AEROSOLS AND CLIMATE: A PERSPECTIVE OVER EAST ASIA

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Aerosol is becoming a central theme in the climate research arena, due to many new findings concerning their significant direct and indirect effects on climate (e.g. by altering temperature, cloud, radiation and precipitation) and to the large uncertainties in our estimates of aerosol forcing on climate. Despite the large loading and complex properties of East Asian aerosols, our knowledge of these aerosols and their climatic effects is so meager that they arguably present the last frontier in aerosol and climate research. While their climate effects are notably strong, the magnitude and mechanisms of their influence are far from being clear. Findings concerning how aerosols interact with energy and water cycles in other regions of cleaner environment may not be valid here. More attention needs to be focused on Asian aerosols in order to examine the existing aerosol-climate paradigms and to explore new ones. This paper provides an overview of Chinese aerosols in terms of their physical, chemical and optical properties and their potential impact on regional climate. Both anthropogenic and natural aerosols are addressed. General discussions concerning aerosol observation methods, research tools and approaches are also given. Findings by Chinese scientists are also reviewed to provide the state-of-the-art of Chinese aerosol research.

1. Introduction

While aerosols have been studied for a long time, it was not until the early 1990s that the role of aerosols in climate was widely recognized. Aerosols were identified as a central missing component in most, if not all, general circulation models (GCMs) that simulated climate changes at odds with observations in terms of long-term trends and spatial distributions (Hansen et al. 1997). After introducing sulfate aerosols estimated from industrial emissions (e.g. Langner and Rodhe 1991), model-simulated climate changes in response to the buildup of greenhouse gases became more realistic (Kiehl and Rodhe 1995; Mitchell et al. 1995). Investigations of a wide-range of aerosol effects ensued and many breakthrough findings were reported in the following years, referred to as the "exploratory phase" of aerosol research (Kaufman et al. 2002a). Major findings include the contribution of aerosols to the suppression of precipitation and the slowdown of hydrological cycles by dust storms (Rosenfeld et al. 2001; Ramanathan et al. 2001); to air pollution (Rosenfeld 2000; Rosenfeld and Woodley 2001), and to fire smoke plumes (Kaufman and Fraser 1997; Rosenfeld 2000). Other important findings include a larger reduction of the solar radiation budget at the surface than at the top of the atmosphere due to absorbing aerosols (Li 1998; Satheesh and

Ramanathan 2000; Li and Trishchenko 2001) and strong radiative heating in the atmosphere due to the mixing state of black carbon (Jacobson 2001). It must be borne in mind that these findings were drawn largely from a handful of cases. It remains an open question whether the findings are fortuitous. If so, it is critical to investigate under what circumstances the effects come into play. As we progress from the exploratory phase to quantitative phase in aerosol research, regional and global effects of aerosols must now be quantified. At present, the global averages of aerosol direct and indirect radiative forcing estimates are subject to very large uncertainties, as assessed by the Intergovernmental Panel on Climate Change (Penner et al. 2001).

While carbon dioxide (CO_2)-induced climate warming still dominates climate change at present, the sum of other anthropogenic forcings, especially ozone (O_3), methane (CH_4) and black carbon associated with air pollution, could exceed the effect of CO_2 over the next century (Hansen et al. 2000, 2002). Unlike the uniformly mixed CO_2, aerosol properties and effects exhibit considerable spatial and temporal variability (Fig. 1, left), of which we have a rather poor knowledge and understanding. This is especially the case in the developing world where many absorbing aerosols originate from air pollution, biomass burning, and dust storms (Streets et al. 2001; Lelieveld et al. 2001; Dickerson et al.2002).

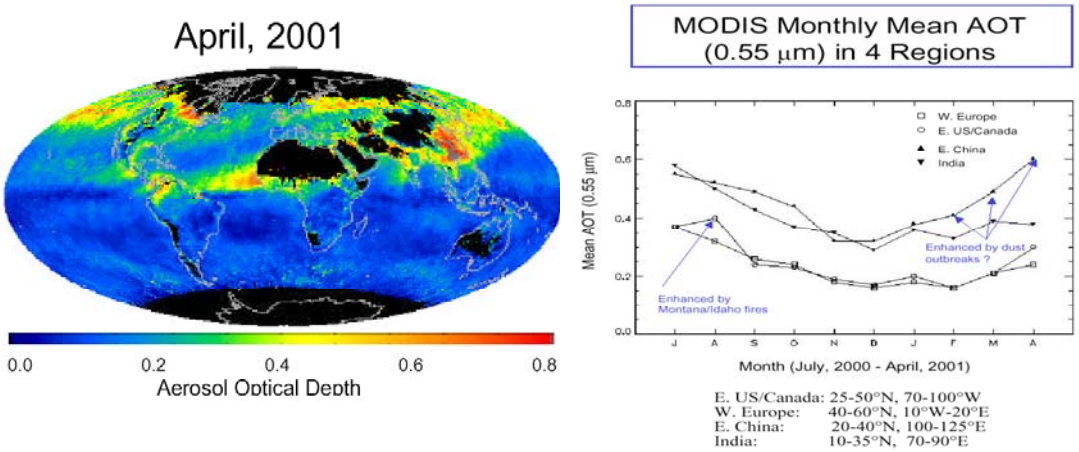

Figure 1. Left: Aerosol optical depth derived from MODIS showing total aerosol loading from all sources. Right: Comparison of mean aerosol optical depths derived over East China, India, West Europe and East United States and Canada from MODIS (Courtesy of Y. Kaufman and A. Chu).

Asia, with 60% of the world's population, is one of the heaviest aerosol-laden regions in the world (Fig. 1 left). At present, aerosol optical depth averaged over India and China is about twice the value in western Europe and eastern United States (Fig. 1 right). As this region is undergoing rapid economic development, increases in the emissions of aerosols and gases may continue for a foreseeable future, especially for black carbon aerosols, a by-product of energy consumption. Demands for energy in Asia are projected to increase rapidly in the future. Coal will remain the dominant source of energy. Dust storms, which are

particularly prevalent in northeast Asia in the spring, are another significant source of aerosol. In recent years, dust storms in this region appear to have increased in severity (Husar et al. 2001). The physical properties and chemical composition of the aerosols in this region differ considerably from those found elsewhere. As a result, not only is the magnitude of aerosol forcing large, the mechanisms by which aerosols interact with energy and water cycles may differ from those identified in relatively clean environments elsewhere. Aerosol effects are not limited to climate. Severe air pollution in China may have significantly reduced crop yields (Chameides et al. 1999).

China, as a primary source of both natural and anthropogenic aerosols in eastern Asia, is drawing much scientific attention. A couple of international aerosol experiments (e.g. ACE/ Asia, TRACE-P) took place in this region. Chinese scientists have conducted many aerosol-related investigations (Zhang and Mao 2001) but few are known to the world since the majority of publications are written in the Chinese language.

This paper reviews aerosol and climate studies, primarily in the context of East Asian aerosols over China. The state of knowledge concerning both natural (mineral dust) and anthropogenic aerosols and major advances in understanding their climatic effects are presented, based on papers published in both English and Chinese. As a self-complete overview article of certain tutorial value to graduate students and junior scientists, necessary fundamental knowledge on aerosol observation and understanding their climate effects is included.

