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Recent progress in understanding physical and chemical properties of mineral dust

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Abstract

This paper presents a review of recently acquired knowledge of the physico-chemical properties of mineral dust from Africa and Asia based on data and discussion during the Third International Dust Workshop, held in Leipzig (Germany) in September 2008. Various regional experiments have been conducted in the last few years, mostly close to source regions or after short-range transport. Significant progress has been made in characterising the mineralogy of iron and the description of particle shape close to source regions. We consider that future research should focus on the evolution of dust properties during transport. In particular, the prediction of the characteristics of the size distribution of mineral dust at emission and their dynamics evolution during transport should be considered as a high-priority. The presence of large airborne particles above 20 µm in diameter, suggested by various studies, has limited implication for radiation, but should be important for deposition-based studies such as those on ocean productivity. Modelling the uplifting and transport of such large particles will represent a new serious challenge for transport models. A critical assessment and standardisation of the experimental and analytical methods is recommended.

1 Introduction

The most recent model estimates indicate that global mineral dust emissions by wind-driven erosion over arid and semi-arid areas range between 1000 and 3000 Tg yr^{-1} (Zender et al., 2004; Cakmur et al., 2006; Textor et al., 2007), representing about half of the annual particle emission at the global scale (Forster et al., 2007).

The global dust belt, where dust sources are located, extents from the west coast of North Africa, through the Middle East, into Central Asia and covers the Sahara, the deserts of the Arabian Peninsula and of Oman, Caspian Sea and Aral Sea regions in Central Asia, and Gobi and the Taklimakan in China. Sources outside the global dust belt are located in the USA and Mexico, Lake Eyre and the Great Artesian Basin in

Australia, Makgadikgadi Depression and Pans in Botswana and the Namibia desert, both situated in Southern Africa, and Southern American deserts Altiplano in Bolivia, as well as desert areas in Patagonia and in Western Argentina.

Once in the atmosphere, mineral dust plumes influence the global climate. Dust interacts with radiation by scattering and absorption in the visible and thermal infrared spectral region (Myhre and Stordal, 2001; Balkanski et al., 2007) and may serve as cloud condensation nuclei (CCN) or ice nuclei (IN) (DeMott et al., 2003; van den Heever et al., 2006). Dust also plays a crucial role in fertilizing large areas of the oceans by deposition and subsequent dissolution (Duce et al., 1991; Jickells and Spokes, 2001). Furthermore, mineral dust has a potential impact on human health directly (e.g., Kwon et al., 2002; Perez et al., 2008) and via the spreading of microorganisms (Kellogg and Griffin, 2006), on regional air quality via the visibility impairment (Prospero, 1999) and on the atmospheric composition through heterogeneous reactions (Bauer et al., 2004; de Reus et al., 2005). An example of environmental impact is the demise of Caribbean Coral Reefs, as suggested by Shinn et al. (2000).

As for any type of atmospheric aerosol particles, the environmental and climatic impacts of mineral dust depend on its physico-chemical properties, that is, composition, shape, surface state, size, and mixing state of the particles (Raes et al., 2000b). This link is made explicit in Table 1.

Mineral dust consists of irregular particles, often aggregates of different composition, of sizes varying from tenths of nanometers to hundreds of microns. The size distribution evolves rapidly according to time after emission (Pye, 1987).

Particle size is a fundamental parameter to understand and predict atmospheric lifetime, transport processes and impacts of mineral dust. The representation of the mineral dust size distribution remains a real challenge due to the large size spectrum. The number size distribution, dominated by sub-micron particles, controls the direct impact on radiation and on cloud processes (Sokolik et al., 2001; Dusek et al., 2006). The mass size distribution, dominated by supermicron particles, controls deposition, hence the biogeochemical effects, as well as interactions with the gas phase (Trochkine et al.,

2003; Zhang et al., 2003a). The surface distribution controls radiative effects. Examples with respect to the direct radiative impact are given by (Otto et al., 2007; McConnell et al., 2010).

Particle shape influences the aerosol optical properties underlying the direct effect (Nousiainen, 2009). Deviation of dust grains from spherical shape can change light scattering by a factor of 2, depending on the scattering angle (Kalashnikova and Sokolik, 2002; Dubovik et al., 2006). The uncertainty in remote sensing of dust optical depth due to particle shape could be even higher due to the particle's refractive index (Kahnert et al., 2007). Particle shape, besides size and density, also influences particle sedimentation. In general, sedimentation velocity decreases with increasing deviation from spherical shape (Cheng et al., 1988). For example, platy particles can be transported over longer distances than spherical ones. In addition, with increasing non-sphericity the orientation of the particle in the air becomes a factor influencing the sedimentation velocity. While it is generally assumed that platy particles orientate horizontally, it was also suggested that due to an asymmetry of the centre of gravity, the particles may orientate with the heavier part pointing downwards (Li and Osada, 2007b,a).

Composition is the third key property. Sokolik and Toon (1999) demonstrated that the variable mineralogical composition (clays, quartz, carbonates, feldspars, sulphates, iron oxides) has to be incorporated into radiative models to estimate the dust optical and radiative properties. When first emitted, dust particles are often composed to a large extent of insoluble or low-solubility components. Referring to Koehler theory, mineral dust particles are considered almost CCN inactive (Kreidenweis et al., 2005). In contrast to this theory-based assumption, several experimental studies suggest that freshly emitted dust particles can serve as CCN or IN (Koehler et al., 2009; Connolly et al., 2009). The nucleation of cloud droplets on dust particles is most likely influenced by the mineralogical composition, notably on their calcite content for CCN ability (Gustafsson et al., 2005; Gibson et al., 2006), and clay content (kaolinite and illite) for IN efficiency (Zimmermann et al., 2008). The iron chemistry is important as it can di-

rectly affect the radiative budget and the marine ecosystem productivity (Claquin et al., 1999; Sokolik and Toon, 1999; Meskhidze et al., 2005; Luo et al., 2005; Fan et al., 2006; Balkanski et al., 2007). More recently, the titanium oxide (TiO_2) has gained in importance in photochemical induced heterogeneous chemistry reactions. As an example, Ndour et al. (2008) showed that in that under a UV flux representative in intensity of atmospheric conditions, NO_2 was uptaken at the surface of dust and lead to the production of HONO.

Many modelling studies point out to the uncertainties which still affect the detailed knowledge of the physico-chemical properties of mineral dust has a limiting factor in estimating their climatic impact (Sokolik et al., 2001; Balkanski et al., 2007; Nousiainen, 2009; Claquin et al., 1998; Myhre and Stordal, 2001). Besides inherent limitations in the in-situ and remote sensing instrumentation, the description of the physico-chemical properties in chemistry and climate models suffer from a lack of understanding of the processes underlying the dust emission and evolution in the atmosphere.

In this paper, we present a review of the current state of knowledge of the physico-chemical properties of mineral dust relevant to estimating its main impacts. The paper reports on the scientific discussion which has taken place within the Third International Dust Workshop, held in Leipzig (Germany) in September 2008, and it is based on the results of the many field experiments which, in the last decade, have been dedicate to mineral dust: ACE-2 (Raes et al., 2000a), SHADE (Tanré et al., 2003), PRIDE (Reid and Maring, 2003), MINATROC (Balkanski et al., 2003), ADEC (Mikami et al., 2005), ACE-Asia (Huebert et al., 2003), BODEX (Todd et al., 2008); AMMA (Redelsperger et al., 2006), DABEX (Osborne et al., 2008), DODO (McConnell et al., 2008), GERBILS (Marsham et al., 2008), and SAMUM-1 (Heintzenberg, 2009). Acronyms are explained in Appendix 1. The need for focused regional closure experiments was highlighted by Sokolik et al., 2001) in the summary of conclusions of the First International Dust Workshop, held in Boulder in 1999.

The aim of this paper is twofold. First, a synthesis of field observations available in the literature is attempted. This cannot be achieved without a critical assessment of

the various experimental and analytical methods. Second, this paper tries to identify remaining gaps and priorities for future research.

By doing so, this paper also completes the recently published review by Redmond et al. (2010) on the physical basis, the instrumental techniques, and most recent observation of the optical properties of scattering and absorption by mineral dust.

2 Sampling and analytical techniques for the study of the physico-chemical properties of mineral dust

Three principal limitations have to be kept in mind when comparing literature data on physico-chemical parameters of mineral dust. First, most of the data found in the literature represent background to haze conditions (concentrations up to 1000 µg m^{-3}). Dust storm conditions are rarely investigated, because a) the source regions are often very remote and hard to access, b) aircraft sampling is prevented by reduced visibility during high dust conditions, and c) state of the art instrumentation may be easily damaged by the extreme atmospheric conditions. An illustration of these difficulties is given by Todd et al. (2008) reporting on the setting up of the BODEX experiment downwind the Bodélé depression in Northern Chad. Another difficulty when sampling mineral dust is related to the episodic character of the emission. At a receptor site in the African Sahel, in Niger, Rajot et al. (2008) have shown that sample exposure upon alert and on times varying from ∼20 min to few hours according to the indication of a simultaneous measurement of total mass had to be undertaken in order to collect samples sufficiently loaded for elemental analysis but at the same time well within the duration of each episodes in order to have the possibility of discriminating their different source region based on composition.

Second, a major problem of mineral dust sampling in field measurements is the wide size range, in particular the sampling of supermicron particles which constituted the larger fraction of the mass (Pye, 1987). In ground-based studies, measurements are performed downstream inlet systems with certified cut-off diameters as small as 2.5 µm

or 10 µm, or with uncertified Total Suspended Particulate (TSP) samplers (Reid et al., 2008; Kandler et al., 2009). Wind-oriented high-volume inlets with high transmission efficiencies are started being used (Rajot et al., 2008; von der Weiden et al., 2009). Recently, dedicated inlet systems for aircraft sampling were developed to improve the even more critical sampling at high air speeds (Wendisch et al., 2004), e.g. the CARIBIC inlet (Hermann et al., 2001), the Big Particles Sampler (Levin et al., 2000), a shrouded inlet system (Dhaniyala et al., 2003), or the PELTI (Huebert et al., 2004).

