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Introduction to special section: Outstanding problems in quantifying the radiative impacts of mineral dust

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Abstract. This paper provides an introduction to the special section of the *Journal of Geophysical Research* on mineral dust. We briefly review the current experimental and theoretical approaches used to quantify the dust radiative impacts, highlight the outstanding issues, and discuss possible strategies to overcome the emerging problems. We also introduce the contributing papers of this special section. Despite the recent notable advances in dust studies, we demonstrate that the radiative effects of dust remain poorly quantified due to both limited data and incomplete understanding of relative physical and chemical processes. The foremost needs are (1) to quantify the spatial and temporal variations of dust burden in the atmosphere and develop a predictive capability for the size- and composition-resolved dust particle distribution; (2) to develop a quantitative description of the processes that control the spatial and temporal variabilities of dust physical and chemical properties and radiative effects; (3) to develop new instrumentation (especially to measure the dust particle size distribution in a wide range from about 0.01 μm to 100 μm , scattering phase function and light absorption by dust particles); and (4) to develop new techniques for interpreting and merging the diverse information from satellite remote sensing, in situ and ground-based measurements, laboratory studies, and model simulations. Because dust distribution and effects are heterogeneous, both spatially and temporally, a promising strategy to advance our knowledge is to perform comprehensive studies at the targeted regions affected by mineral dust of both natural and anthropogenic origin.

1. Introduction

Radiative effects of mineral soil-derived aerosols (or dust), traditionally considered a natural aerosol, have long been the subject of studies [Duce, 1995]. However, only in the past decade have researchers realized the important contribution of anthropogenic dust to the radiative forcing of climate. Various human activities (such as land use practices, construction, etc.) can result in significant dust loading, with the anthropogenic fraction of dust estimated to be as much as 30% to 50% of total dust production, and its direct radiative forcing on both global and regional scales may be comparable to or exceed the forcing by other anthropogenic aerosols [Tegen *et al.*, 1996, Sokolik and Toon, 1996]. However, the large uncertainties in key dust

properties and in the amount of anthropogenic dust make it impossible to quantify the magnitude of radiative forcing, and even the sign of total (solar plus infrared) forcing at the top of the atmosphere remains unclear. In addition, it has been recognized that the presence of dust results in diverse radiative impacts that must also be quantified to predict the overall effect of dust on climate. The existing uncertainties in an assessment of the impact of dust is due in part to the limited data on dust climatology (i.e., spatial pattern of dust distribution); but more fundamentally, they are a consequence of our incomplete understanding of the processes responsible for the production, transport, physical and chemical evolution, and removal of mineral aerosols at various space and time scales. A reduction of these uncertainties requires the integrated and interdisciplinary research efforts of the international scientific community.

This was a motivation behind the organization of this special section on mineral dust. This collection of papers, many of which were presented at an international Workshop on Mineral Dust held in Boulder, Colorado, in June 1999, focuses on the following topics: modeling and measurements of mineral dust production, transport and deposition; modeling and measurements of size-resolved chemical and mineralogical dust composition; individual particle analysis; modeling and measurements of dust optical and radiative properties; regional and global dust modeling for climate change and atmospheric chemistry studies; and satellite remote sensing of mineral dust. A brief report of the Workshop has been published in *Eos* [Sokolik, 1999a].

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Table 1. Direct Radiative Impacts of Mineral Aerosols

Impact	Importance
TOA radiative forcing (solar plus IR)	affects energy balance of the Earth's climate system
Radiative forcing at the surface (solar plus IR)	affects surface temperature and surface-air exchange processes
Radiative heating/cooling (solar plus IR)	affects temperature profile and atmospheric dynamics
Actinic flux (UV)	affects photolysis rates and photochemistry

The ultimate goal of this special section is to report recent progress and new findings in dust studies, covering a wide variety of interdisciplinary issues relevant to an assessment of the dust radiative impacts. In this introductory paper we highlight the important gaps in the knowledge, discuss outstanding problems, and present recommendations for future research on mineral dust. The contributing papers of this special section are introduced in the context of the issues discussed. In section 2 we define the various radiative effects of dust and review the pros and cons of the experimental and theoretical approaches employed to deduce the magnitudes of these effects. Section 3 discusses the outstanding issues in quantifying the radiative impact of dust and recommends strategies to overcome them, followed by a summary.

2. Current Understanding of Radiative Impacts of Mineral Dust: Definitions, Approaches, and Limitations

2.1. Definitions

Both natural and anthropogenic components of mineral aerosols may cause diverse radiative impacts, as summarized in Table 1. The direct radiative effects of mineral particles are due to their ability to scatter and absorb UV, visible, and infrared radiation.

To quantify the effects of aerosols on the Earth's radiative balance, the concept of direct radiative forcing is often employed. Although this concept was introduced originally to characterize the effect by anthropogenic aerosols such as secondary sulfate, it has been extended to include the effects of dust. Typically, the effects due to total dust (anthropogenic plus natural) are analyzed, because it remains difficult to quantify the anthropogenic contribution due to the large uncertainties in defining the anthropogenic fraction of the ambient dust burden, as we discuss below. The radiative forcing of dust is typically calculated as the difference in net irradiance at the tropopause (or at the top of the atmosphere (TOA)) in clean and dust-laden atmospheric conditions at a given moment of time (or averaged over a time period, day, month, or year). The appropriate space scales and timescales for averaging of TOA forcing remain an open issue.

Dust can cause either a positive or a negative radiative forcing leading to a warming or cooling of the climate system. The sign of the direct radiative forcing is determined by the optical properties of the dust and its distribution in the atmosphere as well as atmospheric conditions and surface reflectance. Moreover, the solar and infrared radiative forcings of dust may be of different signs, thus further complicating an assessment of total (solar plus infrared) forcing [Sokolik *et al.*, 1998].

Because dust sources and sinks are not uniformly distributed and because of the short lifetime of mineral particles in the atmosphere, it has been predicted that direct radiative forcing has a complex geographical distribution [e.g., *Teegen et al.*, 1996; *Myhre and Stordal*, this issue]. In addition, the properties of dust vary in time and space [*Husar et al.*, this issue; *McKendry et al.*, this issue; *Chun et al.*, this issue]. As a result, the determination of the sign and the magnitude of TOA direct radiative forcing by dust on regional and global scales remains a key unresolved problem in predicting climate change.

The presence of dust also alters the surface radiation budget (often referred to as direct radiative forcing at the surface). It has been demonstrated that the radiative forcing by dust at the surface is often much larger than that at the top of the atmosphere [*Teegen et al.*, 1996; *Quijano et al.*, 2000]. It implies that surface temperature and hence various surface exchange processes may be affected by dust.

