data from several satellite sensors with ground-based sun photometer measurements (Andreae and Rosenfeld, 2008).

of aerosols.

It is extremely difficult to observe aerosols by using ground or satellite remote sensing techniques, when the aerosols are near or inside clouds. One way to deal with this observational difficulty is to analyze an extended record of satellite measurements in a statistical sense (e.g., Kaufman et al., 2005b). The presence of aerosols inside clouds can be inferred by using other observable gaseous pollutants as a proxy for the aerosol. Carbon monoxide (CO) is a good aerosol proxy because incomplete combustion produces both CO and aerosols, with possible sources including forest fires, coal burning power plants, and fossil fuel-powered automobiles (e.g., Jiang et al., 2008; Su et al., 2011). Additionally, aerosol models can be used as an effective method to offset the limitations of observations. From the early models with only simple schemes for physical and chemical processes (Cooke and Wilson, 1996), a series of aerosol models have been developed, e.g., the dynamic aerosol (Remer and Kaufman, 1998), GLObal Model of Aerosol Processes (GLOMAP; Spracklen et al., 2005), and Spectral Radiation-Transport Model for Aerosol Species (SPRINTARS) models (Takemura et al., 2002; Takemura, 2012; Dai et al., 2014). Recently, the Beijing Climate Center atmospheric general circulation model (BCC_AGCM) has been used to produce simulations of aerosol properties and radiative effects. For example, Zhang et al. (2012b) simulated the aerosol (including sulfate, dust, carbonaceous material, and sea salt) optical properties over globe (Fig. 2). The highest simulated AOD values (0.4–0.7) occurred over the Sahara Desert, followed by Arabia in West Asia (0.2–0.4). The simulated pattern of AOD distribution is generally in good agreement with the satellite data in Fig. 1 for all areas of the globe. For South America, the AOD simulated by BBC_AGCM was lower than observation. For Africa, high AOD values were observed from the Sahara Desert to southern Africa, whereas the simulated high AOD values only occurred around the Sahara Desert. The pattern of simulated AOD over East Asia was in agreement with observations, although the simulated values were lower than the observed. This discrepancy may arise from the parameterization of aerosols in the model. The key factors affecting the aerosol simulation include the land surface properties, soil moisture, wind speed, and parameterization of the aerosol's emission, transport,

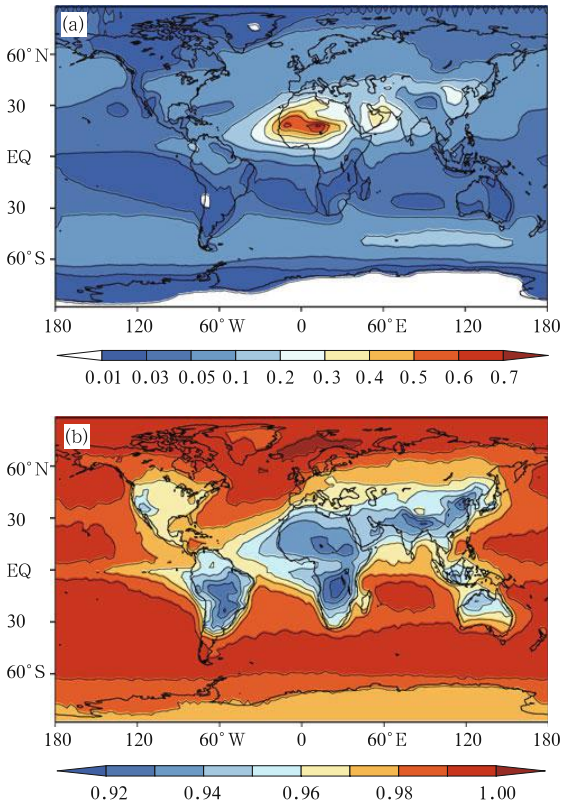

Fig. 2. Global annual mean distributions of simulated (a) total AOD and (b) single scattering at 550 nm (Zhang et al., 2012b).

and deposition.

In addition to satellite remote sensing, model simulation could also provide a global distribution of aerosol properties, which enables the validation using the aerosol observational data, especially for the aerosol global distribution.

Unfortunately, both ground- and satellite-based observations and model simulations contain uncertainties. For example, satellite aerosol retrievals can be contaminated by thin cirrus, resulting in an overestimate of AOD by about 0.02 ± 0.005 (mean ± standard deviation) (Kaufman et al., 2005b). Combination of observation and model simulation (i.e., data assimilation) is a good way to reduce the uncertainties to some degree. Aerosol data assimilation (Schutgens et al., 2010a, b) may overcome the respective weaknesses of observation and simulation, minimize the misfit between them, and produce an optimal estimate of aerosol optical properties.

Under whole-sky conditions, the annual and global average AOD was reported (Yu et al., 2006) to be 0.191 ± 0.017 over land and 0.126 ± 0.046 over ocean. Many studies have focused on aerosol optical properties over Asia (Xin et al., 2005; Huang et al., 2008a, b; Che et al., 2009, 2013; Ge et al., 2010; Wang X. et al., 2010; Bi et al., 2011; Liu et al., 2011; Zhang et al., 2012a; Qi et al., 2013; Alam et al., 2014; Gong et al., 2014), Africa (Diner et al., 2001; Eck et al., 2003; Kim et al., 2011; Queface et al., 2011), and South America (Zhang Y. et al., 2012; Rosário et al., 2013). Among these studies, the AODs show different values depending on the local conditions, e.g., the distance between aerosol sources, characteristics of land surface, and meteorological conditions. The retrieval of AOD is relatively reliable by far. However, big discrepancy on the absorption property of aerosols is presented, even for the same region. The basic reason for the difference in the absorption property may stem from the huge uncertainty on the single scattering albedo retrieval.