2. Aerosol Optical Properties and Direct and Indirect Forcing

2.1. *Basic Aerosol Optical Parameters*

Aerosol direct radiative effects can be determined by three basic optical parameters: the aerosol optical depth, the single-scattering albedo and the asymmetry factor. The indirect effect is much more complicated and cannot be fully defined by any set of aerosol parameters. It depends on both aerosol inherent physical and chemical properties and the atmospheric environment. However, aerosol particle size plays an essential role that is also more readily available. The most widely used aerosol size information is the effective radius and the Ångström coefficient.

Aerosol Optical Thickness

A fundamental aerosol optical property is the total column aerosol optical depth, or thickness. It is essentially the aerosol extinction coefficient (σ_e) integrated over a vertical path through the atmosphere from the top of the atmospheric column (TOA) to the ground and is a function of wavelength, λ:

$$\tau(\lambda) = \int_0^{\mathrm{TOA}} \sigma_e(\lambda, h)\, dh\,. \tag{1}$$

Single- Scattering Albedo

Aerosol absorption properties are usually characterized by aerosol single-scattering albedo, defined as the ratio between the particle scattering coefficient (σ_s) and the total extinction coefficient (the sum of scattering and absorption (σ_a) extinction coefficients):

$$\omega(\lambda) = \frac{\sigma_s(\lambda)}{\sigma_a(\lambda) + \sigma_s(\lambda)}. \tag{2}$$

It is dependent on aerosol chemical composition and particle size distribution.

Phase Function, Asymmetry Parameter and Backscattering Fraction

The aerosol phase function describes the angular distribution of the scattered photons and is given as a function of the angle between the incident radiation and the scattering angle (θ). The asymmetry parameter (g) measures the fraction of forward scattering defined by:

$$g(\lambda) = \frac{1}{2}\int_0^{\pi} p(\lambda,\theta)\cos\theta \sin\theta d\theta . \tag{3}$$

It is equal to zero when the scattering is isotropic or symmetric about a scattering angle of 90°. The asymmetry parameter is equal to 1 when the scattering is confined to the forward direction (θ = 0°) and equal to –1 when the scattering is completely in the backward direction (θ = 180°). The backscattering fraction, or coefficient, is given by

$$\beta(\lambda) = \frac{1}{2}[1 - g(\lambda)]. \tag{4}$$

Effective Radius

The effective radius (r_e) is the area-weighted average radius of an aerosol particle size distribution or, equivalently, the ratio of the third to the second moments of the size distribution:

$$r_e = \frac{\int_0^{\infty} r^3 n(r) dr}{\int_0^{\infty} r^2 n(r) dr}. \tag{5}$$

Ångström Exponent

The Ångström wavelength exponent (α) is an approximate measure of aerosol particle size that can be estimated from aerosol optical depths measured at two wavelengths:

$$\alpha = -\frac{\Delta \ln \tau}{\Delta \ln \lambda}, \tag{6}$$

where Δ refers to the difference between measurements in two wavelength narrowbands. It can be linked to the exponent in an inverse power law, such as the Junge size distribution: $n(r) \sim r^{-4}$.

2.2. *Aerosol Direct Radiative Forcing*

Direct aerosol radiative forcing refers to the difference in the radiation budget with and without the presence of aerosol. While the majority of studies on aerosol direct forcing have been focused on the TOA, increasing attention is now directed toward surface aerosol forcing. Surface forcing is exerted by both scattering and/or absorption due to aerosol particles. Scattering increases the fraction of sunlight reflected to space and consequently reduces the fraction of sunlight reaching the surface. Absorption reduces both TOA reflection and the surface illumination. For conservatively scattering aerosols, the forcing at the TOA is identical to that at the surface. However, for absorbing aerosols, the former is less than the latter, due to the offsetting effects of scattering and absorption (Li and Trishchenko 2001). Because of the offsetting effects, aerosol direct forcing at the TOA may be positive (warming) or negative (cooling), depending on the aerosol single-scattering albedo, backscattering fraction, and surface albedo (R_s), as governed by the following threshold for cooling (Chylek and Coakley 1974):

$$\omega > \frac{2R_s}{\beta(1-R_s^{\,2}) + 2R_s}. \tag{7}$$

The dependence on surface albedo may be understood by comparing dark (e.g. ocean) and bright (e.g. snow) surfaces. Over the dark surface, nearly all types of aerosols cool the earth system due to the overwhelming effect of aerosol scattering. Over the bright surface, aerosol absorption is enhanced by multiple reflections between the surface and the aerosol layer, where even a weakly absorbing aerosol may have a net warming effect. The magnitude of aerosol direct radiative forcing is determined by (Haywood and Shine 1995):

$$\Delta F = -\frac{1}{2} S_0 T^2 (1-C)[\omega\beta(1-R_s)^2 - 2(1-\omega)R_s]\tau, \tag{8}$$

where S_0 denotes the solar constant, T is the atmospheric transmittance, and C is the cloud fraction. Note that this equation only accounts for the aerosol effect under clear-sky conditions. Aerosol direct forcing for cloudy skies is usually very small, since the bulk of aerosols are situated below the cloud and cloud scattering outweighs by far aerosol scattering.

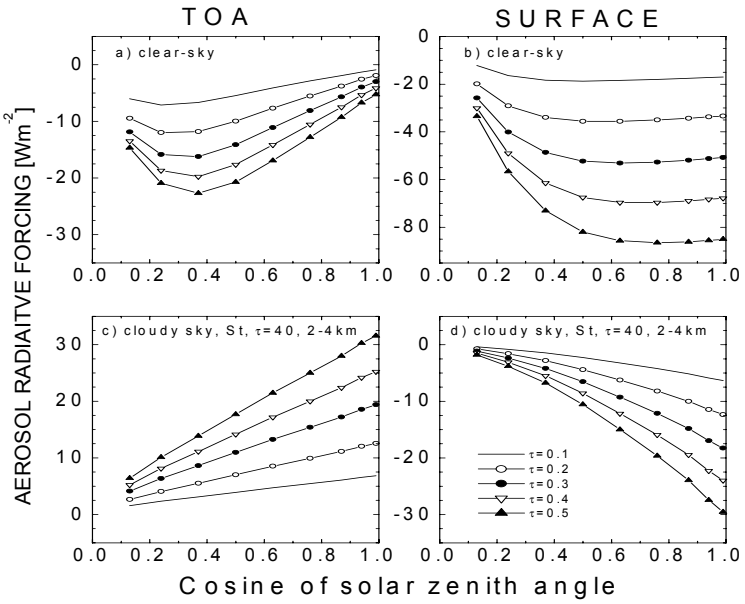

Figure 2. Model simulated aerosol radiative forcing under clear (a and b) and cloudy (c and d) skies at the top of the atmosphere (a and c) and at the surface (b and d) of variable aerosol loading (after Li and Trishchenko 2001).

However, if a significant amount of absorbing aerosol is imbedded inside a cloud layer, multiple scattering by the cloud droplets can substantially strengthen absorption, as demonstrated in Fig. 2. It shows direct aerosol radiative forcing at the top and bottom of the atmosphere for a continental type of model aerosol mixed with a stratus model cloud. For such moderately absorbing aerosols, TOA aerosol forcing under clear and cloudy conditions have opposite signs: cooling and warming for clear and cloudy skies, respectively. The forcing at the surface is always negative, but the magnitude of clear-sky aerosol forcing is much larger than cloudy-sky aerosol forcing. Under clear-sky conditions, surface aerosol forcing is substantially larger than TOA forcing, whereas the magnitude of the forcing is about the same at the surface and at the TOA but have opposite signs. These findings demonstrate the complexity of aerosol direct forcing (Li and Trishchenko 2001).

