

# Asian and Saharan dust from a chemical/mineralogical point of view: differences and similarities from bulk and single particle measurements

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**Abstract.** This paper combines a review on the importance of dust composition with respect to numerous atmospheric impacts with field measurements performed in African and Central Asian dust. In the review part, the most important dust components and their relevance for certain processes are outlined. Typical compositions from bulk measurements for African and Asian dust are presented. Generally the local variation in composition can be higher than the differences between Asian and African dust and their according specific sources. While similar general results are available from individual particle analyses, these investigations add important information on mixing state and homogeneity of composition. Atmospheric aging of mineral dust is observed globally, depending on transport distances from the sources and transport environment. As an illustration, comparative field measurements of African and Asian dust deposition are presented.

## 1 Introduction

Mineral dust undoubtedly is one of the most important aerosol types in the Earth system [1]. Dust derived from soils is among the largest contributors to the global aerosol burden [2]. It dominates climate effects over considerable areas of the world [3-5].

Most of the impacts of mineral dust depend on its microphysical and composition properties [6, 7]. Dust changes the global energy balance by its direct radiative forcing. With respect to the solar forcing in particular iron oxides play a dominant role for radiation absorption [8, 9]. The thermal radiation budget is considerably impacted by dust mineralogy [10, 11]. Dust modifies cloud cycles macroscopically by changing atmospheric stability conditions [12], and microscopically by modifying cloud condensation and ice nucleating processes [13, 14], thus affecting the global water cycle and indirectly climate. Apparently, the latter effect also is depending on dust composition and mixing state [15, 16]. In particular for ice activation in clouds, the composition plays a key role [17-19].

Beyond atmospheric aspects, dust composition impacts the terrestrial and the ocean biosphere. It supplies missing nutrients to tropical and extra-tropical ecosystems [20, 21] as well as to ocean surface waters, typically depleted in iron and/or phosphorus [22-24]. The bioavailability of these nutrients depends highly on mineralogical composition [25, 26]. Via the biosphere, again the climate can be influenced indirectly [27]. On the other side, also adverse health effects are reported, e.g., potentially by transport of microbiomes [28] or toxic substances [29].

Mineral dust in the atmosphere is also subject to chemical reactions, acts as catalyser and provides surface for heterogeneous reactions. For example, the carbonate compounds in dust can lower considerably the atmospheric acidity by reacting with nitric and sulfuric acid [30, 31]. Metal oxide compounds in conjunction with photochemical processes can lead to catalytic ozone decomposition [32] and modify NO cycles [33]. A variety of organic and inorganic reaction can take place on the dust surface [34], while reaction efficiency is apparently affected by dust composition [35].

Finally, also humankind's daily life is affected by dust composition. Health effects strongly depend on composition parameters [36], but also solar energy yield is affected by mineral-specific dust absorption [37, 38], and aircraft operations [39] may be hampered by abrasion or dust melting.

Therefore in the present paper, a synthesis of dust composition as encountered in various regions around the globe is given. In addition to bulk composition properties, a second section deals with individual particle properties, as particle to particle variation may have a considerable impact on dust behavior.

## 2 Dust composition

### 2.1 General aspects

Atmospheric mineral dust is emitted from surfaces of the Earth by different processes, mainly saltation-sandblasting and aggregate fragmentation [40, 41]. Therefore, it could be generally composed of any

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available mineral (i.e. more than a 1,000). However, many of them are rare or confined to certain geographical areas with specific geology. Moreover, sediment composition and dust composition are not identical because fractionation occurs during emission. While the connection between sediment composition and dust composition is not well-characterized, it is for example known that clay minerals are enriched relatively, and quartz and feldspars are depleted [42]. As a result, usually only a much lower amount of mineral groups – around 20 – is identified in atmospheric dust [43, 44].

Different minerals and mineral groups are relevant for different atmospheric processes, so it makes sense to classify the dust accordingly.

1. Clay minerals: clay minerals usually contribute the majority of the atmospheric dust mass and number. Therefore, they are among the most relevant for optical aspects like short-wave radiation scattering and long-wave absorption and emission, but less so for short-wave absorption due to their low imaginary part of the refractive index [8, 10]. Most common clay minerals / mineral groups in atmospheric dust are kaolinite, illite, chlorite, smectites, and micas.

2. Iron oxides and oxy-hydroxides: they are most important for light absorption because their imaginary part of the refractive index is order(s) of magnitude higher than that of clay minerals [8, 45]. They can exist as separate grains, but also mixed with clay mineral aggregates [46-48]. They are most commonly a minor or trace compound of dust by mass. Most common mineral species are hematite and goethite for natural sources.

3. Carbonates: they are the most reactive particles among the dust and can readily interact with acids like sulfuric or nitric acid [49, 50]. By this pathway, they may strongly increase their hygroscopicity. The carbonate contribution is variable and strongly depends on the source. Most common carbonates are calcite and dolomite.

4. Feldspars: feldspars have come recently into focus, as some minerals of the feldspar class are efficient ice nuclei and can therefore influence cloud processes [19, 51]. Feldspars usually only contribute a minor fraction to the total dust. Most common minerals in this group are albite, orthoclase, microcline and the variable plagioclases.

5. Catalytic substances: some compounds in dust can act catalytically for chemical or photochemical processes, mainly iron and titanium oxides. As for the iron oxides, titanium oxides are usually present only in traces. Most common titanium mineral are rutile / anatase (usually not distinguished) and ilmenite.