The SSA, which reflects the scattering and absorbing properties of aerosols, is related to the mixing of different chemical species in atmospheric aerosols. This involves the same type of particles (internal mixing) or different types (external mixing). The mixing state has little effect on scattering (Chylek and Wong, 1995; McMurry et al., 1996); however, it can have a great effect on the absorption efficiency. For example, the absorption of a mixture of BC and transparent particles is significantly higher with an internal rather than an external mixture, resulting in a smaller SSA (Jacobson, 2000, 2001). As seen in the BBC_AGCM model simulation (Fig. 2b), the highest SSA values were observed over Europe, reflecting the high sulfate loading, and the lowest values were over East Asia, South Asia, and South America, where carbonaceous aerosols are the main component. Determining aerosol absorption properties is crucial for reducing the uncertainties associated with aerosol effects. The SSA values vary in different regions, and they are influenced by different aerosol components. As suggested by Xia et al. (2006), the characteristics of SSA are different in different urban locations, and therefore special urban aerosol models should be created and used with satellite remote sensing in different urban regions.

The trends in aerosol optical properties, such as AOD and AE, have also been investigated in various studies (e.g., Kishcha et al., 2007; Meij et al., 2010; Hu et al., 2011; Yoon et al., 2012; Dong et al., 2013). A method using total direct solar radiation to retrieve AOD has been proposed and used to analyze its long-term trends (e.g., Qiu and Yang, 2000; Luo et al., 2001; Wang and Shi, 2010). Atmospheric aerosols are influenced to some degree by the development of local economies and production techniques, and they, in turn, affect local economic development and environmental conditions. Variations in atmospheric aerosols are also partially induced by changes in atmospheric circulation. Studies of AOD trends may also identify the reasons for global dimming and brightening.

Despite the large number of studies, the optical properties of aerosols are still far from being understood (Sokolik and Toon, 1996), especially the absorption properties. With the increase in human activities, the components of aerosols over different regions have become more complex. In regions influenced to a different extent by human activities, the components of aerosols and their optical properties are remarkably

different, which influence the climate in diverse ways.

3. Radiative properties of aerosols

Aerosols may influence the radiation budget of the earth-climate system. Three types of radiative effects have been postulated: direct, indirect, and semi-direct; they are reviewed in Sections 3.1, 3.2, and 3.3, respectively.

3.1 Direct effect of aerosols

The growing awareness of the potential climate impact of aerosols has resulted in a large research effort that has significantly improved our understanding of the role of aerosols in the earth's radiation balance. With a full set of aerosol optical properties available, the direct radiative effect (DRE) of aerosols can be calculated by using satellite-based (Huang et al., 2006c, 2010; Deng et al., 2010), model-based (Takemura et al., 2003, 2005; Wang et al., 2004; Gu et al., 2006; Wang Hong et al., 2010; Zhang et al., 2009, 2010; Wu and Han, 2011; Rap et al., 2013), satellite-model integrated (Yu et al., 2006; Huang et al., 2009), or ground-model integrated (Ge et al., 2010; Liu et al., 2011; Wang W. C. et al., 2013) approaches. Table 2 lists recent estimates of the aerosol direct solar effect and direct climate forcing (DCF). A brief description of each is given in the table, and readers are encouraged to refer to the relevant reference for more details.

Aerosols are derived from a range of sources, both natural and anthropogenic. Natural sources include volcanic emissions, plant vapors, and chemicals released by sea creatures. Anthropogenic aerosols are composed of a variety of aerosol types and components including water-soluble inorganic species (e.g., sulfate, nitrate, and ammonium), condensed organic species, elemental or black carbon, and mineral dust (Penner et al., 1994).

As listed in Table 2, Yu et al. (2006) suggested that the annual mean DRE of natural and anthropogenic aerosols over land at the TOA and surface (SRF) could reach –5.5 and –13.5 W m^{-2}, respectively (based on the integration of MODIS and Goddard Chemistry Aerosol Radiation and Transport (GOCART) databases). The annual mean DRE of natural and anthropogenic aerosols over ocean at the TOA and SRF could reach –6.5 and –11.1 W m^{-2}, respectively (based on the integration of MISR and GOCART databases). Given that the radiative forcing in the atmosphere is the difference between the values at the TOA and those at the SRF (e.g., Liu et al., 2011), the annual mean heating of natural and anthropogenic aerosols in the atmosphere over land can be as high as 8.0 W m^{-2}, which is much higher than that over ocean (4.6 W m^{-2}).

Since the early 1970s, anthropogenic aerosols have been assumed to offset a large portion of the warming resulting from anthropogenic greenhouse gases (GHGs). In the early 1980s, studies based on numerical models estimated that anthropogenic aerosols partially offset more than half of greenhouse warming and this dominated the uncertainty associated with the anthropogenic driving of climate change. As reported by the IPCC, despite the large uncertainty ranges regarding aerosol forcing, there is a high confidence that aerosols have offset a substantial portion of the forcing due to GHGs (Stocker et al., 2013). The best estimate of anthropogenic DCF is –0.35 [–0.85 to +0.15] W m^{-2} (high confidence) according to the evidence from aerosol models with some constraints from observations (Stocker et al., 2013). Using SPRINT-ARS, a radiation-transport model for aerosols, Takemura (2012) simulated the distribution of the adjusted forcing due to aerosol direct effect at the SRF in 2000 (Fig. 3), in which the adjusted forcing was defined as a change in the net irradiance after allowing the atmospheric and land temperature, water vapor, clouds, and land albedo to adjust to the prescribed sea surface temperatures and sea ice cover. The adjusted forcing due to the aerosol direct effect at the SRF is currently negative over most regions, with the maximum negative adjusted forcing over Asia being greater than –5 W m^{-2}. The negative effect due to aerosols at the SRF suggests the influence of some absorbing aerosols (e.g., BC) on solar and thermal radiation.