Observational estimation of aerosol direct radiative forcing is usually only feasible under clear-sky conditions for which the measurements of aerosol optical properties required to compute the forcing are available. Li and Kou (1998) made an attempt to derive aerosol direct radiative forcing inside an atmospheric column under all-sky conditions using coincident surface and satellite observations of solar radiation in the visible wavelengths (400 nm to 700 nm). At these wavelengths, absorption by atmospheric molecules and clouds is negligible so that solar radiation absorbed inside the atmosphere, as determined from satellite and ground observations, is attributed to aerosol forcing inside the atmospheric column. Note that heating by aerosols takes place primarily in the lower troposphere (0-4 km), at the expense of absorption by the surface (Satheesh and Ramanathan 2000). These effects can

have important ramifications for cloud dynamics and climate (Hansen et al. 1997; Ackerman et al. 2000).

2.3. *Indirect Aerosol Forcing*

Aerosol indirect forcing refers to the impact of aerosols on cloud and precipitation processes and is further classified into two categories known as the first and second type of aerosol indirect effect. The former is concerned with the enhancement of cloud reflection due to more but smaller cloud droplets formed upon hygroscopic aerosol particles that serve as cloud condensation nuclei (CCN). Given the increasing numbers of cloud droplets and a limited water supply, clouds tend to become less likely to precipitate. As a consequence, clouds last longer, thereby increasing the solar radiation reflected back to space. The effect of prolonged cloud lifetime (primarily for boundary layer clouds) is generally referred to as the indirect effect of the second kind. Many case studies demonstrated the first and second indirect aerosol effects focused on ship tracks over oceans (King et al. 1993, 1995; Coakley et al. 2000), cloud particle size differences over ocean and land (Han et al. 1994), and suppression of precipitation by air pollution and smoke plumes from fires (Rosenfeld 1999, 2000). However, the overall magnitude of the aerosol indirect effect on the global climate system remains uncertain.

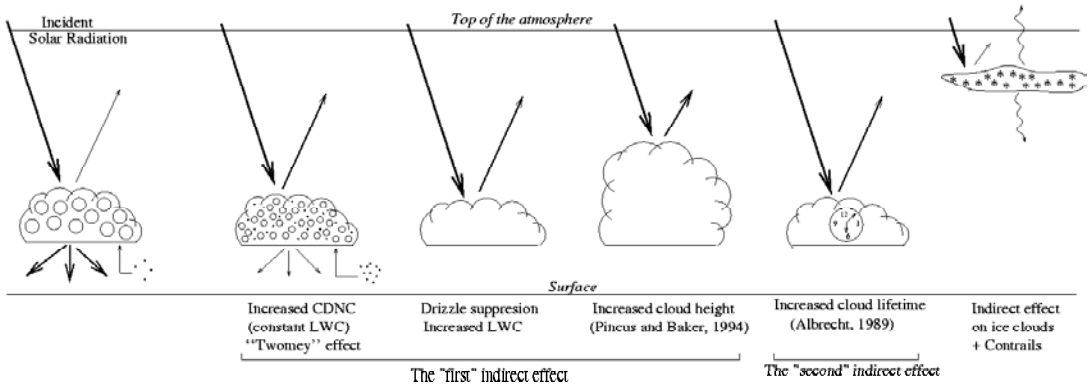

Figure 3. A schematic of aerosol indirect effects (courtesy of Jim Haywood).

The aerosol indirect effect may be determined by 1) taking *in situ* measurements of aerosol size-dependent chemistry, including organics and their dependence on hygroscopicity and CCN concentration; 2) analyzing aerosol data and cloud parameter statistics of the droplet size and cloud albedo as a function of cloud top temperature; and 3) analyzing rainfall data and relating them to cloud microphysics and CCN. Figure 3 is a schematic of four possible aerosol indirect effects: 1) increased CCN, reducing effective radius, 2) drizzle suppression, 3) increased cloud height and 4) increased cloud lifetime. An extensive review of aerosol direct and indirect effects was made by Haywood and Boucher (2000). Satellite retrievals of cloud and aerosol parameters by Nakajima et al. (2001) revealed positive and negative correlations between aerosol and τ and r_e, respectively.

3. Advanced Aerosol Observation Techniques

3.1. *Ground-based Observation*

The most successful ground-based aerosol observation network is the internationally federated AEROsol NETwork (AERONET) employing Cimel scanning sun photometers at multiple wavelengths (Holben et al. 1998, 2000; aeronet.gsfc.nasa.gov). At present, the AERONET includes about 160 instruments distributed with varying spatial density around the world, there are very few AERONET sites in East Asia and none permanent ones in China though. Using AERONET sun photometers, it is possible to characterize the microphysical and optical properties of aerosols. The AERONET system consists of a robotically controlled radiometer that makes direct sun measurements every 15 minutes and diffuse sky scans hourly in eight spectral bands during daylight hours. It has been demonstrated that AERONET data can be employed to characterize aerosol optical, radiative and microphysical properties from spectral direct and diffuse sky radiances (Dubovik and King 2000; Dubovik et al. 2002). The measured and retrieved parameters include aerosol optical thickness, Ångström exponent, particle size distribution (radii from 0.05 to 15 μm), single-scattering albedo, phase function, complex refractive index (n_r, n_i), and spectral flux. These data are valuable for validation of satellite-derived aerosol optical properties, while long-term observations are important in developing trends and climatologies of aerosol optical properties (Holben et al. 2001; Kaufman et al. 2001). Complementary hemispherical solar flux observations at selected AERONET stations allow assessment of direct forcing at the surface by aerosols. It is worth noting that *Qiu et al*. (1983) conducted a pilot study to use sky-scattered radiance to estimate aerosol particle size distribution.

Although aerosol properties derived from AERONET are not direct measurements, they are the most reliable aerosol data obtained so far over large areas on a long term basis. They are subject to uncertainties resulting from retrieval errors and inherent assumptions. *In situ* observations (chemical composition and size distribution, etc.) are useful to validate and interpret the retrievals. At present, we are still confronted with technical challenges in measuring aerosol properties, especially its absorbing properties. One of the most difficult yet most important quantities to measure is black carbon (BC). Inventories of BC are difficult to compile even under the best conditions. Recent analyses of data from the Indian Ocean and the United States (Chen et al. 2001; Dickerson et al. 2002) indicate that measurements of BC and carbon monoxide (much easier to measure) are highly correlated, but their ratio varies with the type of energy consumption. The dominant source of BC in South Asia remains a mystery. Using these data, Dickerson et al. (2002) found that using two different techniques resulted in BC emissions in India that differ by a factor of 2 to 3. Since China has a multitude of small coal-fired sources, a scenario that differs considerably from both the United States and India, a BC inventory must be developed specifically for China. The vertical profile of aerosol extinction can be monitored by a ground-based lidar.