Third, the available bulk and size-segregated sampling methods are based on different aerosol properties (e.g., optical, inertial, electrical), which depend on composition, shape and size. As a consequence, a meaningful comparison of measurements by techniques based on different principles (e.g., number size distribution measurements by optical and aerodynamical methods) requires the knowledge of additional physico-chemical properties (e.g., composition, particle density and shape, Hinds, 1999). When these properties are not measured in parallel, assumptions have to be made to reconcile their measurements.

2.1 Measuring techniques to assess size distribution

The number size distribution of airborne dust particles in the full diameter range from approximately 100 nm to 100 µm can be determined by optical methods such as light scattering techniques or imaging techniques such as electron microscopy.

Light scattering techniques measure the optical equivalent diameter referring to a sphere of given refractive index, which scatters the same amount of radiation into a given solid angle and which has the same volume as the dust particle. The composite refractive index of dust particles is determined from chemical composition analyses combined with appropriate mixing rules (Kandler et al., 2007, 2009; Lafon et al., 2006; Wang et al., 2002) or by a combined analysis of optical and microphysical data (Osborne et al., 2008; Petzold et al., 2009). Particle non-sphericity may cause deviations <10% for dust particles >1 µm by optical particle counters which measure light scattered in the forward to sideward regime (Osborne et al., 2008). The ratio of non-

spherical to spherical particle scattering phase functions is 1.1 for optical particle counters measuring predominately forward-scattered light (Collins et al., 2000), resulting in a slight oversizing if particle sphericity is assumed in the data analysis. Considering realistic dust particle shapes, the deviation of the scattering phase function from that of spheres may increase up to 20% (Reid et al., 2003c). Furthermore, light scattering methods suffer from a limited size resolution at larger diameters due to the fact that the instruments response for super-micron particles is relatively flat, so that uncertainties in particle refractive index and shape may introduce large errors.

Geometric sizing methods are based on particle collection by filtration or impaction followed by individual particle characterization by transmission (TEM) and/or scanning electron microscopy (SEM) (Baron and Willeke, 2001). Individual particle analysis measures properties like the two-dimensional projection area, the volume to surface ratio (Sauter mean diameter) or the gyration diameter, with uncertainties due to pixel resolution, counting statistics, mass losses due to volatility or water evaporation during analysis under high vacuum (Kandler et al., 2009; Baron and Willeke, 2001). This approach also reduces the difficulty in relating the number and the mass size distribution.

Electrical and inertia-based measurements can be used to determine different fractions of the number size distribution. Both methods avoid uncertainties related to assumptions of a distinct particle shape and of an effective complex refractive index. The submicron fraction can be sized in terms of the electrical mobility diameter of a charged particle moving in a static electric field, as done by Differential Mobility Sizer (DMA) or Differential Mobility Particle Sizer (DMPS) methods, respectively. The application of these methods is usually limited to the sub-micron diameter range. Therefore, DMA methods are used in combination with optical particle counters (e.g., Haywood et al., 2003a; Weinzierl et al., 2009; de Reus et al., 2000; Clarke et al., 2001) or with aerodynamic particle sizers (APS; e.g., Wang et al., 2002; Maring et al., 2003). measuring the diameter of a sphere of unit density having the same terminal velocity in an accelerated airflow as the irregularly shaped dust particle (Peters, 2006). A detailed analysis of factors influencing the dust size distributions by means of an APS identified uncer-

tainties in the dynamic shape factor, particle density, and inlet transmission efficiency for different particle sizes. The key conclusion is that APS methods tend to undersize dust particles by at least 10–30% (Reid et al., 2008) even if a dynamic shape factor of 1.10 to 1.25 (Kaaden et al., 2009) is applied. Despite these limitations, APS methods are capable of covering a particle diameter size range form 0.7 µm to 20 µm.

The mass size distribution is measured by multi-stage filtration or impaction sampling coupled with gravimetric or chemical analysis (Ichoku et al., 1999; Formenti et al., 2001a, 2003; Lafon et al., 2006). A combination of the impaction and filtration is also used by streaker sampling (Annegarn et al., 1996). Uncertainties are due to the efficiency of size-separation, due to particles rebound and shattering and accuracy in flow rate control (Hinds, 1999). Further, because the overwhelming fraction of the mass is in the super-micron fraction, the upper size limit and the exposure time have to be optimised in order to allow that all size classes are equally represented.

Comparing size distributions measured by different approaches requires an agreement on aerosol parameters which describe the dust size distribution adequately. The parameterisation of size distributions by multi-modal logarithmic normal distributions is one potential method, if size-binned information on dust particle size distributions is available. The dust particle mode is then characterised by the geometric mean diameter or count median diameter CMD, respectively, and the respective geometrical standard deviation or GSD, respectively (Haywood et al., 2003a; Weinzierl et al., 2009) or by the volume median diameter VMD and the respective geometrical standard deviation GSD (Reid et al., 2008). Whenever possible, a careful analysis of reported equivalent diameters was performed for the determination of reliable particle size distributions by different methods.

Since there is no “true” particle size distribution available, the accuracy of the collected data has to be evaluated carefully. One potential approach is the extinction closure, i.e., the extinction coefficient calculated from the size distribution is compared to a direct measurement of the extinction, e.g. by high spectral resolution lidar (Esselborn et al., 2009). Another approach investigates the discrepancies in measurements of light

extinction and extinction-to-backsatter ratio (lidar ratio) of desert dust with CALIPSO and ground-based lidar systems (Wandinger et al., 2010). If close agreement between both methods is achieved, then the methods used for the determination of the size distribution and the methods used for the calculation of the extinction coefficient are applicable. If the values disagree, then at least one of the two steps (size distribution measurement, extinction coefficient calculation) is not appropriate.

As part of the Saharan Mineral Dust Experiment (SAMUM), dust particle size distributions measured by optical particle counters and geometric sizing methods were evaluated against a direct measurement of the dust aerosol extinction coefficient for Saharan dust by Weinzierl et al. (2009) using refractive indices determined simultaneously (Petzold et al., 2009) and direct measurements of the extinction coefficient by high spectral resolution lidar (Esselborn et al., 2009). The authors report agreement within deviations of <15% between optical extinction from size distributions combined with a detailed determination of the dust refractive index and the application of Mie theory, and extinction measured directly. The finding that even the assumption of spherical particles does not cause a significant deviation from direct extinction measurements can be related to the fact that particle non-sphericity is mostly important for the aerosol backscatter regime (Lacis and Mishchenko, 1995; Mishchenko et al., 1997).

Wandinger et al. (2010) investigated the desert dust extinction coefficient measured by CALIPSO and compared them to measurements with a ground-based Raman lidar systems. The underestimation of dust extinction by CALIPSO by 30% is explained by the influence of multiple scattering which is ignored in the CALIPSO retrievals. Based on recent observations of the size distribution of dust particles from airborne in-situ observations during SAMUM (Weinzierl et al., 2009), model calculations show that the multiple-scattering-related underestimation of the extinction coefficient in the CALIPSO lidar signals ranges from 10–40%.

In another radiative closure study performed during ACE-2 (Collins et al., 2000), size distributions were again measured by optical particle counters, but did not cover the entire size range of dust particles due to inlet cut-off properties and the failure of

an instrument for sizing larger particles up to 100 µm in diameter. No closure was achieved between calculated extinction coefficients and data measured by sun photometry. These cut-off effects for large-particles are discussed as potential explanation for the discrepancies between measured and calculated optical properties.

As part of ACE-Asia, Wang et al. (2002) conducted clear-column radiative closures for different types of aerosols including mineral dust. The chemical composition of mineral dust was derived from impactor samples. Size distributions were measured by means of DMPS-APS instruments. The extinction coefficient derived from size distribution data and refractive index data using Mie theory was compared to extinction coefficient data from sun photometry. The authors report that in the presence of dust layers deviations >35% between measured and modelled extinction coefficients were found, indicating an undersizing of mineral dust by the APS.

Another potential method for data quality assurance is the mass closure, i.e., the mass concentration calculated from size distributions assuming spherical particles of mineral dust is compared to direct mass concentration measurements by gravimetry. This approach however suffers from the fact that dust particles are highly irregularly shaped. Furthermore, this method is applicable only for ground-based measurements because there are no reliable methods available for airborne measurement of particle mass concentrations.

Summarising the instrument intercomparison studies of the particle size distribution, in situ size distributions can be measured in a reliable manner by optical methods if several requirements are fulfilled: Each instrument requires individual calibration for a precise determination of the instrument response. The complex refractive index has to be known in detail for the inversion of OPC data; instrument response requires adjustment to the respective refractive index value. The effect of particle non-sphericity on particle sizing by light scattering methods has to be evaluated for each instrument individually according to the specific optical set-up of the respective instrument.

2.2 Measuring techniques to determine particle shape

The shape of atmospheric mineral dust is assessed mainly by two methods, microscopy and aerodynamic measurements. Only few aerodynamic shape measurements exist for ambient mineral dust (Kaaden et al., 2009). More data sets are available from microscopy measurements, where the size range of atmospheric importance is covered by electron microscopy techniques like transmission and scanning electron microscopy (TEM/SEM). Atomic force microscopy (AFM) is also used, but limited to particles smaller than 1.5 µm in diameter (Chou et al., 2008; Gwaze et al., 2007; Helas and Andreae, 2008).

The particle shape derived by microscopic techniques is usually derived from the two-dimensional outline of the particle. It is mainly expressed as aspect ratio (ratio between longer and shorter axis of an ellipse fitted to the particle outline) or as circularity (ratio of 4π times the particle area to the square of the particle perimeter) (Kandler et al., 2007, 2009). While the aspect ratio is a rather robust measure, the circularity is quickly affected by imaging defects and pixel resolution (Almeida-Prieto et al., 2007; Podczeck et al., 1999). For this reason the circularity is less suitable for a comparison. Finally, also fractal dimension is used as measure for dust shape (Koren et al., 2001).