Although the net effect of anthropogenic aerosols is cooling (negative forcing) (e.g., Chen et al., 2011; Liu and Zhang, 2012), some components may contri-

Table 2. Summary of seasonal and annual (ANN) average clear-sky DRE or DCF (W m^{-2}) at the top of the atmosphere (TOA) and surface (SRF) over the globe derived with different methods and data

Product	Region	Time	Aerosol	TOA	SRF	Reference
			DRE			
POLDER	Ocean	DJF	NAT+ANT	–5.7	—	
		MAM		–5.7	—	(Yu et al., 2006)
		JJA		–5.8	—	
		SON		–5.6	—	
		ANN		–5.7	–7.7	
GMI	Land	ANN	NAT+ANT	–8.6	—	(Bian et al., 2009)
	Ocean			–4.5	—	
	Global			–5.7	—	
MISR_MODIS _CERES	Land	ANN	NAT+ANT	–5.1	—	(Patadia et al., 2008)
			DCF			
HadGEM2–A	Land	ANN	ANT (only in shortwave	–0.91	—	(Bellouin et al., 2008)
	Ocean		spectrum)	–0.51	—	
	Global			–0.63	—	
MODIS_A	Global	ANN	ANT	–1.9	–4.4	(Bellouin et al., 2005)
GISS II-prime	Global	ANN	ANT sulfate	–0.95	—	(Adams et al., 2001)
			ANT nitrate	–0.19	—	
GISS II-prime	NH	ANN	ANT BC	0.52	–0.81	(Chung and Seinfeld, 2005)
	SH		(external mixed)	0.15	–0.28	
	Global			0.33	–0.55	
	NH	ANN	ANT BC	0.93	–1.00	
	SH		(internal mixed)	0.28	–0.34	
	Global			0.60	–0.67	
GISS II-prime	Global	ANN	ANT sulfate	–1.02	—	(Koch, 2001)
			ANT OC	–0.44	—	
			ANT BC	0.19	—	
GISS	Global	DJF	Dust (NH Africa/Arabia	–0.22	–0.75	(Miller et al., 2004)
		MAM	source region)	–0.15	–1.18	
		JJA		–0.04	–1.08	
		SON		–0.06	–0.38	
		ANN		–0.12	–0.85	

GMI: Global Modeling Initiative; CERES: Clouds and the Earth's Radiant Energy System; HadGEM2-A: Hadley Global Environment Model 2-Atmosphere; GISS: Goddard Institute for Space Studies; DJF: December-January-February; MAM: March-April-May; JJA: June-July-August; SON: September-October-November; NAT: natural aerosols; and ANT: anthropogenic aerosols.

bute a warming effect (positive forcing). Jacobson (2000) showed high positive forcing from BC, with the magnitude exceeding that of forcing due to CH4, suggesting that BC may be the second most important contributor to global warming after CO_2 in terms of direct forcing. Chung and Seinfeld (2005) showed (see Table 2) that different mixtures of anthropogenic BC could generate different DCFs, with the annual mean DCFs of externally mixed anthropogenic BC at the TOA over the NH, SH, and the whole global being positive and reaching 0.52, 0.15, and 0.33 W m^{-2}, respectively. Internally mixed anthropogenic BC could induce greater warming (with annual DCF values at the TOA over the NH, SH, and the whole globe being 0.93, 0.28, and 0.60 W m^{-2}, respectively). As noted in Section 2, the absorption property of internally mixed BC is significantly higher than that of externally mixed BC, resulting in lower SSA (Jacobson, 2000, 2001). The strong warming effect of BC derives from its strong absorption property. Although heating effects are postulated, the warming effects of carbonaceous aerosols are still within a wide range (e.g., Zhang et al., 2009; Tian et al., 2013), which contributes to the uncertainties regarding the overall effects of aerosols.

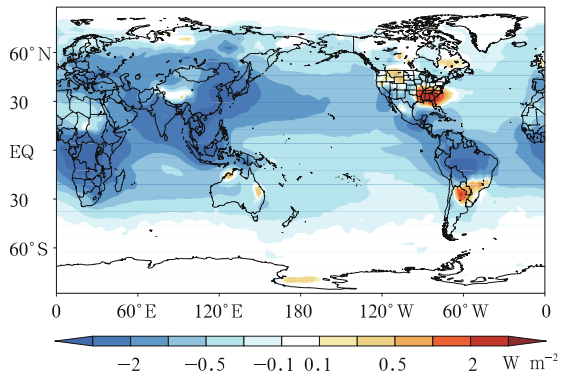

Fig. 3. Global distribution of adjusted forcing (W m^{-2}) due to the aerosol direct effect at the surface in 2000 (averaged for the period 1998–2002), as simulated by SPRINT-ARS (Takemura, 2012).