Furthermore, the radiative heating/cooling occurring in the dust layer itself may affect the atmospheric temperature profile and thus air dynamics and precipitation [*Alpert et al.*, 1998; *Quijano et al.*, 2000]. For instance, *Karyampudi and Carlson* [1988] showed that radiative heating by Saharan dust contributes to maintaining a warmer and deeper Saharan Air Layer (SAL) over the ocean, to enhancing the strength of the midlevel easterly jet, and to reducing the convection within the equatorial zone. On the mesoscale, dust radiative heating rates can affect the evolution of a dust storm leading to stronger surface frontogenesis [*Chen et al.*, 1994]. Consequently, knowledge of radiative heating rates by mineral aerosols may be decisive to better predictions of the dynamics associated with dust transport.

In addition, by altering ultraviolet (UV) radiative fluxes, the presence of dust may perturb the photochemistry of the atmosphere. The strong UV-absorbing dust causes a decrease in photolysis rates, which in turn inhibits ozone production [*He and Carmichael*, 1999]. The ability of dust to inhibit the production of photochemical smog could have an important implication in air pollution studies.

Figure 1 illustrates a standard procedure and the quantities required to perform calculations of the diverse radiative impacts of dust. There are numerous codes developed to solve the radiative transfer equation under various approximations. However, to perform these calculations, one needs to know a set of optical properties of dust such as optical depth, single scattering albedo, and asymmetry parameter as a function of location, time, altitude, and wavelength. The set of optical properties required varies, depending on the radiative transfer scheme used. If radiances are required (e.g., for remote sensing applications), this set should also include the scattering phase function. In turn, the optical properties of dust, as well as other atmospheric aerosols, depend on the fundamental microphysical properties such as particle size distribution, composition and

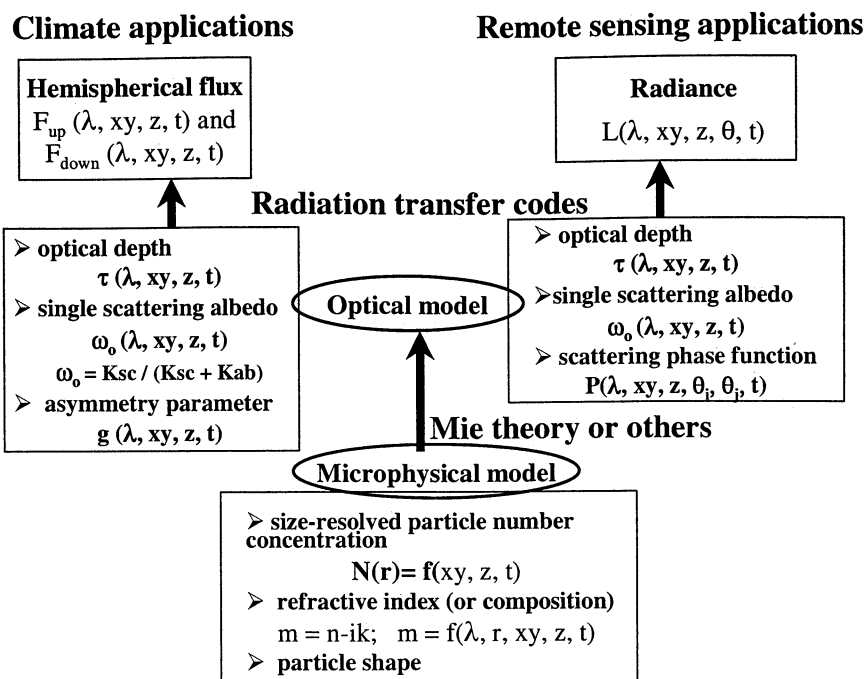


Figure 1. A standard procedure and the quantities required to perform the calculations of diverse radiative impacts of dust.

mixing state (and/or spectral optical constants), and particle shapes. It is the evolution of the fundamental properties which determines how aerosol optics and hence radiative impacts may vary in space and time. Therefore quantification of the radiative impacts of dust must include various processes governing changes of the physical and chemical properties of dust as well as dust emission and atmospheric transport.

It is important to point out that dust may also affect the Earth's radiative budget by altering clouds and atmospheric chemistry. There is strong evidence that mineral particles may serve as cloud condensation nuclei [e.g., Levin *et al.*, 1999]. Dust may thereby affect the Earth's energy balance indirectly by altering cloud properties and their amount (termed an indirect radiative forcing of climate).

In addition to altering photolysis rates, the presence of mineral dust in the atmosphere can also alter photochemical processes by a variety of means, including influencing the partitioning of semivolatile trace species (e.g., the partitioning of nitric acid and ammonia between the aerosol and the gas phases), providing (possible) additional reaction pathways for reactions involving H_xO_y (OH, HO_2 , H_2O_2), N_xO_y (HNO_3 , N_2O_5 , NO_3 , NO_2), SO_2 , O_3 , and semivolatile organics, and transporting trace substances to the oceans. So far, these processes remain largely unquantified. However, new laboratory experiments are beginning to provide the basis to evaluate chemical interactions with dust [Underwood *et al.*, this issue].

In turn, chemical processes occurring on the dust particle surfaces may result in the formation of multicomponent aerosols which can have drastically different optical properties and, hence, radiative impact, from those at the dust source [Sokolik, 1999b]. This adds further complexity to the assessment of the radiative effects of dust.

Although not discussed in this special section, it is worth mentioning that the presence of dust may have various other effects: pose a health threat; affect biogeochemical processes in the oceans; affect terrestrial systems, cause property damage, affect agricultural production, etc. Traditionally, these issues are

studied by scientists from very different and poorly-connected fields. It becomes clear that improvements in the quantification of overall dust impact will require joint efforts from the interdisciplinary scientific community.

2.2. Experimental Studies

Various experimental and modeling studies have been conducted to evaluate the radiative impacts of dust. During the last few decades, several international field campaigns have been organized. However, most existing radiation observations are from ground-based short-duration studies and are available for a limited number of locations (with the exception of satellite observations). The most limited are measurements of radiative heating/cooling rates because just a few aircraft studies of dust layers have been conducted [Haywood *et al.*, this issue]. As a result, the diverse radiative impacts of dust cannot be fully quantified from existing measurements. Therefore current estimations of dust radiative impact are mainly performed by modeling, as illustrated in Figure 1, utilizing measurements of dust optics and microphysics.

Table 2 lists measured optical and microphysical quantities along with the capabilities and limitations of the instruments relative to dust studies. There remain serious deficiencies in the available measurement techniques.