3.2. *Satellite Remote Sensing*

Aerosol optical properties (primarily optical thickness) have been retrieved from several spaceborne sensors such as AVHRR, MODIS, MISR, SeaWIFS, POLDER, and TOMS. King et al. (1999) gave an extensive in-depth review of various aerosol remote sensing methods, while Kaufman et al. (2002a) presented a state-of-the-art review of new aerosol knowledge gained primarily from satellite observations. The essence of aerosol remote sensing is to decompose mixed signals emanating from the surface, atmospheric gases and aerosol particles. Satellite-measured reflectance is composed of contributions from all these atmospheric components and is expressed in the following equation:

$$R(\tau_a,\varpi_0;\mu,\mu_0,\phi)=R_{atm}(\tau_a,\varpi_0;\mu,\mu_0,\phi)+\frac{A_g}{1-A_g r_{atm}(\tau_a,\varpi_0)}\times t_{atm}(\tau_a,\varpi_0;\mu)t_{atm}(\tau_a,\varpi_0;\mu), \qquad (9)$$

where R_{atm} is atmospheric reflectance with a "black surface"; $r_{atm}(\tau_a,\varpi_0)$ is the atmospheric spherical albedo, and $t_{atm}(\tau_a,\varpi_0;\mu)$ is the total transmission. Note that each of these variables is a function of aerosol optical thickness and single-scattering albedo and is, implicitly, a function of aerosol size distribution. From this equation, we can demonstrate many issues associated with aerosol remote sensing.

The first step is to remove the presence of clouds because the equation is only valid for clear-sky conditions. Identification of clear scenes from satellite imagery for the purpose of aerosol remote sensing is a non-trivial task. Any cloud contamination can easily confuse the faint signal of aerosol, whereas excessive cloud screening may remove pixels of heavy aerosol loading. Because of this delicate situation, cloud identification schemes used for cloud studies are usually not adequate for aerosol investigations. The majority of cloud screening methods is threshold-based, while more complicated methods, such as the artificial neural networks (Li et al. 2001), may be applied for aerosol detection on a smaller scale or for a specific aerosol type.

Aerosol remote sensing is often an ill-posed problem for we have much more unknown variables than known variables. When retrieving aerosol optical depth, we need to know the single-scattering albedo. Unfortunately, the latter is most difficult to measure by any means. Recently, remote sensing of single-scattering albedo from satellites was proposed using sun-glint measurements that are most sensitive to aerosol absorption (Kaufman et al. 2002b). Given the mixture of reflection from the surface and scattering from aerosols, one may not be able to retrieve aerosol optical depth at all, even if the single-scattering albedo is known perfectly. This occurs when the single-scattering albedo is equal to or close to the threshold given by Eq. (7). In this case, the aerosol absorption and scattering effects are just about the same so that an increase in aerosol optical depth does not increase TOA radiance. Wong and Li (2002) demonstrated the importance of knowing the single-scattering albedo in the retrieval of smoke aerosol optical depth. In addition to aerosol optical properties, the location of aerosol layer is important to study its direct and indirect effects.

Another complication arises from surface albedo. The premise of remote sensing of aerosol properties is that there is a contrast in TOA reflectance between an aerosol-laden and an aerosol-free atmosphere. As the surface reflectance increases, the contrast decreases and the accuracy of the aerosol properties retrieved is reduced. As a result, the majority of aerosol remote sensing methods are only valid over oceans or dark land. Satellite measurements depend on viewing geometries, which introduces another factor that needs accounting for in aerosol retrievals, namely, the bidirectional reflectance distribution function (BRDF). The BRDF is particularly troublesome over land, since it varies with many land parameters such as the land cover type and vegetation condition (e.g. greenness) (Li et al. 1996). Extensive BRDF models are under development using ample data acquired by the Earth Observation System (EOS) sensors. However, the persistent presence of aerosols poses a serious problem in developing the surface BRDF and in retrieving aerosol properties, as the two factors work together.

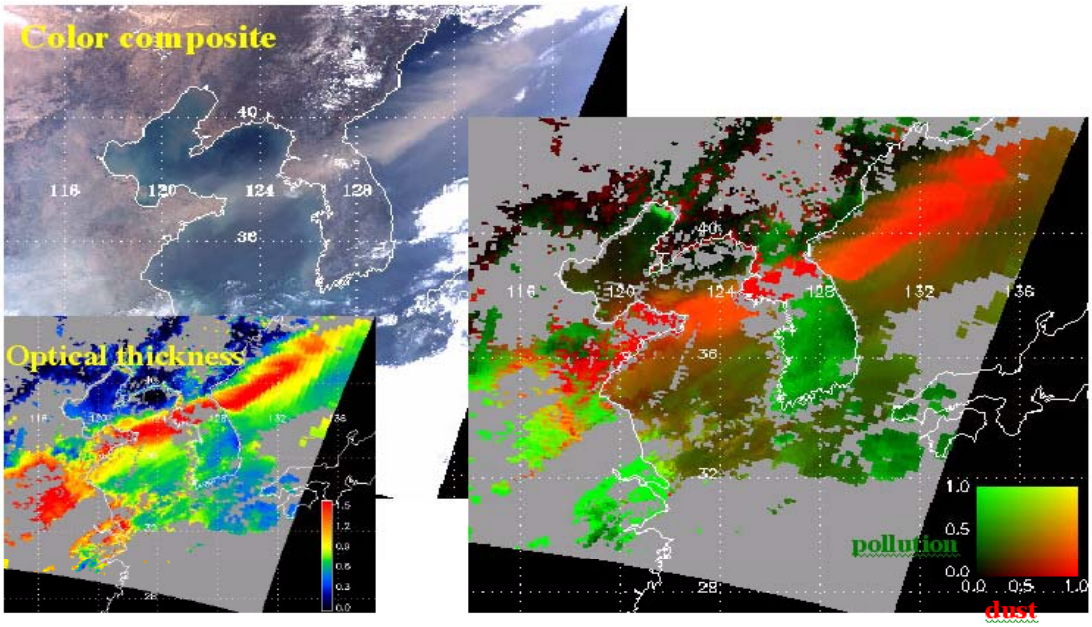

Figure 4. MODIS data from East Asia during a dust episode advected over a pollution layer in March 20, 2001. Top left is a color composite of the MODIS blue, green and red channels. Bottom left is the analysis of the aerosol optical thickness, and the right image is a separation of the fine particle pollution optical thickness (green) and coarse dust optical thickness (red) using the MODIS aerosol spectral signature. A two-dimensional color bar is shown on the bottom-right (after Kaufman et al. 2002a).

The state-of-the-art satellite sensors used for aerosol remote sensing can be classified into three categories: the multi-wavelength sensors (MODIS), the multi-angle sensors (MISR), and the polarization sensors (POLDER). The multi-wavelength sensors take measurements at several spectral channels, which is advantageous because aerosol effects differ from one wavelength to another. By virtue of these spectral differences, one can infer both aerosol size information and surface reflectance (even in the presence of aerosols). Given that aerosol

extinction decreases with wavelength, Kaufman et al. (1997) proposed using a longer-wavelength channel (transparent to aerosol) to estimate shorter-wavelength surface albedo from which aerosol optical thickness of fine particles can be estimated. Figure 4 illustrates the rich information that can be extracted from MODIS by virtue of its multispectral coverage. It detects two distinct aerosol types, a dust episode overlying a polluted layer on March 20, 2001 over eastern Asia. Both optical depth and size information are extracted. Aerosol retrievals for this type of sensor require well-characterized low reflectance, or dark, land surfaces.