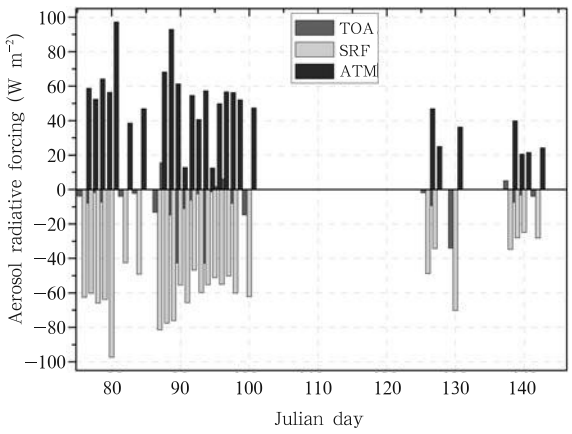

Fig. 4. Daily mean direct radiative forcing due to aerosols at the surface (SRF), top of the atmosphere (TOA), and in the atmosphere (ATM) in spring 2009 over Northwest China (Liu et al., 2011).

Mineral dust, another absorbing aerosol, can have a significant effect on the earth's radiation budget, because it can both scatter sunlight back to space (leading to negative radiative forcing) and absorb solar and infrared radiation (leading to positive forcing) (Sokolik and Toon, 1996). It has been suggested that the DCF due to mineral dust may be comparable to the forcing by other anthropogenic aerosols, and on a regional scale, it can greatly exceed that due to sulfate. Dust aerosol may exert a negative (positive) DCF at the SRF (TOA) by reducing the amount of incoming solar radiation reaching the ground and locally heating the atmospheric column (Islam and Almazroui, 2012). As reported by Highwood et al. (2003), the effect on irradiance due to a dust outbreak over the Sahara was a decrease of 6.5 W m^{-2} at the TOA and an increase of 11.5 W m^{-2} at the surface. Over the Taklimakan Desert, Huang et al. (2009) found that the average daily mean net radiative effect of atmospheric dust was 44.4, 41.9, and 86.3 W m^{-2}, respectively, at the TOA, SRF, and in the atmosphere. As also shown in Fig. 4, over the Loess Plateau near the Taklimakan Desert in Northwest China, the average shortwave DRE of dust for spring 2009 was 0.68, –70.02, and 70.70 W m^{-2}, respectively, at the TOA, SRF, and in the atmosphere. The dust aerosols heat the atmosphere during dust events by about 2 K day^{-1} (daily average), which is 60% larger than the heating (1.25 K day^{-1}) induced by background aerosols (Liu et al., 2011). With the outbreak of dust events in Northwest China, especially in early spring, the mixture of local pollution (especially carbonaceous aerosols from heating) could strengthen the absorption properties of dust aerosols. The uncertainties in the DRE due to dust aerosols (e.g., Su et al., 2011; Ansell et al., 2014) are mainly due to inaccuracies in measurement of dust aerosol optical properties, the SSA.

Additionally, over much of the world, biogenic organic aerosols have been shown to dominate the mass of fine aerosols (Zhang et al., 2007; Jimenez et al., 2009). The properties of biogenic secondary organic aerosols have also been the focus of recent studies (e.g., Mentel et al., 2013; Han et al., 2014). The presence of biogenic organic aerosols can affect the earth's radiative balance and cloud properties. As Scott et al. (2014) suggested, the annual global mean DRE due to biogenic secondary organic aerosols is between –0.08 and –0.78 W m^{-2}. However, the properties of biogenic secondary organic aerosols are still not well understood.

Despite many advances in understanding the direct effect of aerosols, as summarized Table 2, large uncertainties remain regarding several key variables such as surface albedo, aerosol particle size, vertical distribution, optical depth, and the imaginary part of the refractive index (Liao and Seinfeld, 1998).

3.2 Aerosol-cloud interactions

Clouds themselves are important regulators of the earth's radiation budget. Overall, clouds cool the earth-atmosphere system at the TOA. Losses of 48 W m^{-2} at TOA in the solar spectrum by clouds are only partially compensated for 30 W m^{-2} by cloud trapped infrared radiation. Small changes to macrophysical (coverage, structure, and altitude) and microphysical properties (droplet size and phase) of clouds have significant effects on climate. For example, a 5% increase in shortwave cloud forcing would compensate for the increase in greenhouse gases during 1750–2000 (Ramaswamy et al., 2001).

Aerosol particles serve as condensation nuclei for the formation of cloud droplets and atmospheric ice particles. Recent advances in this field have revealed a much more complex interaction of aerosols with clouds (Rosenfeld et al., 2014). Aerosols affect cloud properties in various ways (see Fig. 5). Smaller cloud droplets, which are polluted by aerosols, when there is a constant cloud water path, evaporate faster and cause more mixing of ambient air into the cloud top, which could further enhance the evaporation of cloud droplets (process A in Fig. 5). These small cloud droplets polluted by aerosols could induce an increase in cloud albedo, causing a huge cooling effect through the suppression of precipitation and extending of the cloud lifetime (process B in Fig. 5). Additionally, the aerosols may transport large quantities of small ice particles to the anvils of deep convective clouds, leading to a warming effect through reduction of the thermal radiation emitted to space (process C in Fig. 5). However, the contributions of individual processes to the overall aerosol-cloud interaction cannot be easily separated.

Aerosol-cloud interaction includes the indirect radiative effect (IRE) and the semi-direct radiative effect. The IRE consists of two components: the albedo effect (Twomey effect or the first indirect effect) and the cloud lifetime effect (the second indirect effect). Each affects the size distribution and chemical nature of atmospheric aerosols as well as the chemical composition of clouds and precipitation.