For instance, the single scattering albedo, which is a crucial parameter indicative of the heating or cooling effects of dust, remains poorly quantified. This characteristic is not measured directly but calculated using the light scattering coefficient measured in situ and the light absorption coefficient measured from the difference in light transmission through aerosol-laden and clean filters. Each of these techniques has numerous limitations when used for measurements of light scattering and absorption of dust. For instance, a nephelometer has a truncation error (due to limited angular range), limited particle size coverage (losses of particles greater than $2 \mu m$ diameter), and calibration problems [e.g., Schmid *et al.*, 2000].

Table 2. Optics and Microphysics Measurements Relative to Dust Studies

Measured Quantity	Instrument	Instrument Capability	Instrument Limitations
<i>Dust Optics</i>			
Light scattering coefficient	nephelometer	single or multiple wavelengths in visible variable relative humidity; controlled particle size-cut varying scattering angles	no measurements in IR limit on larger particle size donot cover all range of scattering angles and hence no measurements of scattering phase function problems in interpretation of scattering due to possible nonsphericity and absorption of dust particles
Light absorption coefficient	aethalometer	single or multiple wavelengths in visible ambient relative humidity controlled particle size-cut	no measurements in IR limit on larger -article size limitations in the filter-based techniques and sampling procedure problems in partitioning absorption between species
	photometer		
	photoacoustic instrument	in situ absorption	sensitivity single wavelength 0.802 μm
<i>Dust Microphysics</i>			
Particle size distribution	optical particle counter	size bins from 0.1 to 5 μm	uncertainties due to nonspherical shape and composition
	impactor	size bins from 0.03 to 10 μm	problems with size separation
	electron and optical microscopy	10nm <D<500 μm	dry and stable particles
Particle chemical and mineralogical composition	X-ray fluorescence and diffraction	bulk or individual particle	dry and stable particles
Particle shape	microscope	individual particles	only two-dimensional (if $D < 10 \mu\text{m}$) and for dry particles no systematic measurements have been reported
Mixing state	transmission- and scanning electron microscope	identify internally-mixed species	only for dry particles no systematic measurements have been reported

The absorption of light by dust has been measured using aethalometers and absorption photometers (described by Hansen *et al.* [1982], and Bond *et al.* [1999]) despite these instruments having been designed with black carbon in mind. Both types of instruments have serious problems in calibration and data interpretation [Bond *et al.*, 1999]. A need to separate the absorption due to dust from that due to aerosol black carbon introduces further uncertainty. Recent bulk and size-segregated filter measurements have demonstrated that light absorption by Saharan dust exhibits strong wavelength dependence, decreasing by more than an order of magnitude from 400 to 900 nm [Savoie and Voss, 1999]. However, because of limitations in the filter-based technique and sampling procedures, it remains unknown how well these measurements represent the light absorption by aerosol particles suspended in the atmosphere.

A photoacoustic technique, which eliminates the disadvantages of the filter-based methods, has also been used to measure light absorption by mineral particles [Dinmukhametova *et al.*, 1986]. Recent improvements of photoacoustic instruments make this approach very promising [e.g., Moosmuller *et al.*, 1998].

Just a few studies have reported the scattering phase function of mineral particles measured in the laboratory or atmospheric conditions [e.g., West *et al.*, 1997]. The scattering phase function is an important characteristic of aerosols which directly

impacts climate models as well as our ability to characterize aerosols from remote sensing observations. In particular, this information is critical for the adequate interpretation of lidar measurements [Murayama *et al.*, this issue].

In addition, there is a large gap between optics (and radiation) measurements and measurements of fundamental dust properties such as composition, particle size distribution, etc., which seriously complicates modeling studies. Recognizing this problem, recent experimental efforts have focused on so-called aerosol closure experiments in which an over-determined set of observations is obtained, and then, measured variables are intercompared with those calculated from appropriate theoretical models to test the closure. So far, only a few column-closure experiments have been organized to study mineral aerosols (e.g., ACE-2 [Russell and Heizenberg, 2000; Smirnov *et al.*, 1998], PRIDE [Reid *et al.*, 2000] and INDOEX (S.C. Alfaro *et al.*, Aerosol model derived from measurements performed at the Indian coastal site during INDOEX, submitted to *Journal of Geophysical Research*, 2000). In particular, ACE-2 closure studies revealed that due to existing limitations of dust measurements and modeling, a closure could not always be achieved [e.g., Schmid *et al.*, 2000].

One of the fundamental problems is the complex nature of mineral aerosols. Although the complexity of the mineralogical and chemical composition and morphology of dust has long

been appreciated, the importance of this information for predicting radiative and chemical impacts of dust in the atmosphere is just beginning to be explored [Dentener *et al.*, 1996; Sokolik and Toon, 1999; Claquin *et al.*, 1999]. Mineral aerosols is a collective term referring to widely varying mixtures that may include many constituents such as quartz, various clays, calcite, gypsum, hematite, and others. These minerals each have very different physical and chemical properties (e.g., size distribution, particle shape, density, spectral refractive index, solubility, and chemical reactivity). Moreover, individual particle analysis of samples collected at various locations reveals that dust particles often occur as irregular aggregates of minerals and/or other aerosol species [Parungo *et al.*, 1995; Gao and Anderson, this issue; Falkovich *et al.*, this issue]. No systematic study of irregular dust aggregates has been conducted so far.

Some aerosol optical and microphysical properties can be retrieved from ground-based or satellite remote sensing data [Vaughan *et al.*, this issue]. For instance, the observations acquired at AERONET (Aerosol Robotic Network) sites located downwind from dust sources have been used to calculate the spectral optical depth of dust [Sabbah *et al.*, this issue]. AERONET angular and spectral sky radiances have also been used to retrieve aerosol size distribution [Tanré *et al.*, this issue], refractive index, single scattering albedo [Dubovik *et al.*, 2000], and the spectral flux reaching the surface [Kaufman *et al.*, 2000]. However, these aerosol characteristics are retrieved under various assumptions (e.g., some critical dust characteristics must be prescribed a priori) and averaged over the vertical column. The meaning of the retrieved dust properties averaged over an atmospheric column (ignoring the vertical variations of aerosol properties) remains to be understood. This also applies to dust properties retrieved from satellite observations.

Radiances observed by several satellite instruments have been used to retrieve dust properties and characterize dust transport on the global and regional scales (e.g., TOMS [Alpert and Ganor, this issue; Diaz *et al.*, this issue; AVHRR [Husar *et al.*, this issue], TOMS and AVHRR [Cakmur *et al.*, this issue], SEAWIFS [Moulin *et al.*, this issue], and MeteoSat [Legrand *et al.*, this issue]. In general, the satellite instruments operating at UV and infrared wavelengths can detect dust over both land and oceans, while visible channel instruments are capable of detecting dust over the oceans only.