Besides common measurement errors (e.g., sizing, aliasing effects, instrumental artefacts), some principal limitations apply for the shape measurement. Commonly, a strong simplification of the shape is performed, usually to an equivalent (in volume or in area) ellipse or spheroid. While this limitation is not inherent to the microscopical techniques, shapes are usually given in simplified form to serve as base data for optical calculations (e.g., Kandler et al., 2007). Measurements of shape by microscopy are usually performed two-dimensionally (e.g., Reid et al., 2003b; Kandler et al., 2007) with few exceptions (e.g., Okada et al., 2001), and assumptions are made about the third dimension, which may depend of the method of sampling.

Surface roughness affects the scattering phase function especially for particles larger than the wavelength (Nousiainen and Muinonen, 2007). However, only single measure-

ments of surface roughness by atomic force microscopy are available of soil material (Helas and Andreae, 2008) and airborne dust (Chou et al., 2008), but no systematic studies suitable for modelling purposes have been performed yet.

2.3 Measuring technique to determine particle composition

As shown in Table 1, mineral aerosol composition refers to the analysis of different elements depending upon the characteristics of the aerosol and its impact one is trying to assess. Different techniques can be used to highlight these different aspects of mineral aerosol composition.

The major mineral phases of dust (abundance >5% by mass) can be identified by X-ray diffraction (XRD) commonly used in soil science (e.g., Schütz and Sebert, 1987; Caquineau et al., 1998; Kandler et al., 2009). Owing to its elevated detection limits, the application of XRD to aerosol samples is however limited to loaded samples of mass higher than 800 µg of dust (Caquineau, 1997). Even at high concentrations reached for haze conditions ($\sim$1000 µg m^{-3}), this detection limit can only be achieved by high-volume sampling and relatively long exposure duration which might only be rarely achieved by aircraft sampling (Formenti et al., 2008).

The XRD technique is regarded as a semi-quantitative method due to the difficulty in finding appropriate mineral standards for calibration (Moore and Reynolds, 1997; Caquineau et al., 2002). This is true in particular for clay minerals, whose chemical composition in the aerosol phase can depend on environmental conditions such as weathering, of by cationic substitutions and impurities.

An alternative to the mineralogy, the elemental composition (frequently expressed as elemental ratios) is often used to characterize the dust composition (e.g., Chiapello et al., 1995). Elemental concentrations can be determined by X-ray fluorescence spectrometry (XRF), particle induced X-ray emission (PIXE), or instrumental neutron activation analysis (INAA) (Formenti et al., 2003, 2008; Reid et al., 2003b; Rajot et al., 2008; Zhang et al., 2009; Paris et al., 2010; Müller et al., 2010). These techniques have excellent detection limits for major mineral tracers (lower than 10 ng cm^{-2} for elements

from Al to Fe), allowing analysis of samples with low mass concentration. The well-known X-ray self-attenuation in supermicron particle grains affecting the accuracy of measured light-element concentrations by XRF and PIXE is corrected by using appropriate geological standards, empirical corrections or the complementary use of INAA (Cornille et al., 1990; Formenti et al., 2008, 2010). The dust chemical composition is also determined by inductively coupled plasma atomic emission spectroscopy/mass spectrometry (ICP-AES/ICP-MS) or atomic absorption spectroscopy after acid digestion using hydrofluoric acid (HF) in order to destroy the aluminosilicate matrix (Guieu et al., 2002; Stuut et al., 2005; Castillo et al., 2008; Lazaro et al., 2008; Jiménez-Vélez et al., 2009). However, HF attacks silicon dioxide to produce soluble silicon tetrafluoride (SiF_4) in the gaseous phase, preventing the determination of Si concentrations (Pekney and Davidson, 2005).

In order to estimate the part of salt minerals in the mineralogical composition of dust samples, or to assess the potential processing of dust by adsorbed gases, the ionic inorganic and organic species (e.g., NH_4^+, NO_3^-, SO_4^{2-}, Cl^-, formate, acetate, oxalate) are determined by water extraction. Water extraction by sonication is usually used. However, possible artefacts could be produced by sonication since during implosion of the cavitational bubble nitrites and nitrates could form (Koda et al., 2004). The abundance of water-soluble ions is generally measured by ion chromatography (IC) (Formenti et al., 2003; Paris et al., 2010; Müller et al., 2010). The detection limits are low in order to $\mathrm{ng\,m^{-3}}$ of ionic species, corresponding to about $20\,\mu\mathrm{g\,m^{-3}}$ of dust, or even less in case of mixing during transport (Müller et al., 2010).

Many recent compositional studies of mineral dust have focussed on the characterisation of iron. Iron occurs mostly in the +3 valence state (i.e., ferric iron), either as Fe(III) oxides/hydroxides (hematite or goethite) or as Fe(III) contained in aluminosilicates (e. g., illite and smectite).

Quantification and speciation of iron in dust is achieved by a range of techniques. An adaptation of the classical soil science selective method of Mehra and Jackson (1960) has been developed by Lafon et al. (2004) to quantify the iron oxide content in

samples of mass higher than 500 µg. The speciation of iron oxides/hydroxides can be achieved by diffuse reflectance in the UV-visible (Arimoto et al., 2002) and high-field isothermal remanence (HIRM) (Hatfield and Maher, 2008), or by a combination of both (Lazaro et al., 2008). Synchrotron-based X-ray absorption spectroscopy (XAS) (Ohta et al., 2006; Schroth et al., 2009) and Mössbauer spectrometry (Hoffmann et al., 1996; Zhang, 2003) are also used to quantify the Fe oxidation state and bonding environment. The characterisation of iron in dust samples can be also obtained by sequential leaching (e.g. Chester et al., 1989). This method is particularly interesting for the biogeochemical impact of dust since it provides information on its environmental mobility.

Isotopic composition was used in several papers as source marker. Sr, Nd and Pb radiogenic isotopes, which depend both on the lithology and on the age of the parent rocks from which dust derived, have proved to be the most discriminating array of tracers (Grousset and Biscaye, 2005). A strong grain-size dependence of the different isotope systems (in particular, the $^{87}Sr/^{86}Sr$ isotope ratio) has to be taken into account when comparing isotope data from different dust samples (Grousset and Biscaye, 2005; Chen et al., 2007).

The composition of mineral dust can be also measured by single particle techniques. Scanning and transmission electron microscopy (SEM/TEM) combined with energy-dispersive X-ray microanalysis (EDX) is the most commonly used method for this purpose. Analysis of the chemical composition is practically limited to elements with an atomic number ≥4 (Be), and is restricted to concentrations above approximately 0.1 to 0.5% wt. Quantification of the element concentrations is complicated by the fact that the particle geometry has to be taken into account in the correction procedures (e.g., Armstrong, 1991). Although algorithms for correction of particle effects are available for automated analysis (e.g., Weinbruch et al., 1997; Ro et al., 2005), in most papers on mineral dust the chemical composition is not determined quantitatively (e.g., Ganor et al., 1996; Falkovich et al., 2001; Reid et al., 2003b; Zhang et al., 2006; Kandler et al., 2007; Jeong, 2008; Matsuki et al., 2010), but instead, the net count rates or matrix-corrected data (without geometry correction) are used to categorize the parti-

cles into several groups with similar properties (e.g., chemical composition, refractive index), as for example silicates, carbonates, iron oxides. The recent development of automated recognition procedures enable characterization of large particle numbers (30 000 to 250 000 particles; e.g., Reid et al., 2003b; Kandler et al., 2007, 2009).

Additional single particle techniques that were applied to mineral dust include laser microprobe mass spectrometry (LMMS), micro particle induced X-ray Emission (micro-PIXE) and aerosol time-of-flight mass spectrometry (ATOFMS). The latter technique is especially powerful as in addition to the elemental composition organic species can be also determined on individual particles (Sullivan et al., 2007b).

Finally, Helas and Andreae, 2008 report on the use of AFM and electrostatic force microscopy (EFM) to investigate deposits of distinguishable material on the surface of mineral dust particles.

The previous discussion illustrates the fact that a synergy of experimental and analytical techniques is necessary in represent the physico-chemical properties of mineral dust. The use of these techniques has proven unavoidable to investigate specific aspects such as the regional variability at the source and the evolution after transport. A synthesis of this investigation is reported in the following paragraphs.

3 Physico-chemical properties of African and Asian mineral dust

Most of the data gathered in these last few years concern the African and Asian sources. The Saharan desert and its margins are the largest source of mineral dust on earth, supplying up to 800 Mt per year of material to the atmosphere (Goudie and Middleton, 2001; Laurent et al., 2008). A compilation of the available data on source areas reveals the major potential source areas (PSA; Figs. 1a,d, 2). Dust emission hot spots can be identified using visibility data, back trajectory analysis, satellite observations and isotopic composition studies (e.g., Brooks and Legrand, 2003; Caquineau et al., 2002; Prospero et al., 2002; Israelevich et al., 2002; Goudie, 2003; Chen et al., 2007; Schepanski et al., 2009). The large Saharan sand seas are no prominent sources,

as they do not produce mineral dust that can be transported over long distances due to their grain size distribution. The PSA NAF-1 to NAF-6 shown in Fig. 1 are mainly topographical lows and foothills of mountains.

African sources can be fairly well discriminated by combining isotopic ratios $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}(0)$ data of soil samples (Table 2a). For example, whereas the highest $^{87}Sr/^{86}Sr$ values and least radiogenic $\varepsilon_{Nd}(0)$ values were reported for north-western areas (PSA NAF-2), PSA NAF-6 (Egypt, Sudan) is characterized by low $^{87}Sr/^{86}Sr$ values and significantly higher $\varepsilon_{Nd}(0)$ values (Grousset et al., 1998; Grousset and Biscaye, 2005).