The dark surface requirement is relaxed or even eliminated by the multi-angle and polarization sensors. The multi-angle sensors observe the same ground target from different viewing angles to separate the ground and atmospheric contributions based on their different directional reflectance behaviors. Since the retrieval relies on varying aerosol reflectance associated with changing atmospheric pathlength, dark targets are not absolutely required, and knowledge of ground surface reflectance is not needed for Lambertian surfaces or surfaces with a known BRDF. Polarization sensors measure the polarization of the incoming signal, which is sensitive to the refractive index of the aerosol particles, and not sensitive to the ground contribution. This allows an approximation of the aerosol type without *a priori* knowledge of surface reflectance. The POLDER algorithm, for example, is applicable to both ocean and land regardless of brightness, due to the negligible polarization contribution from the land surface relative to aerosol. Likewise, polarization of larger particles is so weak that POLDER is most sensitive to fine-mode aerosols such as smoke from biomass burning and anthropogenic aerosols from industrial pollution. Figure 5 shows a global distribution of POLDER-derived aerosol index in the spring of 1997 (Bréon et al. 2002). A high aerosol index is observed over Asia and Africa. Air pollution from fossil and bio-fuel combustion is responsible for the high loading of fine mode aerosols in these regions. Note that it remains a major challenge to derive aerosol optical depth from the POLDER aerosol index. The angular properties used by MISR are also yet to be validated together with the MISR operational aerosol products.

The Total Ozone Mapping Spectrometer (TOMS) is another very useful sensor for aerosol monitoring. The TOMS aerosol index is derived from reflectance measured at 340 nm and 380 nm. The TOMS aerosol index provides a mixed measure of aerosol loading and absorption that is insensitive to surface reflectance (surface albedo at ultraviolet wavelengths is small and invariant). It has proven to be useful for identifying major episodes of absorbing aerosols from such events as biomass burning and dust storms. This sensor is especially useful in monitoring dust storms that are usually swept over desert or semi-arid regions and whose high brightness renders spectral techniques useless. The long-term record of TOMS data (since 1979) allows us to reveal the trend of aerosol change due to dust storms over the East Asia. There is a clear increasing trend in the number of strong dust storms over the past few years. However, because of the dependence of the aerosol index on aerosol altitude (usually not known), weak and/or low-flying dust cannot be seen from the TOMS platform. This, together with contamination by clouds, poses a big challenge for the quantitative determination of aerosol optical depth and single-scattering albedo from TOMS. Note that the index is not necessarily correlated with aerosol loading. Li et al. (2001) found that the high

TOMS aerosol index due to biomass burning corresponded to elevated tenuous smoke far from the heavy smoke surrounding the origin of burning. If the aerosol vertical distribution is known, as will be determined from the future CALIPSO spaceborne lidar, aerosol parameters retrieved from TOMS would be more comparable to ground-based AERONET data (Torres et al. 2002).

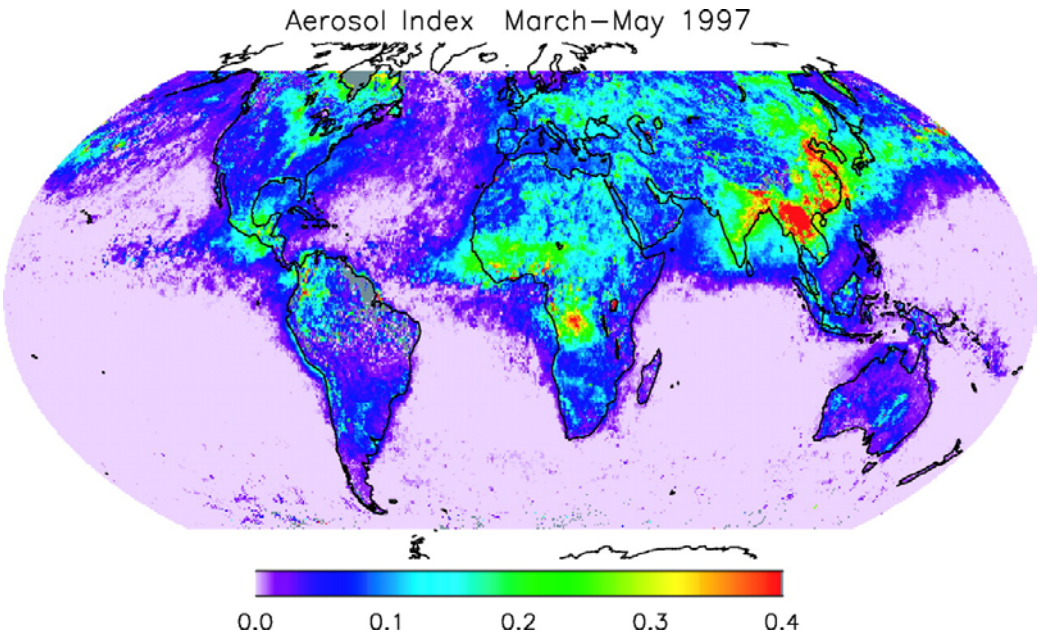

Figure 5. Global distribution of the aerosol index derived from the POLDER (Bréon et al. 2002). The index is most sensitive to fine-mode aerosols.

4. Aerosols and Aerosol Studies in China

Aerosol optical depth over most Chinese cities has increased significantly (*Qiu et al. 1997*). Using routine ground-based solar radiation measurements made across China since the 1960s, Luo et al. (2001) obtained the long-term trend and spatial variations of aerosol optical depth in China (Fig. 6). Similar estimates were also made with the direct solar radiation data by *Qiu et al* (1995). While the retrieved values are likely subject to large uncertainties resulting from numerous assumptions and from cloud screening issues, the patterns of temporal and spatial variations are reasonable. Overall, aerosol optical depth increases steadily from 1960s to 1980s; eruption of the El Chichon volcano might be responsible for the peak observed in the early 1980s. A weak decreasing trend during the 1990s could be an indication of the consequences of emission control enforced by the government. Streets et al. (2001) found a general decrease in sulfur dioxide (SO_2) emissions in China since 1995. On the other hand, emissions of mineral dust in China have an opposite trend. It is thus necessary to separate

dust aerosols from anthropogenic aerosols; fortunately, their respective optical properties are distinct.

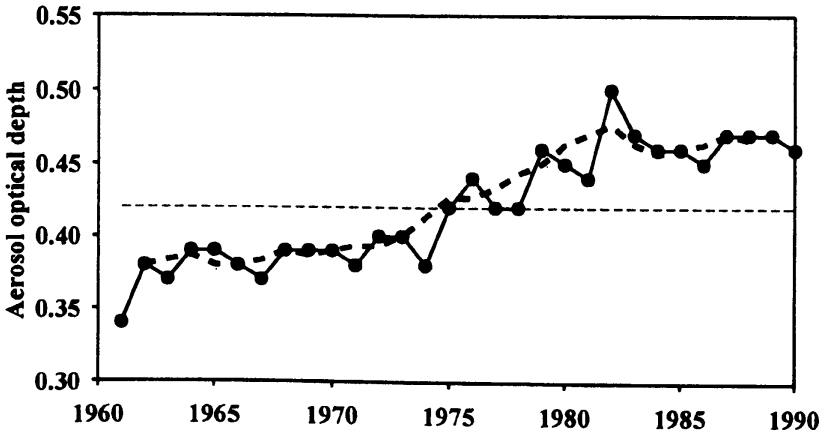

Figure 6. National average of aerosol optical depth over China derived from ground-based radiation measurements (after Luo et al. 2001).