For instance, data from Total Ozone Mapping Spectrometer (TOMS) instruments, operating at UV wavelengths, and designed originally for remote sensing of ozone, have been used to monitor dust transport over land as well as over the ocean since 1979 [Herman *et al.*, 1997a]. TOMS is used to retrieve an aerosol index which is proportional to the aerosol optical thickness, but also sensitive to the aerosol absorption, particle size, aerosol vertical distribution, and clouds. Although the aerosol index is a qualitative characteristic, it is helpful in identifying dust sources and transport routes [Husar *et al.*, this issue].

The aerosol optical depth is another product often retrieved from satellite observations in UV and visible channels. For instance, several techniques, using one or two visible channels of AVHRR (advanced very high-resolution radiometer), have been developed to retrieve aerosol optical depth over the ocean [Stowe *et al.*, 1997; Durkee *et al.*, 2000]. All these techniques require assumptions on one or more of the following: aerosol particle size distribution, optical constants, single scattering albedo, and scattering phase function. Also, information on aerosol vertical structure and surface bidirectional reflectivity is crucial in these retrievals. Because of large uncertainties in satellite sensor characterization and calibration, and numerous

(and often poorly justified) assumptions used in the retrieval algorithms, the aerosol properties retrieved from satellite observations remain insufficiently validated. Several new space sensors, which have been explicitly designed for aerosol study, will overcome some of these problems, as will be discussed in section 3.5 (see also review by King *et al.* [1999]). Yet the problem of how to identify a dust contribution among other factors affecting the radiances observed by the satellite remains to be resolved.

Lidar measurements from the surface and from space reveal complex multilayered aerosol stratification in dust-laden atmosphere [e.g., Murayama *et al.*, this issue; Tratt *et al.*, this issue]. Until recently, lidar measurements have been limited to a relatively small number of surface sites and infrequent aircraft measurements. Although of limited duration, the first lidar system flown in space, the Lidar in Space Technology Experiment (LITE), demonstrated the capabilities of lidar to provide global aerosol observations [Winker *et al.*, 1996]. Lidar provides a very sensitive detection of aerosol over all surface types, at night as well as during the day. Lidar retrievals of aerosol optical depth normally require the assumption of either an aerosol scattering phase function or a value for the ratio between aerosol extinction and 180° backscatter. However, when dust occurs in elevated layers, the optical depth of these layers can be measured directly from the transmittance of the lidar beam through the layer [Young, 1995]. This capability is especially significant because the scattering phase function of mineral dust is largely unknown.

The limitations of current aerosol instrumentation seriously restrict our ability to quantify dust radiative effects and to constrain the parameterizations used in models. Significant improvements in aerosol measurement technology are necessary.

2.3. Modeling Studies

Stand-alone radiative transfer codes or codes linked with atmospheric dynamical models have been used to predict the radiative impact of dust. Dust effects have been simulated using global climate models [e.g., Penner *et al.*, 2001], global general circulation models [e.g., Woodward, this issue; Perlwitz *et al.*, this issue], regional transport models [e.g., Nickovic *et al.*, this issue, Liu and Westphal, this issue], regional chemical transport models [e.g., Song and Carmichael, this issue], and an on-line tracer model coupled with a regional atmospheric model [Uno *et al.*, this issue].

Some of the major processes (such as dust emission, sedimentation, and dry and wet removal) have been identified and included in the models to predict dust radiative impacts while other processes have been only hypothetically formulated (such as heterogeneous chemistry on dust particle surfaces, cloud processing, and interactions with other atmospheric aerosols). Although some atmospheric chemistry models include simplified dust chemistry, these models have not yet been used to calculate the dust radiative impact on the global scale. Because of the complexities involved and the lack of understanding of the interactions between the dust and the chemistry and physics of the atmosphere, model predictions currently rely on oversimplified parameterizations of dust processes.

A common approach simulating dust fields with an atmospheric transport model includes the following steps: (1) characterize a dust source, (2) use a dust production scheme to simulate dust emission, and (3) use transport and dust removal schemes to predict the size-resolved concentration fields or total mass concentration fields as a function of time and space depending on the resolution of a given model. If the dust field is