As shown in Table 3, almost all IRE values are negative. The global annual IRE of anthropogenic aerosols at the TOA and SRF could reach –2.9 (Menon et al., 2002a) and –1.8 W m^{-2} (Lohmann and Feichter, 2001), respectively. The annual IRE of anthropogenic aerosols over land and ocean at the TOA could reach –4.9 (Menon et al., 2002a) and –2.2 W m^{-2} (Rotstayn, 1999), respectively. Shen et al. (2011) modeled the monthly average IRE at the tropopause due to dust aerosols over China, and produced values of –1.26, –2.0, and –2.69 W m^{-2} in March, April, and May, respectively. In some regions, a value of –7 W m^{-2} was simulated. Su et al. (2008) postulated that the combination of indirect and semi-direct shortwave radiative

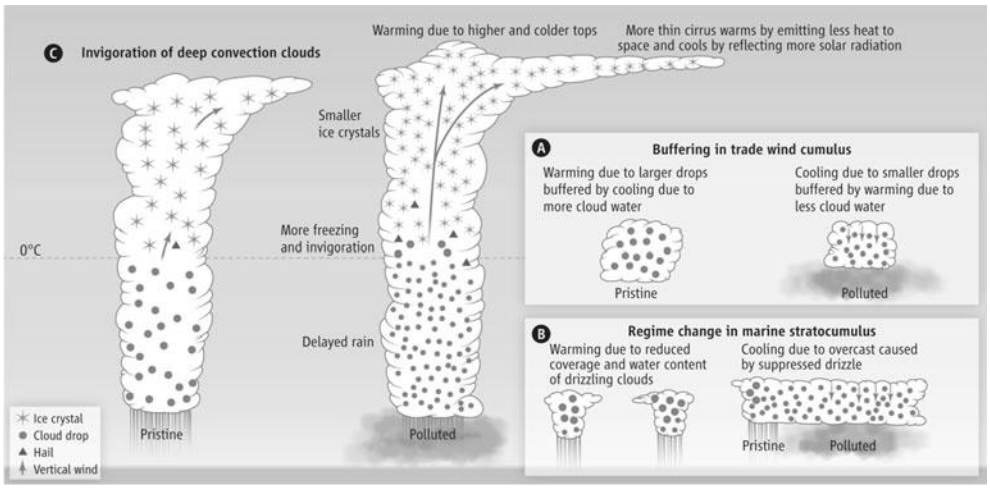

Fig. 5. Schematic diagram of the interaction of aerosols with clouds (Rosenfeld et al., 2014).

Table 3. Summary of the aerosol-cloud indirect radiative effect IRE (W m^{-2})

Product	Region	Time	Aerosol	Layer	1st	2nd	Total	Reference
MIRAGE	Global	ANN	Sulfate	TOA	—	—	–2.1	(Ghan et al., 2001)
ECHAM4	Global	ANN	ANT	TOA	—	—	–1.4	(Lohmann and Feichter, 2001)
				SRF	—	—	–1.8	
GISS II	Land	ANN	ANT	TOA	–2.8	—	–4.9	(Menon et al., 2002b)
	Ocean	ANN	ANT	TOA	–1.3	—	–2.1	
	Global	ANN	ANT	TOA	–1.7	—	–2.9	
CAM-Oslo	Global	ANN	ANT	TOA	—	—	–1.83	(Quaas et al., 2009)
CAM-PNNL	Global	ANN	ANT	TOA	—	—	–1.87	
CAM-Umich	Global	ANN	ANT	TOA	—	—	–1.53	
LMDZ-INCA	Global	ANN	ANT	TOA	—	—	–0.27	
SPRINTARS	Global	ANN	ANT	TOA	—	—	–0.82	
MAM3	Global	ANN	ANT	TOA	—	—	–1.53	(Ghan et al., 2012)
MAM7	Global	ANN	ANT	TOA	—	—	–1.45	
CCM3	Land	ANN	Sulfate	TOA	—	—	–1.28	(Kiehl et al., 2000)
	Ocean	ANN	Sulfate	TOA	—	—	–0.79	
	Global	ANN	Sulfate	TOA	—	—	–0.92	
RegCCMS	China	Jan	Nitrate	TOA	—	—	–1.63	(Li et al., 2009)
				SRF	—	—	–1.61	
	China	Jul	Nitrate	TOA	—	—	–2.65	
				SRF	—	—	–2.65	
MODIS_Terra	Tropical	ANN	ANT	TOA	—	—	–1.38	(Matsui and
	Ocean							Pielke, 2006)
MODIS_CERES	Global	ANN	ANT	TOA	–0.2	—	—	(Quaas et al., 2008)
AVHRR_POLDER	Global	ANN	ANT	TOA	—	—	–0.6 to –1.2	(Sekiguchi et al., 2003)
HadAM4	Global	ANN	Sulfate	TOA	–1.34	–0.49	–1.89	(Jones et al., 2001)
CSIRO	Global	ANN	ANT	TOA	–1.46	–1.32	–2.57	(Rotstayn and Penner, 2001)
GFDL	Global	ANN			–1.5	–0.9	–2.3	
LMDZ	Global	ANN	ANT	TOA	—	—	–0.84	(Quaas et al., 2006)
ECHAM4					—	—	–1.54	