The Chinese and Mongolian deserts are the second largest global atmospheric dust source. Annual emissions vary between 100 Mt yr^{-1} and 460 Mt yr^{-1} (Laurent et al., 2006). This region is a complex and inhomogeneous dust production area with different types of single sources that show distinct properties like soil texture, mineralogical composition, aggregation and crusting all affecting the source strength. Six important potential source areas may be identified in Fig. 2 based on a survey of the recent literature (Chen et al., 2007; Kim et al., 2007; Laurent et al., 2006; Lee and Sohn, 2009; Li et al., 2007; Prospero et al., 2002; Shao and Dong, 2006; Sun et al., 2001, 2007; Wang et al., 2008; Xuan et al., 2004; Yang et al., 2009; Zhang et al., 2003c).

Recently, a large data set for Eastern Asia was produced (see Table 2b), comprising $^{87}Sr/^{86}Sr$ and $\varepsilon_{Nd}(0)$ analyses of various size fractions from different potential source areas. Especially on the basis of $\varepsilon_{Nd}(0)$ values, a clear distinction between areas with low $\varepsilon_{Nd}(0)$ values (PSA EAS-1, PSA EAS-3, PSA EAS-5) and higher $\varepsilon_{Nd}(0)$ values (PSA EAS-2, PSA EAS-6) is possible (Yang et al., 2009). It is also remarkable that local trends in single potential source areas can be revealed by isotope composition (e.g., increasing $\varepsilon_{Nd}(0)$ values from west to east in PSA EAS-5; Chen et al., 2007). Beside these general findings, source discrimination based on chemical composition will not be discussed here (for details see the review by Scheuvens et al., 2010).

3.1 Particle composition

3.1.1 Characterisation of the composition of source regions

Selected mineralogical, elemental, and isotopic parameters of dust from Northern Africa and Eastern Asia are presented in Table 2a,b. In Northern Africa, the calcite (or carbonate) content and the ratio of the clay minerals illite and kaolinite (I/K ratio) are able to serve as "fingerprints" of specific source areas (e.g., Schütz and Sebert, 1987; Caquineau et al., 2002). The highest I/K ratios are observed in mineral dust that originated in north-western source areas (Fig. 1, PSA NAF-1, PSA NAF-2). High carbonate contents were reported for the north-western areas and PSA NAF-3 (Southern Algeria/Northern Mali). The chlorite content and the chlorite/kaolinite ratio (C/K ratio) are generally low and cannot be used for discrimination between different PSA. The Mg-dominated fibrous clay mineral (palygorskite) appears to be a suitable source marker for north-western PSA (PSA NAF-1, PSA NAF-2a) and, to a lesser extent, for PSA NAF-3. Palygorskite is also found in significant amounts in dust emitted from the Arabian Peninsula (Ganor et al., 1991; Kalderon-Asael et al., 2009).

Only a few studies deal with the mineralogical composition of unpolluted aerosol and possible source sediments in China (Svensson et al., 2000; Shen et al., 2005, 2006; Jeong, 2008). Despite the limited data set (in comparison to Northern Africa), major differences between Northern Africa and Eastern Asia are observed. For example, the I/K ratio is generally higher in samples from Eastern Asia (I/K >5) than in samples from Northern Africa (I/K<2), with the one exception of PSA EAS-4 (South-Eastern Mongolia). The C/K ratio is mostly higher in Eastern Asian source regions (C/K>1.5), compared to Northern Africa (C/K<1.5). As all three minerals appear in the same size fraction and have similar densities it can be expected that the I/K and C/K ratios are not substantially changed during long-range transport. Similar to Northern Africa, the I/K ratio in Eastern Asia seems to be a more effective tool to discriminate between different PSA than the C/K ratio. The I/K ratios are highest in western source areas (PSA EAS-1) and lowest in Northern Gobi deserts (PSA EAS-4). In addition, the

carbonate content generally decreases from west to east (with exception of the Gurbantunggut desert (PSA EAS-2), where the carbonate content is very low) and hence can also be used as a compositional fingerprint for the regional provenance of Eastern Asian dust.

Atmospheric concentration ratios for Ca/Al, Fe/Al and K/Al for Northern African and Eastern Asian dust are also presented in Table 2a,b. Al is chosen as a marker element for alumino-silicates. Ca is mainly a tracer for the carbonate minerals (calcite and dolomite), and to a lesser extent, of sulfates such as gypsum and of Ca-bearing plagioclase. K is predominantly a component of clay illite and potassic feldspar.

Though the Si/Al ratio is often reported, its capability to distinguish source regions is limited due to its low variability around 2.3 (Chiapello et al., 1997; Formenti et al., 2001a, 2003, 2008; Reid et al., 2003b; Rajot et al., 2008). An exception is the Bodélé depression source (PSA NAF-5) which, because of the resuspension of detrital diatoms deposited at the surface after the drought of Mega-Lake Chad (Todd et al., 2008), presents values of Si/Al ratio reaching 4 (Formenti et al., 2008),

The higher Ca/Al ratios in the north-west (PSA NAF-2) compared to samples from PSA NAF-3 (Mali-Algerian border region) are probably correlated with the higher carbonate contents in source sediments of the Atlas region (PSA NAF-2). The K/Al ratio shows little variability from one source area to the other. One could infer from the intense weathering in the sub-tropics that the Fe/Al ratio would increase from North to South over the region. Surprisingly, this is not an observed trend of the recent data-set. Kandler et al. (2007) emphasized an opposite north-south gradient of Fe and Ca content in dust samples collected in Canaries Island, showing higher Fe content for dust originating of PSA NAF-3. Compared to Africa, Eastern Asian dust shows higher Ca/Al and K/Al ratios, but similar Fe/Al ratios (Zhang et al., 2003d; Arimoto et al., 2006). The Ca/Al ratio exhibits a geographical dependence with decreasing ratios to the east (see Table 2), following the carbonate distribution in soil. In contrast, the Fe/Al and K/Al ratios are more homogeneous, exhibiting no (Fe/Al) or only a weak (K/Al) regional trend. It can be concluded that based on the currently available data the Ca/Al ratio is the

most promising in discriminating mineral dusts of different source areas.

Most of the last years' research has focuses on iron chemistry, in particular on the partitioning between the structural iron trapped in clay-crystal lattice and the iron oxides/hydroxides in the form of hematite (Fe_2O_3) and goethite (FeOOH). Iron oxides/hydroxides were also assumed to be a major source of water-soluble iron, i.e., iron which is potentially available for dissolution in sea water. However, it was shown recently (Journet et al., 2008) that more than 96% of the water-soluble iron is associated with the dissolution of the abundant clay minerals.

In Northern African source areas, the iron oxide content, determined by the CBD-extraction method (Lafon et al., 2004), ranges from 43% to 65% of the elemental iron content (2.4 to 5% of total mass, respectively) with higher values obtained for Sahelian dust emitted locally from Niger and Mauritania in the Sahel belt and lower ones for the Chad basin (Formenti et al., 2008; PSA NAF-5). The iron oxide content of dust transported to the Canary Islands ranges from 27% to 63%, with the tendency of higher values for source areas between 0° N–20° N (Lazaro et al., 2008; PSA NAF-3). Dust produced in laboratory wind tunnel from Niger and Tunisian soils show that goethite seems to be the most abundant iron species comprising about 65–85% of the total iron oxide mass (Lafon et al., 2006). However, the results of Lazaro et al. (2008) for different African sources (PSA NAF-1, PSA NAF-2 as well as Mali region) show a hematite/goethite ratio with a large variability between 0.5 and 2. Comparison on the hematite/goethite ratio in dust collected in Asian source areas (PSA EAS-3, PSA EAS-5 and PSA EAS-6) show low ratio with a slightly variability ranging between 0.46 and 0.59 (Shen et al., 2006). Thus, goethite can not be used for source discrimination for African or Asian dust.

Electron-microscopical observations of African dust show that iron is both present as Fe-containing silicate and as iron oxide grains in and on silicate particles (notably clay minerals). This is observed close to the dust sources (Lafon et al., 2004, 2006; Kandler et al., 2007) as well as after transport (Falkovich et al., 2001; Blanco et al., 2003; Reid et al., 2003b; Singer et al., 2004; Kandler et al., 2009). Isolated iron oxide particles are

rarely observed (less of 3% in number of total observed particles) and they are usually small (Kandler et al., 2007; Chou et al., 2008). Similar observations were made for Chinese dust (Iwasaka et al., 2003; Trochkine et al., 2003; Gao et al., 2007). For Asian dust, Ohta et al. (2006) found that the iron concentration is rather constant for particle sizes of 1.1–11.0 µm, and a variation is observed below 1 µm, which may be attributed to anthropogenic activities. For example, a lower abundance of iron in the fine fraction is observed by Iwasaka et al. (2003) over the Taklamakan desert. However, the results show no trend of the size-resolved iron distribution in terms of changing source region or distance to the source. Speciation of iron valence state shows that all iron is present as Fe(III) for the size fraction below 11 µm and approx. 93% for the fraction larger than 11 µm (Ohta et al., 2006). It is also observed that the ratios of Fe(III) to total Fe close to Asian source and after long-range transport are similar, indicating that redox reactions of Fe in aeolian dust during transport are negligible. These observations are in agreement with results for soil (representative of coarse aerosol mode), where Fe(II) is usually also low and is contained mainly in aluminosilicates (Ohta et al., 2006; Cwiertny et al., 2008).

3.1.2 Changes of composition during transport

In Asia, strong desert sources are located close to polluted areas leading to intense mixing. Thus, many individual particle studies in this region focus on the mixing state and its evolution during transport. Field measurements have shown that many dust particles contain sulphates or nitrates which are formed by heterogeneous reactions involving sulphur dioxide and nitrogen oxides, particularly in the marine atmosphere where the relative humidity is high (Trochkine et al., 2003; Zhang et al., 2003a). Most dust particles take up sulphate in the marine air during transport from the Asian continent to the Japanese islands and, thus, act as a sink for gaseous sulphur (Okada and Kai, 1995, 2004; Fan et al., 1996; Zhou et al., 1996; Trochkine et al., 2003; Zhang et al., 2003a,d). An increase of nitrate-containing Asian dust particles during transport was also described, although it is not as pronounced as for sulphate (Zhang et al.,

2003a,c). For African dust, the Mediterranean basin is the major zone of transport with high polluted areas. A strong internal mixing of dust and sulphate was also reported for the Eastern Mediterranean and Western Africa (e.g., Falkovich et al., 2001; Sobanska et al., 2003; Koçak et al., 2007). The nitrate formation on dust particles has also been observed as a result of the interaction with HNO_3 (Putaud et al., 2004; Koçak et al., 2007).