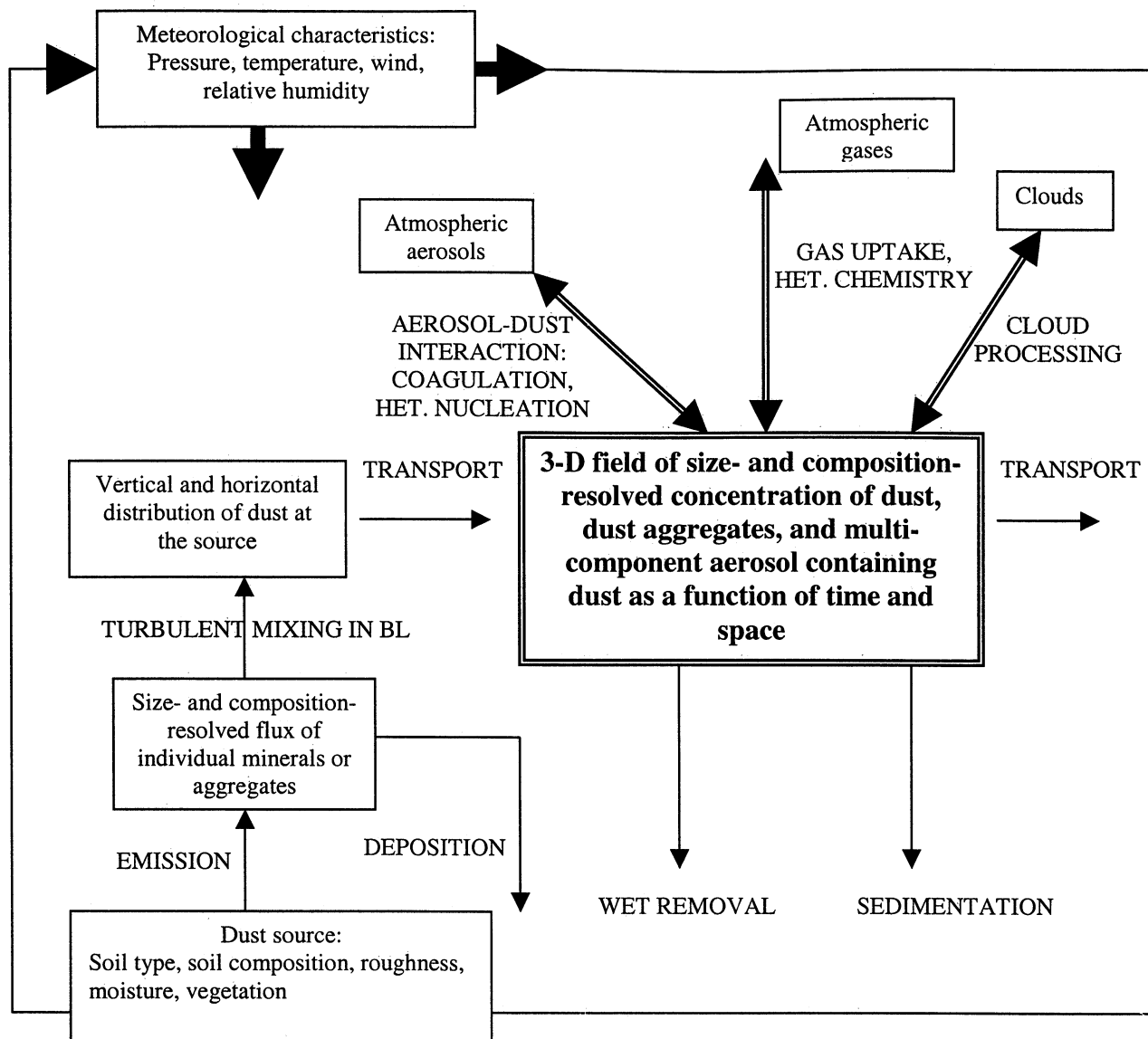


Figure 2. Processes governing the evolution of the physical and chemical properties of dust in the atmosphere.

simulated with size-resolved particle concentration, then the optical properties and radiative effects are calculated following the procedure illustrated in Figure 1 for a given refractive index [e.g., *Woodward*, this issue]. If the model predicts total dust mass concentration as a function of time and space, then the optical depth can be deduced assuming a fixed mass extinction coefficient [*Liu and Westphal*, this issue]. No radiative transfer calculations can be performed in the latter case.

Each of the above steps has a number of challenges that make this problem very complex. For example, in spite of having specified a monthly average dust source, the recent IPCC (Intergovernmental Panel on Climate Change) model intercomparison exercise found that the annual average dust burdens ranged from 0.4 Tg to 3.2 Tg for particles with a diameter of $D < 2 \mu\text{m}$ and from 0.5 Tg to 2.6 Tg for $D > 2 \mu\text{m}$ [*Penner et al.*, 2001]. This large variation must be attributed to differences in model treatment of wet and dry deposition. Furthermore, none of the models were able to adequately represent observed optical depths. As a result, uncertainties in the present estimates of the radiative impacts of dust remain

3. Outstanding Issues in Quantifying Radiative Impacts of Mineral Dust

Several strategies could be suggested to overcome the existing problems with measurements and modeling of dust impacts. The ultimate goal is to link the properties of dust to its source, production mechanisms, and life cycle, i.e., quantify the evolution of the chemical and physical properties and hence radiative effects of dust during its life cycle. The key processes, which need to be accounted for, are illustrated in Figure 2. The ability to carry this out will require resolving certain key problems discussed below.

3.1. Issues in Quantifying Dust Sources and Production Mechanisms

Despite the large diversity of dust sources, a common feature is that mineral dust is mainly injected in the atmosphere during sporadic events of high intensity [e.g., *Prospero*, 1996]. This is caused by emission processes that involve interactions between wind and surface features which exhibit a large spatiotemporal

Table 3. Mechanisms of Dust Production by Wind Erosion

Mechanism	Effects
Meteorological	
synoptic scale	surface winds: momentum source of erosion regional high winds
mesoscale	modification of wind by topography, thermally driven circulations
localscale	small-scale wind erosion, e.g., dust devils (not persistent) or small scale topography (persistent)
Air-particle interactions	
wind speed pdf and threshold wind speed	
airborne particle effect on flow (Owen effect)	aerodynamic roughness height (z_0) changed by airborne particles
Interactions of surface/air	
threshold friction velocity	
size distribution of particles	threshold friction velocity u_{*t} on a smooth surface is minimum (~22 cm s ⁻¹ for 60-120 μ m particles)
momentum partitioning	increase of u_{*t} with z_0 effect of clasts, vegetation and microtopography
drag coefficient	drag coefficient increases with z_0
Changeable physical properties of surface/particles	u_{*t} , z_0 changed; mass flux changed
aggregation- disaggregation	
crusting/crust destruction	
trapping	
particle supply limitation	
soil moisture	

After *Gillette* [1999]

several orders of magnitude) in atmospheric dust concentrations over short time periods. Because of the short lifetime of mineral dust in the atmosphere (about 2 weeks), which does not allow complete mixing at a global scale, an estimate of atmospheric dust emissions requires a relatively high resolution in time and space

Surface soil properties, vegetation, and production mechanisms are key factors determining the emission of dust for specific meteorological events. They all must be included for dust production schemes to be realistic. We consider the mechanisms and properties listed in Table 3 to be the most important in predicting wind erosion, and they are at present poorly specified in models.

What is known in dust emission modeling is largely from microscale experimental and theoretical studies. Relations of saltation sand flux to wind stress for sand sheets have been well described theoretically and experimentally. The effect of soil roughness elements reducing the sand flux has been modeled quite well along with the dominating physical mechanisms that control the threshold velocity for dust emission. Less well known (but known enough for simple models) is the relation between sand flux and dust emission by sandblasting. Using this knowledge, dust models have been constructed which have been partially successful in predicting mineral dust concentrations [e.g., *Marticorena and Bergametti*, 1995; *Marticorena et al.*, 1997a, b; *Shao and Leslie*, 1997]

However, several serious problems remain. One of the crucial issues is a lack of input data. Because of this lack, the above models operate well only for limited areas (Saharan and Australian deserts). Data are largely lacking for surface features such as roughness length or soil types. The best way to progress significantly is probably in the development of relationships allowing us to link surface properties to selected aircraft and satellite observations.

Another issue is that some physical processes are poorly or not-at-all included in the dust emission models. For instance, the emission processes of supply limited material or crusted surfaces are not well represented in these models. Because crusting and

supply limitation are very frequent in semiarid areas, modeling of these areas is presently unsatisfactory. Examples of the effects of crusting and supply limitation are given by *Gillette et al.* [this issue].

Crusting and supply limitation are clearly related to soil composition of the source. There is a clear need for a new data set to provide information on size distributions and composition of soils on global and regional scales. Existing global data sets of soil properties currently include soil texture (three size classes: clay, silt, and sand) and soil types, but they do not provide information on size-resolved mineralogical composition of the soil [*Webb et al.*, 1991]. In a recent paper by *Claquin et al.* [1999], a first attempt has been made to characterize the mineralogical composition of dust sources on a global scale by relating mineralogical experimental data and types of surface soils in arid areas.