MIRAGE: Model for Integrated Research on Atmospheric Global Exchange; ECHAM4: European Centre/Hamburg Model; CAM: Community Atmosphere Model; PNNL: Pacific Northwest National Laboratory; LMDZ-INCA: Laboratoire de Météorologie Dynamique Zoom-Interaction with Chemistry and Aerosols; CAM-Oslo: The model based on the NCAR CAM3 with large-scale condensation and a diagnostic cloud cover scheme; CAM-PNNL: A development version of CAM; CAM-Umich: A coupled aerosol and atmospheric circulation model and consists of the NCAR CAM3 and the LLNL/Umich IMPACT aerosol model; MAM3/MAM7: Modal Aerosol Model; RegCCMS: Regional Climate Chemistry Modeling System; CSIRO: Commonwealth Scientific and Industrial Research Organization; GFDL: Geophysical Fluid Dynamics Laboratory.

forcing for Asian dust was 82.2 W m^{-2}, 78.4% of the total dust effect. Huang et al. (2006a) suggested that during the growth of clouds in the presence of dust over East Asia, the instantaneous TOA net radiative forcing for the no-dust region was about –208.2 W m^{-2}, 42.1% larger than the value in cloud-over-dust regions. The presence of dust aerosols under clouds could significantly reduce the cooling effect of clouds.

Despite considerable advances in our understanding of the interactions of aerosols with clouds, further progress has been hampered by limited observational capabilities and coarse-resolution climate models. Differences in the cloud microphysics scheme, especially in the auto conversion rate, have led to uncertainties in estimates of the indirect aerosol effect (Lohmann and Feichter, 1997; Jones et al., 2001; Menon et al., 2002a). For the indirect effects due to aerosols, most studies have only considered the cloud albedo effect (e.g., Jones et al., 1994; Lohmann and Feichter, 1997). Even when both the cloud albedo effect and the cloud lifetime effect are considered, some discrepancy in their importance still exists. For example, Williams et al.

(2001) and Kristjánsson (2002) concluded that the albedo effect at the TOA was four times as important as the cloud lifetime effect, whereas Ghan et al. (2001) simulated a cloud lifetime effect that was larger than the albedo effect. In the following section, the albedo effect and cloud lifetime effect are discussed separately in review of the aerosol indirect effect. The semi-direct effect is not included in this section but is discussed separately in Section 3.3.

3.2.1 Albedo effect

Because solar radiation is mainly scattered but only minimally absorbed by cloud droplets, an increase in cloud condensation nuclei (CCN) at constant liquid water content leads to a large concentration of small-radius cloud droplets. This enhances cloud reflectivity, rendering the radiative forcing negative. This aspect of the aerosol indirect effect is referred to as the first indirect (or albedo) effect (Twomey, 1974; Twomey et al., 1984).

The Third Assessment Report (TAR) of the IPCC concluded that the first indirect albedo effect of anthropogenic aerosol particles amounts to between 0 and –2 W m^{-2} as a global mean (Ramaswamy et al., 2001). The IPCC later refined this range from –0.5 to –1.9 W m^{-2} (Lohmann and Feichter, 2005).

The cloud albedo depends on both the cloud droplet size and the cloud thickness. Many studies have investigated cloud droplet size (Brenguier et al., 2000, 2003; Schwartz et al., 2002) to study the impact of polluted clouds on albedo.

Most studies (e.g., Langner and Rodhe, 1991; Jones et al., 1994; Boucher and Lohmann, 1995) relate cloud droplet number concentration to the mass of sulfate aerosols, assuming that the dominant source of CCN is sulfate aerosols, and estimate the change in cloud albedo by calculating shortwave forcing with and without anthropogenic sulfur emissions. In the studies referred to above, the simulated indirect aerosol effect including the first indirect albedo effect ranges between –0.5 and –1.6 W m^{-2}. Lohmann and Feichter (1997) fully coupled a sulfur cycle module to a cloud microphysics scheme and estimated the albedo effect of sulfate aerosols. Le Treut et al. (1998) obtained the indirect forcing by considering the albedo effect only in the range of –0.4 to –1.6 W m^{-2}. Lohmann et al. (2000) found that the change in radiative cooling by the cloud albedo effect between present-day and pre-industrial conditions ranged from near zero to –0.4 W m^{-2} due to anthropogenic sulfate and –0.9 to –1.3 W m^{-2} due to anthropogenic carbonaceous aerosol.

Many studies have suggested that the cloud albedo effect contributes most to the indirect effect due to aerosols. For example, Lohmann and Feichter (1997) showed that 60% of the indirect forcing of –1.4 W m^{-2} was due to the cloud albedo effect. However, there are very large uncertainties in the albedo effect due to the presence of different types of aerosols in the atmosphere. Lohmann et al. (2000) and Chuang et al. (2002) reported that the impact of carbonaceous aerosols on cloud albedo was 3–6 times of that due to sulfate aerosols. Conversely, Ghan et al. (2001) suggested that the cloud albedo effect was dominated by sulfate. In addition to the model simulations, some studies have considered the impact of aerosols on cloud droplet size (e.g., Huang et al., 2006c).

In future studies of the albedo effect of aerosols, the issues of (i) albedo effects of aerosols on different cloud types, (ii) albedo effects of different aerosols, and (iii) a validation of the albedo effect between satellite analysis and modeling studies are key to understanding and reducing the uncertainties associated with the aerosol indirect effect.

3.2.2 Lifetime effect

The lifetime effect (Squires, 1958; Hudson and Frisbie, 1991) is an aerosol-cloud interaction mediated by precipitation. It is based on the hypothesis that changes in the atmospheric aerosol lead to changes in cloud droplet size, which then influence precipitation formation and the residence time of clouds. The distinguishing quality of the lifetime effect hypothesis is the idea that the macrostructure of the cloud (such as its spatial extension or liquid water content) is determined by the efficiency with which precipitation develops, which is in turn regulated (at least in part) by the aerosol. This differentiates the cloud lifetime effect from the cloud albedo effect (Twomey, 1977).