4.1. *Dust Aerosols*

Mineral dust aerosols originate from desert regions, agricultural fields, barren land, and semi-arid regions. Dust storms are of primary concern due to the magnitude of influence and spatial coverage. Major dust storm tracks have been identified, as shown in Fig. 7. (Sun et al. 2001). There are two major sources of dust outbreaks in China: the western Xinjiang territory that includes the Taklamakan Desert, and northwestern Inner Mongolia that includes the Gobi Desert. In Xinjiang, desertification has increased by 400 km^2 every year. Dust emitted from China's nine deserts and semi-arid regions may affect the environment and climate of the mid-latitudes regions in the northern hemisphere. The annual mean dust emission from China is estimated to be around 800 teragrams (Tg). Drought in northern China may have played an important role in dust outbreaks. It remains a critical question, though, whether the aerosol indirect effect has anything to do with the drought trend. The direct cause of dust storms is vegetation cover lost in farming and ranching activities. 27% of the total land area of China is desert or is undergoing desertification with an economical impact of approximately $0.6 billion annually. Dust storms tend to intensify during daytime and weaken at night (*Xu et al.* 1979).

To probe the vertical concentration of dust aerosol, a handful of lidar instruments were deployed in China (*Qiu et al. 1984*). Long-term lidar observations of aerosol profiles have been obtained over Beijing and Hefei (*Zhou et al. 1998b*), supplemented by some short-term campaigns. A unique and seemingly ubiquitous maximum in aerosol concentration between 5 km and 8 km was found both in China and in the United States. This is because dust is usually lifted to the upper atmosphere by cold fronts in the lower atmosphere (below 4 km) and traveled with the westerly wind at higher altitudes.

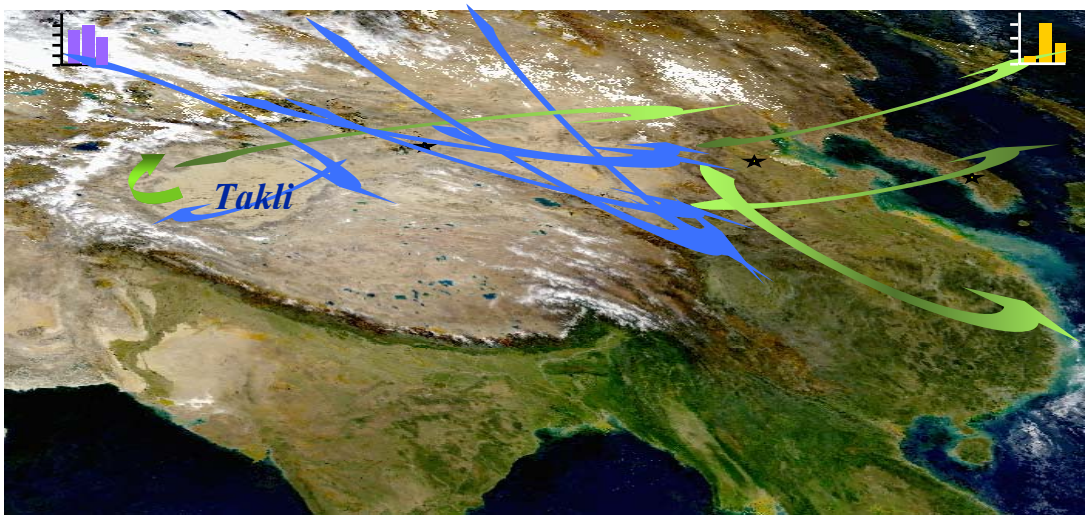

Figure 7. Major dust storm tracks in China (after Sun et al. 2001) superimposed upon MODIS real-color image (courtesy of S.-C. Tsay).

The radiative forcing of dust aerosol was evaluated using AVHRR and ground observations made during the HEIFE experiment (*Shen and Wei 1999a, 1999b; Wei and Shen 1998*). In Beijing, solar radiative heating of the atmosphere is larger on dust storm days than on dust-free days by 80-318% (*Yi and Han 1989*). *Zhao et al.* (*1983*) monitored aerosols in Beijing with a seven-band sun photometer, from which the aerosol size distribution was retrieved. The radii of dust particles were found to be generally larger than 2.1 μm, while the radii of pollutant aerosols were generally less than 2.1 μm (*Yang 1995*). The average refractive index in Beijing was found to be 1.517–0.034i during heating periods and 1.533–0.016i during non-heating periods. Note that these values are very crude estimates from broadbance solar direct and sky diffuse radiance measurements using shadowband radiometers for the simultaneous retrieval of aerosol optical thickness, size distribution, refractive index, and surface albedo (*Lu et al. 1981; Qiu 1983, 1986*). The imaginary part of the refractive index is dictated primarily by anthropogenic aerosols that are often mixed with mineral dust in Northern China. For pure dust aerosol, its value is much smaller (near zero revealed by the ACE-Asia studies). Dusty weather can reduce visibility to less than 50 m, increase local TOA albedo by 50-100%, and reduce total solar radiation by 10-40% (*Zhou et al. 1994*).

Dust originating in East Asia can travel a long distance, influencing a large area of the Asia-Pacific rim (Arimoto et al. 1996). During the major dust episodes observed in the spring of 1998, dust storms swept from the Takamoukou desert crossed the Pacific and reached the United States within one week. Extensive documentation has shown the impacts of these aerosols (and gases) at the Mauna Loa Observatory in Hawaii (Hubert et al. 2001) and in the Midway Island (Prospero et al. 2002). High loading of transported aerosols was found in the optically important 0.3 to 1.0 μm size range (Perry et al. 1999).

4.2. *Anthropogenic Aerosols*

As the most populated and fastest developing country of vast territory, China is a major source of anthropogenic aerosols. At present, SO_2 emissions from China account for about 20% of the world's total (Lefohn et al. 1999) and is expected to double between 1995-2050 (Streets and Waldhoff 2000). About 50% of the anthropogenic SO_x emitted over East Asia is removed from the continental source regions (Tan et al. 2002). The remaining could be carried to Hawaii or even further (Perry et al. 1999; Propero et al. 2003). The primary energy sources in China are coal (74.8%, or 1.36 billion tons per year in 1997), oil (17.9%), natural gas (1.77%), hydroelectric power (0.09%), and nuclear-generated power (5.44%). The dominance of coal combustion combined with a generally low burning efficiency results in large quantities of black carbon emitted into the atmosphere (Streets et al. 2001). Elemental carbon and organic aerosol concentrations in China are also exceptionally high and may contribute substantially to aerosol direct forcing at both regional and global scales. Other emissions have also increased due to a dramatic rise in automobile use and the rapid expansion of private industry. Major pollutants include SO_2, nitrogen oxides, soot, and suspended particles. The total annual emission of SO_2 amounts to 20.9 Tg (76.2% industrial and 23.8% domestic) and the countrywide average atmospheric concentration of SO_2 is 0.056 mg m^{-3} and 0.037 mg m^{-3} for nitrogen oxides. Note that nitrogen oxide concentrations in Chinese cities may increase rapidly with a drastic increase in private vehicles.

Heavy pollution has a severe adverse impact on the environment, on human life, and on the economy. For example, the large concentration of SO_2 over China has caused widespread acid rain (*Wang 1997*). The total area affected by acid rain is approximately equal to one third of China's total territory, including more than half of Chinese cities, mostly in southern China. In the foreseeable future, the composition of the energy infrastructure in China is unlikely to change dramatically, but the demand for energy will continue to grow, likely leading to more pollution and more anthropogenic aerosols. The problem is exacerbated by the increasing numbers of small, inefficient power plants owned privately or by local governments. Such small enterprises usually do not have coal-cleaning and sulfate-removal facilities essential to produce "clean energy." Currently, the overall consumption of energy per capita in China is very low, about 1/48 the level of the United States; the potential for increase in energy consumption is phenomenal.