In the same way, the effect of vegetation and its change over time (especially seasonal change, such as observed in Sahelian area) is not directly included in these models. The effect of vegetation has been successfully modeled by approximating vegetation as solid hemispherical roughness elements on the soil [*Stockton and Gillette*, 1990]. For randomly placed small vegetation, the effect has also been modeled by considering the vegetation to primarily cause a change in aerodynamic roughness length [*Marticorena and Bergametti*, 1995]. However, for some vegetation, random placement is not observed, and the above models do not apply. For improved models of dust emission, better parameterizations of vegetation effects on wind erosion must be developed. For global scale climatic models, the approach will require (1) coupling between dust emission models and vegetation models in order to simulate vegetation characteristics and their change with climate and (2) studies allowing a better parameterization of the effect of vegetation in terms of protection of the surface (increase of roughness length and surface cover)

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One more critical point concerns the relation of the vertical flux of fine particle mass to the horizontal mass flux of the coarser airborne particles. *Shao et al.* [1993] expressed the

particle flux of dust as a primary function of the flux of saltating particles, and binding energy of fine particles held in aggregation in the soil. Shao theory was consistent with data on the ratio of the vertical flux of dust mass to the horizontal flux of sand of *Gillette et al.* [1997]. Another approach following the lines of the Shao theory has been proposed by *Alfaro et al.* [1997]. In their theory the size distributions of the saltating particles are vitally important. The kinetic energy of individual saltating grains, along with the kinetic energy required to release suspendible particles, determines the quantity and size distribution of the suspended aerosol produced by sandblasting [*Alfaro and Gomes*, this issue]. Although it seems that the sandblasting theory describes the general features of dust fluxes, an improved knowledge is needed.

Finally, it must be stated that to sort out the climate effects of dust, we must be able to quantify an anthropogenic fraction of total dust loading. At the present time, this is uncertain by at least an order of magnitude [*Tegen and Fung*, 1995]. Various human activities can extend the geographical area of dust sources and increase dust loading into the atmosphere. For example, in the United States the highest dust production occurs in regions where the soil has been disrupted by cultivation and other intensive land uses (e.g., the dust bowl years in the 1930s and 1950s). In turn, future climate change caused by human activities may affect the emission and distribution of dust because of the dependence of dust emissions on climatic parameters such as wind speed and rainfall. In particular, dust emission strength is a function of the third power of the wind friction velocity, indicating that small changes in wind speed will be strongly amplified in terms of dust emissions. Therefore the fraction of the mineral dust particles which results from both the man-made climate change and the intensive land use (or other direct dust production resulted from human activities) should be considered as an anthropogenic climate forcing factor. However, this issue has yet to be studied quantitatively. Perhaps it will be possible to use model simulations and satellite observations together with historical data to identify the impact of humans. In particular, ice core and marine sediments data can provide valuable information on dust in past climate [*Chylek et al.*, this issue; *Hinkley and Matsumoto*, this issue].

3.2. Issues in Linking Dust Microphysics and Optics

The physical and chemical properties of dust particles, determined by a given source and production mechanism, evolve during the dust life cycle. The extent to which these variations are important for dust radiative impact is poorly quantified. Two main reasons why we have not been able to establish reliable empirical relations between the radiative impact of dust and its properties are the following: deficiencies of individual measurements and missing coordination among measurements of optical properties and dust composition, size distribution (and other physical and chemical characteristics), and dust radiative effects as a function of time and space.

Referring again to Table 2, improvements are sorely needed in the techniques used to determine the size, composition, mixing state, and shape of dust particles from ground-based and aircraft measurements. In particular, measurements of size-resolved mineralogical and chemical composition of dust particles in a size range from about 0.01 μm to 100 μm in coordination with dust optics measurements are urgently needed. Although chemical composition cannot be used to directly calculate optical properties, this information is useful to characterize dust sources, transport routes, and chemical evolution during the life cycle [*Choi et al.*, this issue; *Zhang et al.*, this issue, *Vaughan et al.*, this issue].

One of the promising approaches to address this issue is bulk and individual particle analysis based on a combination of X-ray diffraction, X-ray fluorescence, and electron microscopy techniques (scanning and transmission microscopes). These techniques enable the determination of size-resolved mineralogical composition, mixing state, and particle shapes [*Koren et al.*, this issue; *Ganor and Foner*, this issue; *Gao and Anderson*, this issue]. Yet some improvements in these techniques will be required to better quantify size-resolved mineralogical composition [*Falkovich et al.*, this issue]. Another limitation is that individual particle analysis gives only a two-dimensional image of dried particles of $D < 10 \mu\text{m}$.

Systematic studies of individual dust particles should be aimed at identifying major mineral species, determining whether they are aggregated with other aerosol species and characterizing the particle shapes. This analysis should be performed for dust samples collected at the ground and from aircraft at various distances from the dust source to characterize the evolution of dust particles during transport under specific atmospheric events.

These experimental data are urgently needed to identify the appropriate theory to model the optical characteristics of irregular dust aggregates. Currently, calculations of dust optical properties are done using Mie theory or, in a few cases, assuming spheroidal particles. In the case of mineral dust Mie theory is hardly reliable because particles can be made of several minerals (or other aerosol species) and have irregular shapes [*Kalashnikova and Sokolik*, 2000]. Improved analytical and numerical methods for predicting scattering from nonspherical particles are currently available [e.g., *Mishchenko et al.*, 2000]. However, empirical information on shape and composition of dust aggregates is largely missing.

To constrain model predictions of dust optics, well-coordinated measurements of dust optical properties along with dust morphology and composition will be required. In particular, laboratory measurements of optical properties of nonspherical dust aggregates of controlled particle size, shape, and composition could be helpful.

Another critical issue is how to adequately measure light absorption and scattering by mineral particles. New instruments must be developed to measure the scattering phase function of dust particles under atmospheric conditions. Also, new techniques to measure in situ absorption by dust are urgently needed.

Given the current limitations in techniques to measure aerosol scattering and the current inability to directly measure the absorption of the ambient aerosol, techniques to measure the effects of absorbing aerosols on the atmospheric radiation field gain added importance. A technique combining coordinated satellite radiometers and ground-based Sun photometers has been demonstrated [e.g., *Tanré et al.*, this issue]. Improved techniques based on new satellite observations need to be developed (see section 3.4).

3.3. Required Improvements of Dust Treatments in Climate and Chemistry Models

Process-oriented models represent the most comprehensive tool available for evaluating the impact of aerosols on chemistry and climate. To be realistic, the models will need to include explicit information on the particle size distribution and mineralogical composition of dust to allow for better physically based treatments of key dust processes shown in Figure 2. This information is crucial to adequately predict dust impacts on both Earth's energy budget and atmospheric chemistry.

Improved dust emission algorithms are required to provide

input for model transport schemes, as discussed previously. Another important consideration is the initial height to which the particles are lofted. Particle size distribution and vertical extent in the source region are critical factors in determining transport characteristics and dust impacts. Surface velocities and vertical lofting processes are very sensitive to model resolution, and further research is needed to quantify resolution issues and to build scale-dependent, sub-grid-scale parameterizations [Liu and Westphal, this issue].