The enrichment of sulphate and nitrate on dust particles in the marine atmosphere is linked to the high humidity. In marine air, it can be expected that dust particles become soon wetted by formation of thin water films on the surface or partial deliquescence (i.e., deliquescence of the soluble fraction contained in agglomerates). The presence of liquid water will result in more efficient transformation of sulphur dioxide to sulphate and nitrogen oxides to nitrate. Nonetheless, the results of Putaud et al. (2004) indicate the nitrate formation on dust particles occur even when the relative humidity is low.

Formation of sulphate and nitrate is also closely related to the mineral composition. Laboratory experiments and field observations both revealed that sulphate formation is favoured on aluminum silicate particles compared to calcium carbonate (Krueger et al., 2003; Laskin et al., 2005a; Shi et al., 2008). The opposite behaviour is observed for nitrate formation (Ro et al., 2005; Sullivan et al., 2007a; Matsuki et al., 2010; Fairlie et al., 2010). The preferential nitrate formation on carbonates can be explained by the different hygroscopicity of Ca nitrate and Ca sulphate. Calcium sulphate (the reaction product on carbonates) is poorly water soluble preventing further uptake of water and other gaseous species. Consequently, the transformation of sulphur dioxide to sulphate is suppressed in these particles. In contrast, the major product of nitrate formation on carbonate particles is highly hydrophilic (Sullivan et al., 2009). Thus, uptake of water is enhanced resulting in a positive feedback. This feedback process will be operating until all calcium in the particles is transformed to calcium nitrate. The frequent observation of calcium-rich spherical particles in Asian dust plumes in Japan and in polluted urban air masses in China (Matsuki et al., 2005), as well as in the Eastern Mediterranean (Laskin et al., 2005a) is a strong indication for the feedback process outlined above.

Calcium-rich spherical particles have also been observed during AMMA campaigns in convective system over Sahel (Matsuki et al., 2010) in parallel with a strong internal mixing of dust with nitrate or sulphate suggesting an in-cloud processing (Crumeyrolle et al., 2008; Matsuki et al., 2010).

A few studies found chloride on sea-salt free dust particles and suggested that the absorption of hydrogen chloride was responsible for the chloride formation. This phenomenon was first observed and its importance with respect to the formation of sulphate evaluated by Zhang and Iwasaka (2001). Recent observations with ATOFMS during ACE-2 at the North Atlantic and ACE-Asia at the Northern Pacific provided more quantitative data to support this finding (Sullivan et al., 2007a,b). A study by Tobo et al. (2009) found that the chloride formation on dust particles was more pronounced in episodes of weak sulphate and nitrate formation, suggesting the presence of an additional process of dust modification by gaseous species. ACE-Asia results emphasize also a possible mixing between dust and organic dicarboxylic acid (DCA), as oxalic and malonic acid due to heterogeneous and aqueous oxidation of DCA to the alkaline Asian dust (Sullivan et al., 2007a). Internal mixing between dust and organic material has also been suggested for African dust in source areas during monsoon period by in-cloud process in convective system (Desboeufs et al., 2010) and in zones of transport in Israel (Falkovich et al., 2001) and over Alps (Aymoz et al., 2004).

Mixing of dust with particulate matter is expected to include sea salt, soot, and primary biological particles (e. g., microorganisms). The mechanisms responsible for the mixing of dust particles and sea-salt have not been elucidated (Andreae et al., 1986; Zhang et al., 2005). In mineral dust collected at the Japanese islands (i.e. after several hours of transport in the marine air) more than 60% of the particles were internally mixtures of mineral dust and sea-salt (Okada et al., 1990; Niimura et al., 1998; Zhang et al., 2003b). This abundance is much higher than that predicted from particle-to-particle collision theory based on Brownian motion, implying that additional processes assist the mixing (e.g., Ma and Choi, 2007). While in-cloud processing was suggested as a major route for agglomerate formation (Andreae et al., 1986; Niimura

et al., 1998), the high abundance of internally mixed particles were observed during cloud-free days. Small-scale phenomena, such as streak flow or warm rolls in the marine boundary layer may change the settling of dust particles to the surface and be responsible for the mixing, however, the exact pathway and the dominating effect is still unclear (Andreae and Rosenfeld, 2008; Zhang, 2008). In contrast, in the Saharan dust plume outflow of Western Africa, mixtures of dust and sea-salt were reported to occur only to a minor degree (e.g., Niemi et al., 2005; Kandler et al., 2007; Reid et al., 2003b). This is probably due to the fact that dust is transported above the marine boundary layer (Karyampudi et al., 1999).

Only few publications reported the presence of internal mixtures of soot and mineral dust (e.g. Parungo et al., 1994; Petzold et al., 2009). The origin of these mixtures is still not clear. The presence of microorganisms on dust particles has been confirmed at various sites for Asian dust (Iwasaka et al., 2009), which is not surprising for surface soil particles. The impacts of these internal mixtures are unknown.

3.2 Particle size distribution

Newly available field data of airborne dust size distributions are compiled in Table 3 with respect to source areas and long-range transport regimes. Applied methods and the altitude above ground over land and above sea level over sea are listed as well. A complementary compilation has been published by Reid et al. (2008) based on APS datasets available worldwide.

The analysis of the available data set is not straightforward due to the uncertainties in the experimental techniques and the difficulties in reconciling them (Sect. 2.1).

The prediction of the size distribution of mineral dust is an outstanding modelling issue. Various emission models based on different physical schemes (Alfaro et al., 1998; Lu and Shao, 1999; Alfaro and Gomes, 2001; Shao, 2001) describe the size distribution of mineral dust as determined by the saltation and sandblasting processes, which depending on the surface wind speed, and on soil characteristics such as dry size distribution, humidity and mineralogy (Marticorena and Bergametti, 1995; Fécan

et al., 1998; Alfaro and Gomes, 2001; Prospero et al., 2002).

Recently, Sow et al. (2009) showed from the analysis of size-resolved vertical fluxes that only the fine mass particle fraction (<2 µm in diameter) is sensitive to wind speed and becomes relatively more important as wind speed increases.

The presence of submicron dust particles is also put into evidence. The fine dust mode (Alfaro and Gomes, 2001), has been up to now a debated issue in the literature (Gomes et al., 1990; Reid et al., 2008). However, the Sow et al. (2009) observations are confirmed by most of the new data available for airborne dust after short-range transport (Chou et al., 2008; Osborne et al., 2008; McConnell et al., 2008; Weinzierl et al., 2009; Kandler et al., 2009), which all agree in showing that aluminosilicate dust particles in the diameter range between 0.1 and 0.5 µm exist and might represent up to 40% of the total particle number. This fine mode is often mixed, internally or externally, with soot or sulphate particles (Chou et al., 2008; Kandler et al., 2007, 2009; Weinzierl et al., 2009).

The dynamics of coarse particles larger than 2 µm in diameter shown by Sow et al. (2009) is different. In contrast with previous laboratory observations (Alfaro and Gomes, 2001), these authors suggest that the size distribution of the dust released by a given erosion event is fairly constant and insensitive to even relatively important variations of the friction velocity.

Then, the dynamics of the mass size distribution of advected dust after 1–2 days of transport has been investigated by Rajot et al. (2008) and Reid et al. (2008). Rajot et al. (2008) show that the coarse fraction is composed by two modes of geometric mean equal to 8.7 µm and 4.7 µm, respectively. The first mode is dominant at the beginning of the event, but it loses importance with time after transport. At contrary, the second mode gains in importance with time. This is consistent with the conclusions of Reid et al. (2008) suggesting that, for particles smaller than 10 µm in diameter, the size characteristics of dust events from a single source are rather independent of wind speed and remain practically unchanged after 1–2 days of transport in the atmosphere. This would imply that, in the absence of wet deposition or cloud processing, the size

distribution of mineral dust does not change during long-range transport. These results are in accordance with observations of the size distribution dynamics during long-range transport by Maring et al. (2003).

State-of-the-art emission models also consider that only the PM_{20} dust fraction is entrained vertically by sandblasting, while larger particles are only entrained horizontally in the saltation flux, being too heavy to become airborne by sandblasting (Alfaro et al., 1998). However, near-source ground-based data in dust storm conditions show a giant mode of particles larger than 20 µm (Rajot et al., 2008; Kandler et al., 2009; Sow et al., 2009), and, in the proximity of source regions, airborne measurements in the boundary layer below 500 m show a large mode below 40 µm diameter (Kandler et al., 2009; Weinzierl et al., 2009; Formenti et al., 2010). Coarse particles of tenths of micron in diameter are observed over Portugal after 3–4 days of transport (Wagner et al., 2009) and in the Caribbean after long-range transport (Formenti et al., 2001b; Reid et al., 2002).

3.3 Particle non-sphericity

Mineral dust at the emission stage has a rather constant median aspect ratio between 1.6 and 1.7 for Saharan dust (Chou et al., 2008; Kandler et al., 2009) and between 1.4 and 1.5 for Asian dust (Okada et al., 2001). For comparison, ammonium sulphate particles usually have aspect ratios smaller than 1.3 (Kandler et al., 2009).

The size-dependence of the aspect ratio was found to be weak to negligible (Okada et al., 2001; Chou et al., 2008; Kandler et al., 2009) a sufficiently large number of particles is analyzed, an aspect ratio distribution can be determined. For mineral dust, this distribution follows a shifted log-normal shape (Kandler et al., 2007, 2009), independent of particle size or mineralogical composition. While silicates in the Saharan desert usually have median aspect ratios of 1.6, halite (sodium chloride) and titanium oxide particles tend to have lower aspect ratios (1.4 to 1.5), whereas gypsum usually has higher ones. Carbonate particles are reported to have slightly lower values (Coz et al., 2009).