Due to air pollution, the imaginary part of the aerosol refractive index is generally high, and decreases from cities to the countryside (*Hu et al. 1991*). The single-scattering albedo is generally low, as is shown in a comparison between Beijing and Washington, DC (Fig. 8). The chemical composition of urban aerosols in China differs considerably from that in North America and Europe, with significantly higher concentrations of sulfate and black carbon (*Wang et al. 1981; Ren et al. 1982*). This is consistent with the findings of *Zhang and Shi* (2000) showing that the fine mode aerosols have increased more rapidly in the past decade, associated with a sharp increase in coal combustion and automobile emissions. Industrial and urban emissions are present year-round and are likely exacerbated by the hot, humid summer monsoon season in the heavily populated southeastern provinces. Note that the differences in

single-scattering albedo as shown in Fig.8 are tremendous in terms of the influence of aerosol on the solar radiation budget.

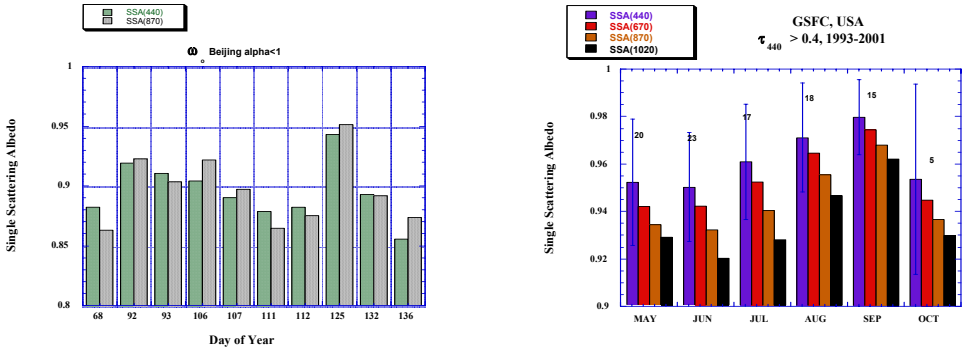

Figure 8. Comparison of single-scattering albedos in Beijing and Washington as retrieved from AERONET measurements (courtesy of B. Holben, 2002).

The highly hygroscopic nature of the pollutants compounds its climate effect due to the influence of humidity (Li et al. 1996, 2000). Observations in Chongqing indicated that aerosol optical depth is doubled as relative humidity increases from 65% to 90%. Because of their high soot content, these aerosols have a warming effect (over 1 °C per day) in the upper boundary layer and a cooling effect in the lower boundary layer, leading to a more stable atmosphere during the daytime. At night, the effect is opposite. Chameides et al. (2002) even argued that the world thickest clouds found in Northwestern Pacific is caused by air-pollution from China.

4.3. *Climate Effects*

Direct aerosol forcing is strong and complex due to high loading and strong absorption of aerosols overlying bright surfaces. One significant effect is a reduction in direct solar radiation at the surface (*Xu, 1990*). According to Luo et al. (2001), this reduction from the 1960s to the 1980s is larger than 20% in all cities, with the largest decrease of 29.2% occurring in Guangzhou, where visibility has diminished by more than 50% during the last two decades! Using the aerosol optical thickness data estimated from surface radiation measurements, Luo (1998) computed aerosol radiative forcing across China and found that the largest forcing (−13 Wm^{-2}) occurred in spring and the smallest (−8 Wm^{-2}), in winter. Note that these estimates are subject to considerable uncertainties due to errors in the retrieval of optical thickness and lack of observations for other aerosol attributes (e.g. single-scattering albedo). The distribution and radiative forcing of anthropogenic aerosols was also modeled, based on an inventory of emission sources and strengths (*Hu and Shi 1998a, 1998b; Zhang and Gao 1997*).

The strong radiative forcing resulting from large aerosol loading impinges substantially upon regional climate (Zhou et al. 1998b; Hu and Shi 1998). One direct effect is surface cooling that can exceed the warming effect of increased greenhouse gases. For example, a

generally decreasing trend of surface temperature observed in the heavily industrialized Sichuan Basin (Fig. 9 left) was attributed to the influence of exceptionally high aerosol concentrations. Statistical studies (Xu and Wang 1993; Xu 1997) revealed a strong link between air pollution and China's monsoon regime. Significant correlations were found between an increase in aerosol loading (air pollution), a decrease in solar radiation, and the abnormal location of the subtropical high-pressure system located in the western Pacific. He argued that the "northern drought, southern flood" anomaly is associated with increases in air pollution (Xu 2001).

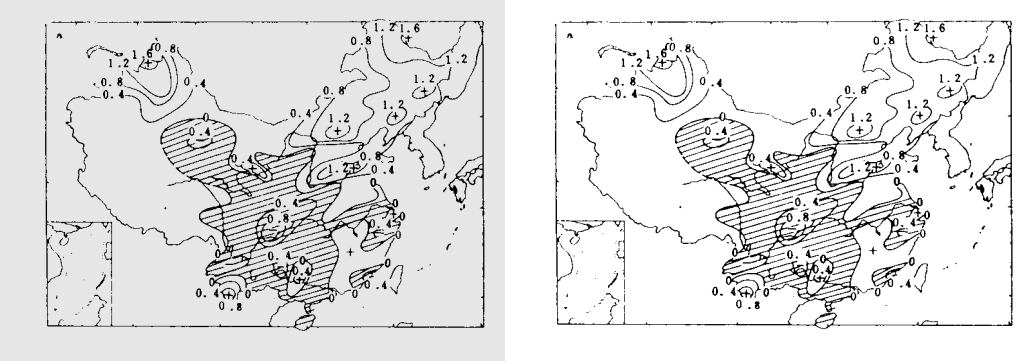

Figure 9. Temperature and precipitation changes in China. Left: Yearly mean temperature difference between 1980s and 1950s (Luo 1998); Right: Rainfall anomaly in 1980-1993 relative to long-term mean (1951-1980) (Xu 1997).

The argument was further substantiated by Menon et al. (2002) using the NASA/GISS general circulation model. They simulated three aerosol scenarios: no aerosol, sulfate aerosol, and sulfate plus black carbon aerosols. Given that the BC concentration in Asia, in particular India and China, is very high, they adopted a very low single-scattering albedo of 0.85, invariant with time and location for the third scenario. Using the aerosol optical depths estimated by Luo et al. (2001), they obtained two distinct climates in the Asian region in terms of changes in temperature and precipitation distribution. It was found that the temperature decrease in East Asia due to sulfate and BC aerosols is much more than that due to sulfate aerosol alone. This is because both types of aerosols lower the amount of solar radiation reaching the ground. Changes in precipitation due to sulfate aerosol are relatively minor. However, when an absorbing aerosol is added, the difference is dramatic. Precipitation decreases in northern China but increases in southern China. The changes in precipitation are induced by changes in atmospheric convection initiated by inhomogeneous aerosol heating in the boundary layer. In the Sichuan basin located in southern China, high aerosol loading and humidity accelerates the upwelling convection leading to excessive precipitation. In northern China, the subsiding air causes a precipitation deficit. While this theory may have oversimplified the workings of the climate in the region (e.g. the monsoon dynamic is not

included), it demonstrates a potentially very large role played by aerosols in dictating the regional climate.