In addition to quantifying dust sources, the development of physically based treatments of dust requires quantification of the rate of the processes governing the evolution of mineral particles during their transport in the atmosphere. The relative role of cloud processing, heterogeneous chemistry, dust-aerosol interaction, and size- and composition-resolved removal processes must be quantified. This is a difficult problem due to the nonlinearity of the processes, various feedbacks, and the overlap in timescales between aerosol processes and atmospheric dynamics. A promising strategy is to perform comprehensive coordinated measurements of the physical and chemical properties of dust in targeted regions, on various space and timescales, as we discuss below.

The need to calculate the chemical interactions with dust adds further complexity to model development. In particular, it creates demand for additional data. For example, the calcium content of the dust plays a key role in constraining the gas-aerosol partitioning of semivolatile inorganic components such as ammonia and nitric acid, while the reactivity of the particle depends on the mineralogy. Thus models must track the composition of key species in the primary aerosol, thus creating additional needs from the emissions and computational viewpoints.

Assessing chemical interactions with dust particles is important because processes, which alter the water uptake properties of the dust, provide new pathways for chemical conversion, and/or effect the partitioning of species between the gas and the aerosol phases, result in modification of the size, chemical composition, and radiative properties of the ambient aerosol. To account for these effects, size-resolved inorganic aerosol modules that consider aerosol dynamics, surface chemistry, and gas to particle exchange must be developed [Song and Carmichael, this issue].

The diverse radiative effects of dust may result in various feedbacks on the climate system, which are potentially important but hard to predict and quantify [Perlwitz *et al.*, this issue]. To address the tight couplings between processes linking dust emissions and feedbacks caused by dust impacts, it will be necessary to represent aerosol processes and forcing "online" in models in order both to represent correlations between aerosol loading and forcing and the pertinent meteorological variables, such as cloudiness and humidity, and to capture the feedbacks of aerosols on the climate system. The correlation between dust loading and local source resolution is especially important. For example, the use of a monthly average source term for dust can lead to a factor of 2 error in the total abundance of dust [Penner *et al.*, 2001]. Furthermore, as coupled models are developed, a strategy for validating the model's temperature response to dust loading is needed. At the present time, model simulations using the GISS GCM have shown that the surface temperature is cooled below the main dust cloud, but the amount of cooling is reduced where convection is strong [Miller and Tegen, 1998].

Well-coordinated observations are necessary to test the assumptions on which models are based. In situ measurements are required for validation of many of the parameterizations built into these models. The global transport of aerosols is highly dependent on the vertical mixing predicted by the

models. Ground-based and satellite lidars are the only means of testing model predictions of vertical distribution on a global basis. Also, satellite observations of global aerosol distribution and loading are required for testing the overall model predictions. However, any comparison of satellite and model data involves assumptions about dust radiative properties, which are highly uncertain. There are problems in how models should assimilate the dust properties retrieved from satellite observations. One assimilation issue (currently unsolved) is whether the model should assimilate retrieved optical properties or the observed radiances themselves. Satellite aerosol retrievals all rely on certain assumptions. If satellite radiances were assimilated directly, these assumptions would become part of the model. With the current state of the parameterization of aerosol processes in models, it is not clear that useful additional constraints could be provided by the model.

3.4. How to Make the Best Use of Remote Sensing of Dust

Satellites with new capabilities, which were specifically designed to study atmospheric aerosols, have been recently launched or will be launched soon (see review by King *et al.* [1999]). The data to be obtained from these new satellites promise to greatly improve our understanding of the properties of mineral dust.

The ADEOS satellite, launched in 1996, had the first passive space sensor (a polarization-sensitive charge-coupled device camera, POLDER) designed for aerosol observation over both ocean and land [Deschamps *et al.*, 1994]. POLDER combines polarization, several spectral channels below 1 μm , and two-dimensional multiangle observations to derive the aerosol optical thickness and a measure of the aerosol size over the ocean and an aerosol index over the land [Herman *et al.*, 1997b]. The advantage of polarization measurements is that they provide additional sensitivity to the aerosol refractive index over the ocean and sensitivity to submicron aerosol particles over the land despite the higher surface reflectance. Dual-view observations are also collected by the Along-Track Scanning Radiometer (ATSR) [Veefkind *et al.*, 1998]. A combination of angular, spectral, and polarization capability over a given footprint was suggested on EOSP [Mishchenko and Travis, 1997] but awaits implementation by NASA in space.

The new NASA Earth Observing System (EOS) Terra satellite, launched in December 1999, has two instruments obtaining aerosol measurements: the Moderate-Resolution Imaging Spectroradiometer (MODIS) and the Multiangle Imaging Spectroradiometer (MISR). MODIS has eight well-calibrated spectral channels in the solar spectrum between 0.41 and 2.1 μm which are used for aerosol retrievals [Tanré *et al.*, 1997; Kaufman *et al.*, 1997]. MISR acquires observations at nine angles and at four wavelengths in the visible and near IR [Martonchik and Diner, 1992; Kahn *et al.*, this issue]. A combination of MODIS spectral and MISR angular data is expected to improve the characterization of the aerosol size distribution and single scattering albedo and thus to retrieve much more accurate aerosol optical thickness and mass loading. It is expected that the accuracy of the optical thickness derived from MODIS on Terra will be $\Delta\tau = \pm 0.03 \pm 0.05\tau$ over the ocean and $\Delta\tau = \pm 0.05 \pm 0.2\tau$ over the land. Measured radiances from MODIS will also be used to derive the aerosol impact on the radiative spectral fluxes at the top of the atmosphere, probably with accuracy higher than that of the optical thickness. A combination of aerosol properties from MODIS with total solar and IR fluxes measured by the CERES instruments (also on Terra) will enable direct measurements of aerosol radiative

forcing, at least in regions with significant aerosol concentration (e.g., optical thickness larger than 0.2).

Following the successful demonstration of space lidar by LITE, two long-term space lidar missions are now planned. The first long-term lidar system in space is expected to be IceSat in 2001 with the primary objective of measuring the height of ice and snow layers but with some ability to measure aerosol with a one-channel lidar. A two-wavelength polarization lidar system dedicated to atmospheric investigations and optimized to detect the vertical distribution of aerosol and their properties is planned to fly in 2003 on the PICASSO-CENA mission [Winker and Wielicki, 1999]. PICASSO-CENA will fly in formation with the EOS-Aqua satellite. A combination of multi-spectral passive measurements of the entire aerosol column from MODIS on EOS-Aqua with near-simultaneous lidar measurements in two spectral channels has the potential to identify the presence of layers of aerosol in the atmosphere along with their specific properties and interaction with cloud layers.