Only little work was carried out so far on the evolution of the particle shape during dust transport. The median aspect ratio does not change significantly during short-range transport (Kandler et al., 2007; Coz et al., 2009). Two studies report data for the long-range transport situation. At Puerto Rico, long-range transported Saharan dust showed a significantly higher aspect ratio of 1.9 while preserving the shifted log-normal shape of the density distribution (Reid et al., 2003a). This suggests a preferential removal of spherical particles. In contradiction, aerosol deposit analyses of Asian mineral dust in Japan resulted in significantly lower aspect ratios around 1.27, leading to the conclusion that preferentially non-spherical particles are removed (Li and Osada, 2007a).

Various processes have a potential to change the average shape of the dust particles during transport, shape-selective removal (i.e. deposition), heterogeneous processing and clouds and ice nucleation. Shape-selective removal can either directly affect particles of the same composition, but different shape, or particles with different composition (and density), which is reflected as different shape. Thus, it could increase as well as decrease the average aspect ratio. In any case, it does not change the shape of single particles. In contrast, heterogeneous processing which can occur as in-cloud and out-of-cloud processing can change the shape of individual particles. The deposition of secondary material as well as the aggregation of different dust particles in a cloud droplet would lead to lower aspect ratios due to the surface tension of evaporating droplets. On the other hand, a slow crystallization of, for example, sulphates may lead to more needle-like particles and would increase the aspect ratio (Kandler et al., 2007). In addition, the heterogeneous processing depends on the particle composition. While silicates (e.g., clay minerals, quartz) usually are rather inert, especially carbonates react with acids in liquid phase, particularly nitric acid, resulting in formation of particles with lower aspect ratios (Laskin et al., 2005a,b).

Currently, only one publication is available presenting statistical information on the third dimension (Okada et al., 2001). The authors report the prevailing presence of platy particles for Asian dust with a median height-to-width ratio of 0.3, and a limited

dependence on particle size. Due to its relevance for modelling the optical properties (Mishchenko, 2009), the particle thickness should be given further attention in future work.

4 Synthesis and recommendations

This paper presents a synthesis of the recent data on physico-chemical properties of mineral dust presented at the Third International Workshop on Mineral Dust in September 2008 in Leipzig (Germany). Most of the data stems from a number of regional studies which have been conducted in the last few years to target some of the major dust emission and export regions, i.e., the African and Asian deserts, the Atlantic Ocean and Japan Sea, as it had been recommended by Sokolik et al. (2001).

Synopsis of literature data is complicated by a strong dependence of the results on sampling and analytical methods.

Mineral dust is a multi-component, strongly size-dependent aerosol species which requires the combination of various analytical techniques and sampling methods based on different physical principles. The qualitative characterisation of the composition of bulk dust mainly via X-ray spectroscopy techniques is well established but its quantitative assessment is affected by the absence of calibration standards truly representative of the real chemical form and size distribution of the mineralogical components in the ambient situation. That is particularly true for clays, which account for the large majority of the mineral dust mass in the suspended fraction, but whose chemical form is extremely variable due to weathering, substitutions and impurities. To date, the only quantitative determination of the dust clay mineralogy based on calibration has been achieved by Caquineau et al. (1998). Nonetheless, minerals have different refractive indices and solubility properties, and their quantification is essential to estimate their impact.

We encourage the definition of common procedures for data treatment and presentation. Log-normal fitting is a common practice for representing the particle size dis-

tribution (e.g., Haywood et al., 2003a; Weinzierl et al., 2009). However, there is no common practice in the way that log-normal fitting is performed. In particular, the number of modes is often increased to optimise the curve fit, regardless of their physical meaning. We recommend that, in addition to customised fits, authors should present fit parameters for physically-based modes in order to improve comparison with model results and help in evaluating the evolution with time of the size distribution at emission. Also, reporting systematically on the skewness of the fit could help in evaluating the extent of the modifications during transport (Reid et al., 2008). In order to serve both for comparison purposes and geophysical discussions, fitting parameters of the optically-based size distribution should be presented for the refractive index of the aerosol species under investigation. Furthermore the conversion of instrument response into particle size, i.e., the calibration function needs to be reported.

Because of the dependence of particle number and mass on size, the use of certified collectors is recommended in order to reduce ambiguities when comparing results on composition, concentrations and size distribution. Sampling size limits of collectors should be indicated systematically.

Some significant progress has been made in characterising the dust properties close to source regions. Much of the investigation has focussed on the mineralogy of iron. Our synthesis on iron mineralogical speciation between oxide and structural iron concerns mainly African sources but very little the Asian dust. Recent field works confirm the laboratory conclusions of Journet et al. (2008) and emphasizes the iron speciation in dust samples is critical to estimate its solubility (Paris et al., 2010). Thus, the ratio between oxide and structural iron could constitute a simple approach to in order to better model the solubility of iron for dust transport models that focus on the role of iron in ocean biogeochemistry. For that, the determination of the oxide and structural iron content in dust samples should be systematic. The apportionment of iron oxide between goethite and hematite should be pursued as it might have implications for the optical properties (Lafon et al., 2006; McConnell et al., 2010). Currently, modelling radiative transfer studies do not take into account the hematite/goethite ratio (Soko-

lik and Toon, 1999; Miller et al., 2004; Balkanski et al., 2007; Heinold et al., 2008; Mallet et al., 2009). In general terms, the inclusion of those field observations of the dust composition into regional and global climate models will require the research of a physically-based and systematic relationship to describe the chemical and mineralogical fractionation between soil and airborne dust (Rahn, 1976) so to allow the prediction of the dust composition based on maps of soil mineralogy.

Only few data sets on the shape of dust particles are available to date. Despite the physical diversity of the shape of individual particles, there are hints that the particle aspect ratio is a rather universal parameter which only weakly depends on particle composition, size, and source region. Data agree in showing that the particle aspect ratio distribution extends up to 5, whereas state-of-the art modelling of the optical properties of randomly oriented spheroids only allows take into account values of the aspect ratio up to 3 (Dubovik et al., 2006). Veihelmann et al. (2004) suggests that extending the optical models from moderate to large aspect ratios should reduce the errors in the estimation of the optical properties. We recommend that data are gathered on a broader statistical basis in order to provide with a robust parameterization of shape and the role of potential key parameters which could control its natural variability (like chemical composition and transport distance). In parallel, sensitivity modelling using the field data should be undertaken to evaluate if, although little, the variability of aspect ratio has an impact of the optical and radiative properties of mineral dust. Data on the third dimension of particles and the surface roughness are very rare but are needed for optical modelling (Mishchenko, 2009; Nousiainen and Muinonen, 2007). Currently, there are contradictions in the evolution of the aspect ratio during transport, though the data basis is quite small. Future analyses should focus systematically on the causes of shape change (due to particle removal or due to processing) as well as the resulting direction.

The evolution of mineral dust properties during transport deserves further and systematic investigation in order to highlight and parameterise the processes responsible for those changes. This recommendation was already part of the list of high-priority

research needs which had been identified by Sokolik et al. (2001). Much of the field observations on the evolution of dust properties concern the description of changes of the single-particle composition due to particle mixing and heterogeneous reactions (Sect. 3.3.2). Surface ageing is one of the factors imputed for the large range (4 orders of magnitudes) of accommodation coefficients reported for various heterogeneous reactions (Atkinson, 2008).

In this field of research, we recommend that further attention is given to the deposition of organic acids on dust particles (Hatch et al., 2008). This process, which could be relevant in areas where dust and biomass burning might mix, like the African Sahel, could increase the ability of the particles to absorb water vapour (Hatch et al., 2008). No technique is yet available to identify organic acids accurately in individual particles.

Finally, because of its relevance in ruling the optical and radiative properties, the solubility and CCN capacity, the gaseous uptake efficiency, and the deposition of mineral dust, we consider that future research should focus on the outstanding issue of the prediction of the size distribution of mineral dust at emission and its dynamics evolution during transport. There are inherent experimental limitations in the measurements of the aerosol size distribution, and of that of the coarse fraction of dust in particular (Reid et al., 2003c). Nonetheless, the challenge of representing the full extent of the dust size distribution has to be pursued. Recent studies show the evidence of a submicron fraction of mineral dust (Chou et al., 2008; Osborne et al., 2008; McConnell et al., 2008; Weinzierl et al., 2009; Kandler et al., 2009). This fraction, surely persistent in the atmosphere in the absence of wet deposition, should be better constrained because of its radiative efficiency. This fraction is currently represented by models but it is very variable (Huneeus et al., 2010). The presence of large airborne particles above 20 µm in diameter is suggested by various studies reported in Sect. 3.2. This has limited implication for radiation, but it is important for modelling the mineral dust cycle and the impact of dust on ocean productivity. Modelling the uplifting and transport of such large particles will also represent a new serious challenge for transport models.

An open issue remains in the prediction of the anthropogenic fraction of emitted

mineral dust, that is, the fraction of dust which is emitted by human-disturbed surfaces or as a result of anthropogenic-induced climatic change (Forster et al., 2007). The anthropogenic fraction of dust only should be accounted when estimating the radiative forcing effect relevant to climate change (Forster et al., 2007). Possible differences between properties of mineral dust emitted from undisturbed to those of disturbed soils could be used to trace the relative magnitude of the anthropogenic fraction of the dust global emission. An indication could be the enhanced presence of organic material in soils which would be periodically laboured for agricultural purposes, or differences in the size distribution due to the modification of the friction velocity due to labouring. There is currently no experimental evidence of these effects.

This research should motivate dedicated near–source and medium-to-long range transport studies to be planned in the next few years. Rather than gathering new datasets, we recommend that these studies focus on the representation of the physical and chemical processes which determine the dust physico-chemical properties.