Two approaches have been taken to studies of the lifetime effect hypothesis (Albrecht, 1989; Stevens

and Feingold, 2009). The first considers statistical measurements of the lifetime effect, either in observations or by enforcing relationships among the aerosol, clouds, and precipitation in large-scale models. The second considers the effect to be hypothetical in order to test it or the various assumptions upon which it is based, e.g., through dedicated field or modeling studies that explore how individual clouds or fields of clouds respond to changes in the ambient aerosol.

In 1966, a study of the relationship between cloud nuclei produced by cane fires and the cloud droplet concentration was performed by using a cloud droplet sampler and thermal diffusion chamber installed in an aircraft (Warner and Twomey, 1967). It had been suggested that aerosols suppressed the precipitation in warm (ice-free) boundary layer clouds (Albrecht, 1989). Therefore, many studies were initiated to determine the cloud lifetime effect due to aerosols. Lohmann and Feichter (1997) included the cloud lifetime effect in their estimate of indirect forcing, and estimated that 40% of the forcing of –1.4 W m^{-2} was due to the cloud lifetime effect. Rotstayn (1999) suggested that the cloud lifetime effect of anthropogenic aerosols was –1.0 W m^{-2}. Le Treut et al. (1998) included the cloud lifetime effect of aerosols by regarding it as a feedback.

Recently, many studies have investigated the cloud lifetime effect with high-resolution models and satellite data analyses (e.g., Suzuki et al., 2008; Small et al., 2009; Wang et al., 2012). Although large uncertainties still remain in estimations of the cloud lifetime effect, it should not be ignored in future calculations of the indirect forcing due to aerosols.

3.3 Semi-direct effect (cloud evaporation)

The term "semi-direct effect" was postulated by Hansen et al. (1997) to describe the impact of absorbing aerosols on clouds, but relatively little attention was devoted to it until the INDOEX field campaign, which highlighted its importance (Ackerman et al., 2000). The absorption of solar radiation by aerosols leads to a heating effect in the atmosphere, which can result in the evaporation of cloud droplets. Such heating can partially offset the cooling due to the aerosol indirect effect. Conversely, Penner et al. (2003) and Johnson et al. (2004) indicated that the semi-direct effect could result in a cooling depending on the location of the absorbing aerosols with respect to the clouds. Thus, there is a lack of consensus regarding the sign and magnitude of the global semi-direct effect (Morgan et al., 2006), as shown in Table 4, whereas estimates of the direct and indirect effects are relatively more consistent. However, reducing the uncertainties associated with the semi-direct effect is important, as some studies have found that the magnitude of the semi-direct effect could exceed the direct forcing (Johnson et al., 2004).

The semi-direct effect of absorbing aerosols has been studied not only in small regional cloud simulations but also in global models, because aerosol heating can drive large-scale motions that affect clouds (Ackerman et al., 2000; Allen and Sherwood, 2010). Lohmann and Feichter (2001) provided the first assessment in the global annual mean semi-direct forcing and revealed significant reductions of liquid water path in regions where the absorbing aerosols burden was high. Huang et al. (2006b) postulated that the cloud water path of dust-contaminated clouds was considerably smaller than that of dust-free clouds because of the semi-direct effect of dust aerosols over East Asia (Fig. 6). Their results indicated that the mean ice water path (IWP) and liquid water path (LWP) of dusty clouds were less than those of their dust-free counterparts by 23.7% and 49.8%, respectively. Sakaeda et al. (2011) indicated that the total aerosol radiative effect of biomass burning aerosols over the southern African/Atlantic region at the TOA was significantly altered by inclusion of the semi-direct effect. Their results also indicated that the semi-direct effect was primarily driven by changes in cloud cover and to a lesser extent by changes in liquid water path over the ocean and land (Sakaeda et al., 2011).

When considering the effect of cloud evaporation, a 0.17-K warming in the global annual mean surface temperature is expected due to the reduction in low clouds, whereas the warming without cloud feedback is –0.41 K (Hansen et al., 1997). Although many studies (e.g., Chýlek et al., 1996; Hansen et al., 1997; Cook

Table 4. Summary of the semi-direct radiative forcing (W m^{-2})

Product	Region	Time	Aerosol	TOA	SRF	Reference
MAM3	Global	ANN	ANT	0.08	—	(Ghan et al., 2012)
MAM7				–0.09	—	
OMI_CERES_ AMSR-E_MODIS	Subtropical Southeast Atlantic Ocean	Winter	Smoke	–0.7	—	(Wilcox, 2012)
INDOEX	Indian Ocean	Feb to Mar	ANT	3.7	–2.8	(Koch and Del Genio, 2010)
CAM3	Land	JJA	ANT	0.99	1.77	(Allen and Sherwood, 2010)
		ANN		0.43	1.04	
	Sea	JJA		–0.08	0.41	
		ANN		–0.03	0.35	
GISS_MATRIX	Global	ANN	ANT	–0.1	—	(Bauer and Menon, 2012)
ECHAM5-HAM	Global	ANN	H_2SO_4/H_2O nucleation	–0.24	—	(Kazil et al., 2010)
COSMO-MUSCAT	Europe	19–26 July 2006	Absorbing aerosol	2.4	2.6	(Meier et al., 2012)
	Europe land			2.7	—	
	Europe ocean			2.3	—	
RegCCMS	China	Jan	BC	0.04	–0.05	(Zhuang et al., 2010)
		Apr		0.10	–0.14	
		Jul		0.09	–0.12	
		Oct		0.06	–0.08	
CanAM4	Global	DJF	BC (online)	0.79	—	(Li et al., 2013)
		JJA		1.92	—	
		ANN		1.32	—	

COSMO-MUSCAT: Model of the Consortium for Small Scale Modeling and Multi-Scale Atmospheric Transport Model; CanAM: Canadian Global Climate Mode; OMI: Ozone Monitoring Instrument.