Despite the great recent progress in the capabilities of satellite remote sensing, it is important to point out that satellite data are inherently limited in the information they can provide on dust (e.g., dust chemistry, morphology, and relationship to sources). Satellites by themselves cannot measure all of the aerosol characteristics required. Therefore to benefit from the wealth of data to be provided by new space aerosol sensors, a complete strategy must include a combination of satellite and ground-based remote sensing with in situ measurements and modeling efforts, as discussed below.

3.5. How to Properly Combine Measurements and Modeling

The innovative combination of a well-planned network of surface stations and short-term aircraft measurements, coordinated with satellite platforms, would be the optimum approach to fulfill the need for a global data set of dust properties. Yet challenges remain in relating dust climatology to the processes controlling the evolution of dust at all relevant spatial/temporal scales needed for chemistry and climate models.

A feasible approach to address this problem is to conduct regional closure experiments at targeted geographical locations. These experiments should go beyond quantifying what is there to identifying and quantifying the mechanisms responsible for the evolution of the physical and chemical properties of dust. Each targeted area should be centered on a particular dust source and extended to the distances to which dust can be transported over a 1-week time period. The regions of main interest are the North Sahara – Atlantic Ocean; North Sahara – Mediterranean Sea; Arabian Peninsula – Indian Ocean; central Asia- North Pacific; Australia – Tasman Sea; and the southwestern United States.

Dust properties must be studied at regional scales to elucidate the link between the given source and production mechanism and the initial size-resolved composition of dust particles. Initial properties of dust along with specific transport routes (e.g., clean air, urban polluted air, humid marine environment) will further determine the possible chemical and physical evolution of dust. For instance, in contrast to Saharan dust, Asian dust, originating in the arid zones of China and Mongolia, might be transported through polluted industrial urban regions and hence interact with abundant anthropogenic sulfates and soot prior to reaching the marine environment [Parungo *et al.*, 1995]. Thus it is important to establish the relative roles and characteristic scales of various processes affecting the properties of dust in a given region. Understanding of the unique temporal and spatial variations of dust in each region is of special importance to better quantify the regional as well as global impacts of dust.

These tasks will require the coordinated efforts of interdisciplinary researches to perform the comprehensive measurements of diverse dust properties at the selected locations. In particular, well-coordinated measurements of dust chemical, physical, and optical properties and related radiative effects at various space scales and timescales are highly desirable. The ultimate goal is to integrate collected experimental data sets to derive physically based parameterizations of dust chemical, physical, and radiative properties needed for climate models and for satellite retrievals.

There is a strong demand for new techniques capable of interpreting and merging the various data provided by new satellite instruments, in situ and ground-based observations, laboratory measurements, and state-of-the-art models. It is probable that data assimilation techniques developed in different disciplines could be modified for dust applications. For instance, four-dimensional (4-D) (latitude, longitude, altitude, and time) assimilation of meteorological characteristics (such as temperature, pressure, etc.) has been successfully used in weather forecasts. Assimilation methods have also been developed and used for atmospheric gases. However, mineral dust as well as other atmospheric aerosols, by virtue of their nature, are more complicated to assimilate into the models. Thus new approaches are urgently needed to take full benefit of the better data and improved models becoming available in the near future.

4. Summary

Recently, there have been substantial advances in our understanding of mineral aerosols and their impacts on the climate system as demonstrated by the collection of papers in this special section on mineral dust. However, large uncertainties in the prediction of dust radiative impacts still exist. In this paper we have highlighted the various outstanding issues and have recommended a few strategies for future research.

The sources of the current uncertainties involve deficiencies in our understanding of physical and chemical processes as well as the lack of adequate data. In particular, a better understanding of the main processes governing the emission, transport, removal, and evolution of dust is required. Examples of poorly understood processes that would make critical contributions include dust emission via saltation and sandblasting mechanisms at a variety of scales, interactions of dust with other atmospheric aerosols, gases, and clouds, and heterogeneous chemistry on dust particle surfaces. These are difficult problems due to the nonlinearity of these processes, possible feedbacks, and the overlap in timescales between aerosol processes and atmospheric dynamics. In addition, model predictions of dust processes are hard to verify because available data on dust characteristics are still woefully inadequate. Improvements are sorely needed in the methods employed to determine the composition of individual particles and their morphology, light absorption, and scattering phase function.

To summarize our discussion, we identify the high-priority research needs. Table 4 lists these needs grouped into three broad categories: dust emission and sources, dust properties, and measurements and modeling capabilities. It is clear that accomplishing these tasks will require a great deal of effort. We believe that a promising strategy is to perform comprehensive coordinated measurements of the physical and chemical properties of dust in targeted regions. Elucidating the linkage between dust impacts and dust properties evolving during transport under various atmospheric conditions (such as clean air, polluted air, marine environment) is urgently needed.

Table 4. High-Priority Research Needs

	Research Needs
Dust emission and sources	develop dust production schemes for size- and composition-resolved fluxes accounting for soil features (such as roughness, texture, composition, moisture), vegetation, and land use develop a new data set of surface properties needed for improved dust production schemes
Dust properties	quantify the anthropogenic fraction of dust burden and its trends elucidate the linkage between morphology (size and shape), mineralogical composition of dust particles and their optical properties quantify the relative role of processes governing the evolution of the physical and chemical properties of dust during transport in the atmosphere
Measurement and modeling capabilities	develop new instrumentation (e.g., to measure dust particle size distribution in a range from ~0.01 to 100 μm ; scattering phase function and light absorption by dust particles) design and implement comprehensive field studies in selected regions develop new techniques for interpretation of diverse data being available from satellite, in situ and ground-based observations, laboratory studies, and state-of-the-art models

Although this paper focused on dust radiative impacts, the importance of other diverse impacts of dust upon the environment and the overall climate system should not be underestimated. Several other major impacts have been pointed out in section 2.1. Because of numerous interlocking issues across various disciplines dealing with dust, integrated analysis of climate and socioeconomic systems will eventually be required to fully address this problem. One way this might be achieved is by incorporating mineral dust into integrated assessment modeling. Recently, the Integrated Assessment Models have focused primarily on the greenhouse gases buildup [e.g., *Alcamo*, 1994]. However, various human activities, land cover change, certain industries, and climate change (especially changes in wind speed and precipitation) all might extend the geographical area of dust sources and increase dust loading into the atmosphere. Because the presence of both anthropogenic and natural dust may rival or enhance the radiative forcing of greenhouse gases on the climate system as well as pose a health threat, affect agriculture production, etc., it is important to consider dust impacts in overall assessments of policy and cost options concerning the climate-change impacts.

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