In particular, we prone for (1) an improved representation of the size-fractionation between the soil and the aerosol phases at the emission as a function of wind speed and soil type and its implication on the mineralogy and the size distribution of suspended dust; (2) a Lagrangian-type investigation of size-changes due to gravitational settling during transport; and (3) the mechanisms of water-uptake by mineral dust as a function of the evolution of its mineralogical composition due to heterogeneous reactions and mixing.

In agreement with the recommendation by Redmond et al. (2010), a systematic linkage between field observations and laboratory studies should be seek in order to advance our knowledge on those processes.

Appendix A

Acronyms

ACE-2	=	The Aerosol Characterisation Experiment – 2 (Raes et al., 2000a)
ADEC	=	Aeolian Dust Experiment on Climate Impact (Mikami et al., 2005)
BoDEx	=	Bodélé Dust Experiment (Todd et al., 2008)
SHADE	=	Saharan Dust Experiment (Tanré et al., 2003)
PRIDE	=	Puerto Rico Dust Experiment (Reid and Maring, 2003)
MINATROC	=	Mineral Dust and Tropospheric Chemistry (Balkanski et al., 2003)
ACE-Asia	=	Aerosol Characterisation Experiment – Asia (Huebert et al., 2003)
AMMA	=	African Monsoon Multidisciplinary Analysis (Redelsperger et al., 2006)
DODO	=	Dust Outflow and Deposition to the Ocean (McConnell et al., 2008)
DABEX	=	Dust and Biomass-burning Experiment (Osborne et al., 2008)
GERBILS	=	Geostationary Earth Radiation Budget Intercomparison of Long-wave and Shortwave radiation (Marsham et al., 2008)
SAMUM	=	Saharan Mineral Dust Experiment (Heintzenberg, 2009)
CARIBIC	=	Civil Aircraft for Remote Sensing and In-Situ-Measurements in the Troposphere and Lower Stratosphere Based on the Instrumentation Container Concept (Hermann et al., 2001)
PELTI	=	Passing Efficiency of an Airborne Low Turbulence Aerosol Inlet (Huebert et al., 2004)

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Table 1. Definition of the dust physico-chemical properties and their role in climatic and environmental impact.

Parameter	Direct radiative effect	Indirect effect	Biogeochemical impact	Heterogeneous reactions
Size distribution	Optical efficiency (number size distribution)	CCN/IN activation (number size distribution)	Wet and dry deposition (Mass number distribution)	Gas adsorption (surface number distribution)
Composition	Refractive index (mineralogical and elemental composition)	Hygroscopicity (soluble fraction and elemental composition)	Wet and dry deposition (soluble fraction and elemental composition)	Gas adsorption (mineralogical composition)
Shape	Scattering phase function (aspect ratio, surface roughness)		Dry deposition (aspect ratio)	

Table 2a. Selected mineralogical, elemental, and isotopic parameters for Northern African mineral dusts and source sediments. Analyses given as mass fraction (weight percentage) were discarded, because they are very scarce for Asian dusts and are not directly comparable to atmospheric concentration ratios.

Potential source area	Illite/kaolinite ratio	Chlorite/kaolinite ratio	Carbonate content	Ca/Al	Fe/Al	K/Al	$^{87}Sr/^{86}Sr$	$\varepsilon_{Nd}(0)$
PSA NAF-1	1.0–2.0 (Coudé-Gaussen, 1991; Paquet et al., 1984; Caquineau et al., 2002)	1.5 (Paquet et al., 1984)	High (10–50) (Coudé-Gaussen, 1991; Paquet et al., 1984)		0.52–0.70 (Chester et al., 1984; Bergametti et al., 1989b)		0.714–0.717 (Grousset et al., 1992, 1998)	−13.5 to −9.5 (Grousset et al., 1992; Grousset and Biscaye, 2005)
PSA NAF-2	>1.6 (Avila et al., 1997; Caquineau et al., 2002; Kandler et al., 2009)	0.0–0.8 (Glaccum and Prospero, 1980; Avila et al., 1997; Kandler et al., 2009)	High (5–70) (Avila et al., 1997; Khiri et al., 2004; Kandler et al., 2009)	0.60–0.92 (Bergametti et al., 1989a; Chiapello et al., 1997)	0.99–1.24 (Bergametti et al., 1989a,b)	0.31–0.35 (Bergametti et al., 1989a)	0.720–0.738 (Grousset et al., 1992, 1998)	−17.9 to −13.5 (Grousset et al., 1998)
PSA NAF-3	0.3–0.7 (Paquet et al., 1984; Caquineau et al., 1998, 2002)	0.2–0.9 (Paquet et al., 1984; Alastuey et al., 2005)	Variable (0–30) (Paquet et al., 1984; Falkovich et al., 2001; Alastuey et al., 2005)	0.36–0.40 (Chiapello et al., 1997; Alastuey et al., 2005)	0.52–0.88 (Bergametti et al., 1989b; Alastuey et al., 2005)	0.22 (Alastuey et al., 2005)	(0.721–0.726) (Grousset et al., 1998)	−12.4 to −12.1 (Grousset et al., 1998)
PSA NAF-4	0.2–1.9 (O'Hara et al., 2006)	0.0–2.6 (occasionally higher) (O'Hara et al., 2006)	Variable (1–21) (O'Hara et al., 2006)		0.55–0.65 (Guieu et al., 2002b)		0.715 (Grousset and Biscaye, 2005)	−15.4 to −10.7 (Grousset and Biscaye, 2005)
PSA NAF-5	0.0–0.4 (Mounkaila, 2006)	Chlorite not detected (Mounkaila, 2006)	Very low (Mounkaila, 2006)				No data	−12.7 (Grousset and Biscaye, 2005)
PSA NAF-6	0.7 (Caquineau et al., 2002)	No data	No data (probably low)	0.75 (Eltayeb et al., 1993)	1.08 (Eltayeb et al., 1993)	0.24 (Eltayeb et al., 1993)	0.706–0.718 (Krom et al., 1999; Grousset and Biscaye, 2005)	−11.0 to −3.9 (Grousset and Biscaye, 2005)

Table 2b. Selected mineralogical, elemental, and isotopic parameters for Eastern Asian mineral dusts and source sediments. Analyses given as mass fraction (weight percentage) were discarded, because they are very scarce and are not directly comparable to atmospheric concentration ratios.

Potential source area	Illite/ kaolinite ratio	Chlorite/ kaolinite ratio	Carbonate content (* analyzed by titration)	Ca/Al	Fe/Al	K/Al	$^{87}Sr/^{86}Sr$	$\varepsilon_{Nd}(0)$
PSA EAS-1	12.5–13.9 (Shen et al., 2005)	3.3 (Shen et al., 2005)	Intermediate (9–15) (Wang et al., 2005*)	2.04–4.14 (Zhang et al., 1996, 2003b; Makra et al., 2002)	0.87–1.74 (Zhang et al., 1996, 2003b; Makra et al., 2002)	0.54–0.94 (Zhang et al., 1996, 2003b; Makra et al., 2002)	0.712–0.717 0.717–0.720 (TSP) 0.722–0.730 (<5 µm) (Sun 2002a,b; Bory et al., 2003; Honda et al., 2004; Nakano et al., 2004; Kanayama et al., 2005; Chen et al., 2007)	−12.7 to −8.4 (Bory et al., 2003; Honda et al., 2004; Nakano et al., 2004; Kanayama et al., 2005; Chen et al., 2007)
PSA EAS-2	No data	No data	Very low (<2) (Wang et al., 2005*)	1.69 (Zhang et al., 1996)	0.48 (Zhang et al., 1996)	0.43 (Zhang et al., 1996)	0.711–0.714 0.717 (<5 µm) (Sun 2002a,b; Honda et al., 2004; Nakano et al., 2004; Chen et al., 2007)	−6.3 to −1.2 (Honda et al., 2004; Nakano et al., 2004; Chen et al., 2007)
PSA EAS-3	7.0–8.0 (Shen et al., 2006)	2.0–2.2 (Shen et al., 2006)	Intermediate (8–11) (Wang et al., 2005*; Shen et al., 2006)	0.94–1.80 (Zhang et al., 1996, 2003a,b)	0.65–3.53 (Zhang et al., 1996, 2003a,b)	0.26–0.44 (Zhang et al., 1996, 2003a,b)	0.713–0.719 0.727–0.733 (<5 µm) (Sun, 2002b; Nakano et al., 2004; Chen et al., 2007)	−13.9 to −9.1 (Chen et al., 2007)
PSA EAS-4	2.3–3.0 (Svensson et al., 2000)	1.3–2.1 (Svensson et al., 2000)	(Very) low (0–5) (Wang et al., 2005*; Jeong 2008)				0.713–0.716 0.714–0.719 (<5 µm) (Biscaye et al., 1997; Bory et al., 2003; Nakano et al., 2004)	−9.9 to −4.9 (Biscaye et al., 1997; Bory et al., 2003; Nakano et al., 2004)
PSA EAS-5	5.0–5.6 (Shen et al., 2005)	1.4 (Shen et al., 2005)	Low (0–10) (Wang et al., 2005*; Jeong, 2008)	0.67–2.43 (Zhang et al., 1996, 2003a,b; Alfaro et al., 2003; Arimoto et al., 2004; Xu et al., 2004)	0.36–1.60 (Zhang et al., 1996, 2003a,b; Alfaro et al., 2003; Arimoto et al., 2004; Xu et al., 2004)	0.20–1.00 (Zhang et al., 1996, 2003a,b; Alfaro et al., 2003; Arimoto et al., 2004; Xu et al., 2004)	0.713–0.726 0.721–0.732 (<5 µm) (Bory et al., 2003; Honda et al., 2004; Nakano et al., 2004; Yokoo et al., 2004; Chen et al., 2007)	−19.7 to −7.2 (W-E trend) (Bory et al., 2003; Honda et al., 2004; Nakano et al., 2004; Yokoo et al., 2004; Chen et al., 2007)
PSA EAS-6	7.5–9.7 (Shen et al., 2005)	1.7–2.0 (Shen et al., 2005)	Very low (<1) (Wang et al., 2005*)	0.43–0.76 (Cheng et al., 2005; Shen et al., 2007)	0.70–0.77 (Cheng et al., 2005; Shen et al., 2007)	0.36–0.42 (Cheng et al., 2005; Shen et al., 2007)	0.710–0.714 0.716–0.717 (<5 µm) (Honda et al., 2004; Nakano et al., 2004; Chen et al., 2007)	−7.9 to −0.8 (Honda et al., 2004; Nakano et al., 2004; Chen et al., 2007)

Table 3. Dust size distributions covering the full size range; references are listed if parameterised size distributions are given.