There are very few studies on the aerosol indirect effect in China, although the effect could be substantial, especially with regard to suppression of precipitation. If so, aerosols would have a significant impact on the hydrological cycle and on the monsoon regime that governs summer weather and climate across China. These arguments are essentially hypotheses that are yet to be substantiated with more sound physical mechanisms following rigorous scientific investigations.

Of course, the impact of pollution is more far-reaching than just climate. Air pollution in China is likely to reduce crop yield by 5-30% due to reduced photosynthetically active radiation (PAR) reaching the surface, according to a model study by Chameides et al. (1999). On the other hand, aerosols increase diffuse solar radiation, thus improving the efficiency of canopy photosynthesis and boosting productivity. There is also a significant linkage between pollution and mortality. It is acknowledged that today more people die from pollution than cancer in China.

5. Challenges, Prospects and Recommendations

Despite the numerous encouraging advances in characterizing aerosols and understanding their climate effects in China, we are confronted with many daunting tasks such as unraveling the complex relationships between aerosol and climate on both regional and global scales. Previous studies provided preliminary insights into the basic characteristics of
aerosols in East Asia, but our knowledge remains very much uncertain for the following reasons.

First, many of the instruments used in the aforementioned studies are outdated with low or un-quantified accuracies. Some advanced instruments were deployed for a short period over few locations. There are no operational aerosol observation stations equipped with rigorously calibrated instruments. China recently purchased 30 Cimel sun photometers mainly for monitoring dust storms. The lack of long-term seasonal and large-scale measurements of aerosol microphysical and radiative properties means there is no baseline against which to gauge changes in aerosol loading and climate impacts into the future. Given the uniqueness of the aerosols in the region, measurements and information acquired over other source regions cannot be substituted to fill the gap. With major advances in ground-based observational techniques and analysis methods, it is highly desirable to establish a long-term routine aerosol observation network equipped with advanced instruments such as those used in the AERONET. At present, there exists a big gap in the coverage of AERONET in China.

Few attempts have been made to utilize satellite data to quantify the spatial and temporal variations of aerosols in China. Only a handful of attempts were made by Chinese scientists to use satellite data to monitor aerosols (Zhao and Yu 1986). Most studies have been limited to qualitatively browsing imagery for major aerosol episodes, such as dust storms (Zhou et al. 1994). A quantitative investigation was made recently to retrieve aerosol optical thickness

over major lakes across China using the GMS-5 and MODIS satellite data (Liu 1999; Mao et al. 2000). There are several major challenges in the retrieval of aerosol properties from satellite. A combination of bright surfaces and absorbing aerosols that are often encountered in China makes the retrieval of aerosol optical thickness particularly cumbersome, unless aerosol properties are known reasonably well.

Lack or insufficient knowledge of ground truth information pertaining to aerosol optical properties, surface reflectance and BRDF all hinder remote sensing of aerosol. *In situ* aerosol measurements are required to properly characterize the physical and chemical properties of major aerosol types found in the region. A major piece of missing information is the composition of pollution aerosols. A large uncertainty is probably linked to the source of carbonaceous aerosols. Although its contribution to total optical thickness is relatively small, the black carbon concentration is often a determinant factor for aerosols to warm or cool the atmospheric column.

The largest source of uncertainty lies in estimating aerosol direct and indirect effects and understanding their impact on regional and global climate. Many aspects of aerosol direct forcing are yet to be explored, especially its impact and relation with the Asian monsoon system. In China, aerosols may have an even stronger effect on atmospheric dynamics through which both precipitation intensity and pattern may be severely affected. Aerosols could have a significant impact on the monsoon regime that governs summer weather and climate across China. Physical mechanisms that may link air pollution and the monsoon episode await discovery. Reducing solar illumination at the surface, aerosols may weaken the monsoon atmospheric circulation by lowering the land-ocean thermal contrast, a condition unfavorable for the development of monsoon weather. Since the annual rainfall in northern China comes chiefly from precipitation associated with the summer monsoon, any retreat/weakening of the monsoon system could have tremendous impact on the hydrological cycle. To tackle this problem, one sound approach is to develop a more reliable climatology of aerosol radiative forcing (at the TOA, the surface and inside the atmosphere) across the region over a long period of time, incorporate them into regional climate models capable of generating the monsoon regime (e.g. Tao et al. 2002) and compare the model- simulated and observed monsoon time series.

Given the high concentration and chemical composition of aerosols in China, there may be particularly strong aerosol indirect effects in China. Yet, the aerosol indirect effects are likely more complex than what we have known from studies conducted in relatively clean environments. So far, aerosol indirect effects are mainly confined to alteration of cloud microphysics and lifetimes, although the indirect effect may be coupled with the direct effect and atmospheric dynamics. In China, the indirect effect may be a direct cause of precipitation reduction in northern China, where there is limited water vapor but plentiful CCN, whereas in the southern region, an abundance of both water vapor and CCN may produce excessive precipitation. The latter is corroborated at least in part by acid rain observations that indicate that most precipitation in southern China has rather high values of pH. This hypothesis needs rigorous testing using comprehensive *in situ* cloud and aerosol measurements in cloud

microphysical models containing aerosol-cloud interaction schemes like those proposed by Chen and Lamb (1999) and Khain et al. (2000).

In summary, there are plenty of investigations that can be conducted to unravel the complex aerosol-energy-hydrology relationship. China is an excellent testbed for examining aerosol climate effects that may well go beyond the current paradigms. To this end, we need to tackle the following fundamental questions (to name a few):

- What are the spatial and temporal variations of single-scattering albedo?
- What is the aerosol radiative forcing at the surface and at the TOA in the region?
- How do mineral dust aerosols interact with anthropogenic aerosols?
- Do Asian aerosols modify cloud microphysics and precipitation patterns, and if so, how?
- Are the radiative and hydrological effects of anthropogenic aerosols large enough to alter atmospheric circulation patterns and regional climate?

To address these questions, it is recommended that extensive research on the following basic issues be conducted:

(1) Aerosol characterization

- *In situ* observation and ground-based remote sensing (especially AERONET) of aerosol microphysical, chemical, and optical properties;
- Identification of aerosol types (dust, soot, sulfate, organic and carbonaceous aerosols);
- Use measurements of aerosols and trace gases to better understand the origins of the particles and to test and improve emissions inventories;
- Satellite-based mapping of aerosol spatial (including vertical) and temporal variations from such sensors as MODIS, TOMS, SeaWiFS, AVHRR, etc.;
- In situ and remote sensing of broadband and spectral surface albedos, radiative fluxes, precipitable water, etc.

(2) Aerosol direct radiative forcing

- Direct radiative forcing at the top, bottom and within the atmosphere;
- Radiative forcing of different aerosol types (dust, soot, sulfate, mixed);
- Combining observations and modeling studies to estimate aerosol forcing under cloudy conditions.

(3) Aerosol indirect effects

- Indirect radiative forcing; hygroscopic (humidification) effect; hydrological suppression and distribution;
- Acid rain and acidity effects;
- Cloud lifetime.

(4) Impact on regional/global climate

- Absorbing/non-absorbing aerosol in regional/global models for EOS-data assimilation; aerosol-climate-atmospheric chemistry;
- Regional/global climate simulation; feedback effects (absorbing aerosol).

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