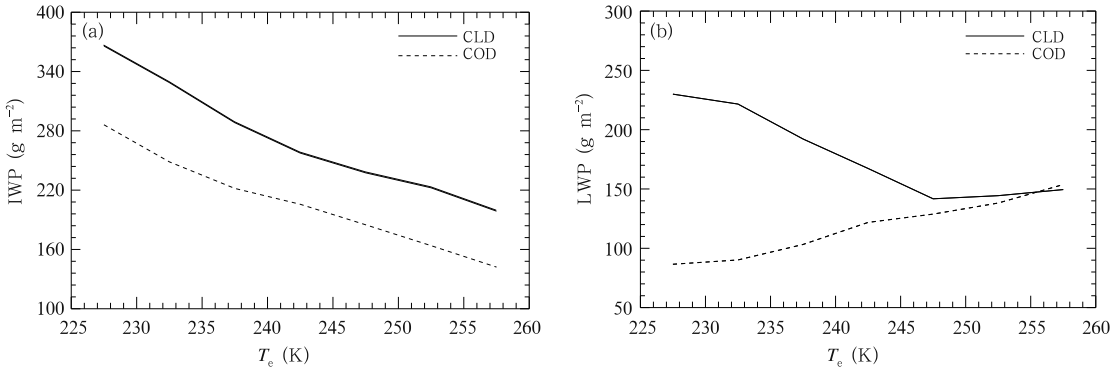

Fig. 6. Comparison of the cloud water path over the pure cloud (CLD) region and the cloud over dust (COD) region as a function of effective cloud top temperature (T_e) for the (a) ice water path (IWP) and (b) liquid water path (LWP) (Huang et al., 2006b).

and Highwood, 2004; Johnson et al., 2004; Huang et al., 2006b, c) have identified a positive semi-direct effect, negative semi-direct forcing results have also been reported (e.g., Penner et al., 2003).

For the absorbing aerosols, the semi-direct effect is an important aspect to consider alongside the direct effects. The semi-direct effects of aerosols in different regions are significantly different, and their signs

mainly depend on the vertical profile of aerosols. In different regions, the impact of aerosols on precipitation through the semi-direct effect varies, which is important for future studies of the hydrological cycle on both global and regional scales.

4. Summary and discussion

In this review, we have summarized the major progress so far achieved in studies of aerosol optical properties and radiative effects. Main conclusions are presented in the following two subsections, and important issues for future studies are highlighted respectively.

4.1 Optical properties

Aerosol optical properties have been studied through ground-based observation, satellite remote sensing, and numerical modeling. These approaches can also be used to validate each other. With ground- and satellite-based observations and model simulations, data assimilation is expected to reduce the uncertainties regarding aerosol optical properties. However, large uncertainties still remain, especially concerning the absorption properties of different aerosols in different regions. Some important considerations include:

(1) For in-situ measurement of aerosols, the representativeness of the extent of the observed optical properties at each station is an important issue.

(2) When using satellite data, validation with ground-based observational data is required.

(3) Despite significant improvements in cloud screening from satellite retrievals, it remains difficult to observe aerosols when they are near or inside clouds. Alternative methods, such as the use of an aerosol proxy and the use of aerosol models, have been developed to compensate for this observational weakness.

(4) Determining aerosol absorption properties when the aerosols are polluted by absorbing aerosols, such as BC, is important to reduce the uncertainties associated with aerosol radiative effects.

4.2 Radiative effects

The aerosol radiative effects can be divided into the direct effect on the radiative balance and the indirect effect through interaction with clouds. Aerosol radiative effects can be further divided into those that exert a positive perturbation on the radiation budget and those that exert a negative perturbation.

Many studies have attempted to calculate the direct effect due to aerosols. Although the net direct effect of anthropogenic aerosols is cooling (negative forcing), some components may contribute a warming effect (positive forcing). For simulation of the aerosol direct effect, the uncertainties in the type of aerosols and their precursors, parameterizations of a variety of sub-grid aerosol processes (e.g., wet, dry, and gravitational deposition, cloud convection), and assumptions of aerosol size, absorption, mixture, and humidification of particles, should be further reduced.

Some studies have attempted to determine the indirect effect due to aerosols, whereas some have ignored the cloud lifetime effect (e.g., Jones et al., 1994; Lohmann and Feichter, 1997). A large number of studies have indicated that both the albedo effect and cloud lifetime effect act to cool the earth-atmosphere system by increasing cloud optical depth and cloud cover, respectively.

Some important considerations for future studies include:

(1) Determination of the cloud lifetime effect and the aerosol effect on mixed-phase and ice clouds is currently insufficient and should not be neglected when attempting to calculate the total radiative effect.

(2) When assessing the aerosol indirect and semi-direct effects from data, the shift in emissions of aerosols and their precursors may be a critical problem. In the future, due to climatic and environmental changes across the globe, determining the main emission sources will be crucial for predicting the aerosol indirect and semi-direct effects.

(3) To fully understand the interaction of aerosols with clouds, better observational capabilities and high-resolution climate models are needed.

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