	Dust source	Sampling location	Method	Altitude	Dust coarse mode #1 CMD (µm)	GSD	VMD (µm)	Dust coarse mode #2 CMD (µm)	GSD	VMD (µm)
Sahara near source										
Osborne et al. (2008)	Lake Chad/Niger	Niger	OPC	BL	2.0	1.3	2.4	3.9	1.4	5.1
					1.2	1.6	2.3	4.4	1.4	6.2
Chou et al. (2008)	Lake Chad/Niger	Niger	TEM/SEM	BL	0.8	1.5	1.3	2.0	1.7	4.2
					0.8	1.5	1.3	2.0	1.6	3.9
					0.7	1.4	1.0	1.5	2.0	6.3
Weinzierl et al. (2009)	NW Sahara	Zagora Level#1	DMPS-OPC	562 m a.g.l.	1.0	2.0	4.1	5.7	1.7	12.4
		Zagora level #2	DMPS-OPC	1390 m a.g.l.	1.1	2.1	5.4	5.9	1.6	10.8
		Zagora level #3	DMPS-OPC	2730 m a.g.l.	0.9	2.1	4.6	5.2	1.7	13.1
		Zagora level #4	DMPS-OPC	4485 m a.g.l.	1.0	2.0	3.7	6.2	1.9	19.5
		Ouarzazate level#1	DMPS-OPC	1165 m a.g.l.	0.8	2.1	4.2	4.8	1.7	10.9
		Ouarzazate level#2	DMPS-OPC	1875 m a.g.l.	1.0	2.0	4.4	5.8	1.8	15.5
		Ouarzazate level#3	DMPS-OPC	3215 m a.g.l.	1.1	2.0	5.1	6.3	1.8	16.7
		Casablanca level#1	DMPS-OPC	3900 m a.g.l.	0.8	2.3	5.8	5.0	1.6	9.0
Kandler et al. (2009)	NW Sahara	Zagora dust storm	DMPS-APS-impactor	Ground level	1.7	3.7	3.5	153.9	1.4	210.4
		Zagora medium dust			0.7	1.8	3.5	31.1	2.5	374.1
Rajot et al. (2008)	Sahara	Niger	OPC	Ground level	4.5			8.7		
Sahara transport regime										
McConnell et al. (2008)	Mauritania	Dakar coast	OPC	1000 m a.s.l.	2.0	1.3	2.5			
McConnell et al. (2008)	Mauritania	Dakar coast	OPC	1000 m a.s.l.	1.9	1.3	2.4			
Haywood et al. (2003a)	W Sahara	Cape Verde islands	OPC		2.1	1.3	2.6	3.6	1.5	5.9
Bates et al. (2002)	NW Sahara	Portuguese coast	DMPS-APS	Sea level	0.9	1.8	2.6			
de Reus et al. (2000)	NW Sahara	Tenerife	DMPS-OPC	FT	0.6	2.5	6.8			
Wagner et al. (2009)	NW Sahara	Portugal	APS, DMPS-OPC	FT	1.2	2.0	5.1	6.6	1.5	11.2
Maring et al. (2003)	NW Sahara	Canary Islands	APS	Izana (2360 m a.s.l.)	0.3	2.4	3.4			
Maring et al. (2003)	NW Sahara	Puerto Rico	APS	Cabras Island	0.4	2.2	2.5			
Arabian Peninsula										
Reid et al. (2008)	UAE sand fields	UAE coast	APS-cascade impactor		0.7	2.0	2.9			
Reid et al. (2008)	Iraq	UAE coast	APS-cascade impactor		1.2	1.9	4.1			
Reid et al. (2008)	Yemen/Oman	UAE coast	APS-cascade impactor		0.5	2.2	3.5			
Bates et al. (2002)	Arabian Peninsula	Arabian Sea	DMPS-APS	Sea level	1.1	1.8	3.0			
Namibian desert										
Haywood et al. (2003b)	Aged dust	Namibia	OPC		1.6	1.9	5.5			
Haywood et al. (2003b)	Dust+regional haze	Namibia	OPC		2.0	1.9	6.9			

Table 3. Continued.

	Dust source	Sampling location	Method	Altitude	Dust coarse mode #1 CMD (μm)	GSD	VMD (μm)	Dust coarse mode #2 CMD (μm)	GSD	VMD (μm)
Chinese deserts										
Quinn et al. (2004)	Gobi/Taklimakan	Sea of Japan	DMPS-APS	Sea level	0.9	1.8	2.5			
Clarke et al. (2004)	Gobi/Taklimakan	Sea of Japan	DMA-OPC	BL	1.8	1.9	5.5	8.7	1.5	14.2
Kobayashi et al. (2007)	Gobi/Taklimakan	Kofu, Japan	Coulter Mulitsizer	Ground level	0.5	2.4	5.6			
Kobayashi et al. (2007)	Gobi/Taklimakan	Kofu, Japan	Coulter Mulitsizer	Ground level	0.8	2.1	4.3			
Kim et al. (2004)	Gobi/Taklimakan	Dunhuang, China	OPC	FT	1.0	1.5	1.6			
Zhang et al. (2003c)	Gobi/Taklimakan	Zhenbeitai, China (dust storm)	cascade impactor	Ground level		>2.0	≅11			
North American deserts										
Peters (2006)	Arizona desert	Phoenix, Arizona	APS	Ground level	1.6	1.9	5.4			
Global model data sets										
Dentener et al. (2006)	AeroCom coarse mode				1.3	2.0	5.5			
Hess et al. (1998)	OPAC coarse mode				3.8	2.2	22.0			

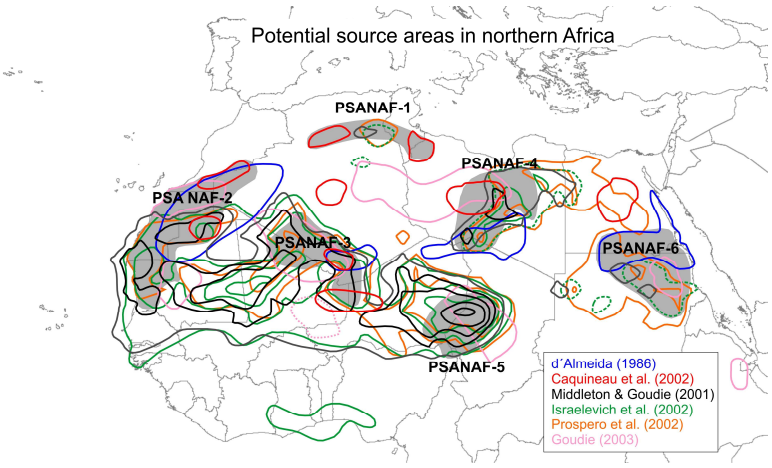

Fig. 1. Potential source areas in Northern Africa (PSA NAF) based on work by Brooks and Legrand (2000), Caquineau et al. (2002), Prospero et al. (2002), Israelevich et al. (2002), Goudie (2003), Schepanski et al. (2009). These are PSA NAF-1: Zone of chotts in Tunisia and Northern Algeria; PSA NAF-2: Foothills of Atlas mountains (PSA NAF-2a) and western coastal region (PSA NAF-2b; Western Sahara, Western Mauritania); PSA NAF-3: Mali-Algerian border region; PSA NAF-4: Central Libya; PSA NAF-5: Bodélé depression (Western Chad); PSA NAF-6: Southern Egypt, Northern Sudan.

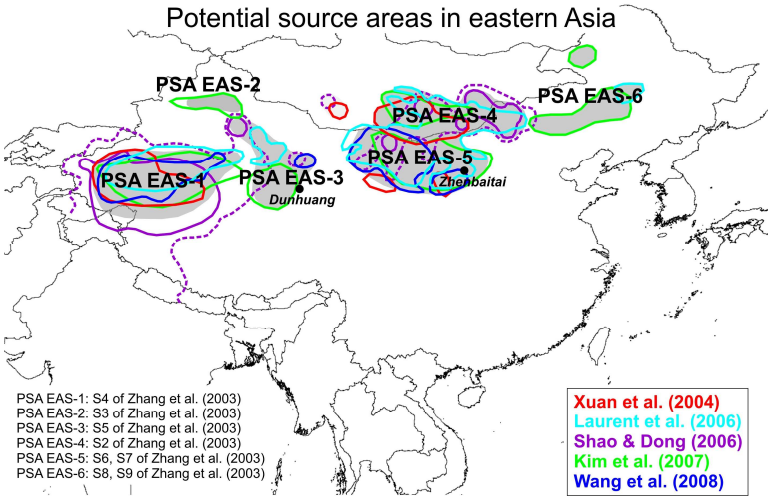

Fig. 2. Potential source areas in Eastern Asia. PSA EAS-1: Taklamakan; PSA EAS-2: Gurbantunggut; PSA EAS-3: Kumtaq, Qaidam, Hexi corridor; PSA EAS-4: Mongolian (Northern Gobi) deserts; PSA EAS-5: Inner Mongolian (Southern Gobi) deserts: Badain Jaran and Tengger (PSA EAS-5a), Ulan Buh, Hobq, Mu Us (PSA EAS-5b); PSA EAS-6: north-eastern deserts (Otindag Sandy Land, Horquin Sandy Land, Hulun Buir Sandy Land).