6. Nutrients: for marine ecosystems usually iron and phosphorus compounds are of interest [52], whereas for terrestrial ones phosphorus, calcium and magnesium are discussed [53, 54]. Which compound can act as efficient nutrient, depends on the ecosystem state [22]. Common phosphate compounds are the apatites, magnesium can be present in dolomite or in different clay minerals.

7. Other substances like directly toxic or abrasive materials: the most abrasive and still major or minor compound of dust would be quartz. However, due to its

large particle size, it is usually not transported far in this potentially abrasive size range [55]. Other substances reported for toxicity are for example copper compounds [29]. Human health aspects are frequently related to quartz and asbestos, for different reasons [56], but also toxic substances in natural dust such as chromium and other heavy metals are reported [57, 58]. However, most of the health-related investigations are focused on individual substances, so no generalizations can be derived here.

## 2.2 Dust bulk composition and sources

The bulk composition of mineral dusts (and their source sediments) can be given as a mineralogical composition mainly determined by X-ray diffractometry (XRD) or as a chemical composition determined by vastly different analytical methods (e.g., AAS, ICP-MS, XRF).

XRD analyses revealed that the mineralogical bulk composition of mineral dust is generally dominated by silicates, carbonates, and iron and titanium oxides [44, 59]. The most important silicate phases are quartz, feldspar (plagioclase, K-feldspar), and different phyllosilicates (mica, chlorite, clay minerals). Other silicates (e.g., amphiboles, pyroxenes, palygorskite) only occur in minor or trace amounts. Quartz is the major mineralogical phase of mineral dust and generally contributes between approximately 10 and 60 wt.% to the mineralogical bulk composition. Feldspar minerals are only a minor component and rarely exceed 10 wt.%. The evaluation of a compiled data set from the literature reveals no correlation between quartz or feldspar abundance, the quartz/feldspar ratio or the plagioclase/K-feldspar ratio and specific source areas [43, 44]. Better suited for the assignment of (far-travelled) mineral dust samples to specific potential source areas are the occurrence and abundance of chlorite, members of the illite, kaolin and smectite group, and palygorskite. The latter is a rarely observed magnesium aluminium phyllosilicate which is characteristic for source regions in north(west)ern Africa [60] and dusts transported to the Mediterranean region (e.g., [61]) and the Canary islands [62].

Depending on source sediment composition, and fractionation during entrainment and transport, the amount of clay-sized phyllosilicates in dust samples varies widely from minor amounts to significantly more than 50 wt.% (up to 87 wt.% (average) for western northern African dust samples [59]). Whereas smectite group, illite/smectite mixed layer minerals and pyrophyllite are with a few exceptions rare in mineral dust samples, the abundance and ratios of chlorite, kaolinite, and illite can be a sensitive tracer and compositional fingerprint of a source region [63]. For example, the ratio of illite to kaolinite (I/K ratio) is higher for dust samples originating in northwestern Africa ( $> 1.0$ ) than in the sub-Saharan (Sahelian) region ( $< 0.5$ ) [44, 59]. Concerning Asian dust, [64] reported elevated kaolinite abundances in Asian desert soil samples and hence significantly lower I/K (0.7 to 3.7)

and C/K ratios (0.3 to 1.1) compared to earlier reports (see compilation in [65]).

Apart from the phyllosilicates, also the carbonate contents in mineral dust samples can be used as fingerprints of potential source areas. For example, in northern Africa dust samples enriched in carbonates (calcite and dolomite) are mainly uplifted in northwestern source areas (northern Algeria, Morocco), whereas in eastern Asia carbonate-rich dust samples mostly originate in more westerly source regions (e.g., Taklamakan Desert; [65]).

Additional minerals that were detected by X-ray diffraction in mineral dust samples include Fe (hydr)oxides as magnetite, hematite or goethite, and salt minerals as gypsum or halite [44].

A very helpful tool for the understanding of the global distribution of the mineralogical composition of dust source sediments is the construction of world-wide mineralogical maps by [66].

The mineralogical composition of dust is strongly coupled to its chemical composition. Hence, most dust samples are characterized by elevated Si (and Al) contents with Si/Al ratios between 1 and 7 for northern African and eastern Asian dust samples [43]. High Si/Al ratios ( $> 3$ ) are observed for example for dust samples from the largest single dust emission spot on earth, the Bodélé depression in Chad [59]. The compiled data set of [44] reveals that the elemental ratio (Ca+Mg)/Fe is a good indicator of dust provenance in northern Africa with values  $> 1$  for dusts from the 'northern belt' (Atlas region, central Algeria, Libya, and Egypt) and values  $< 1$  for dusts from the sub-Saharan region (see also [59], their Figure 3). Other elements or elemental ratios do not show or only exhibit weak regional trends. TiO<sub>2</sub> concentrations are generally low ( $\sim 1$  wt.%). TiO<sub>2</sub> is incorporated in Ti-dominated minerals as rutile and anatase or in FeTi oxides as ilmenite. Elevated K contents in Sahelian samples may be correlated with higher abundances of kaolin group minerals in this area but may also be due to an additional biomass-burning component. Dusts from the eastern Asian region are comparable in composition to dusts from northern Africa. In eastern Asia, an unequivocal trend with decreasing Ca/Al ratios from western (Taklamakan desert) to eastern source regions is observed which is again associated with the mineralogical composition ([65], see above).

As a final remark, it should be noted that in mineral dust the 'nutrients' Fe and P are at least partially structurally bound to phyllosilicates as illite, smectite, or chlorite (Fe) and apatite (P), respectively. This causes highly variable solubilities of these nutrients and hence bioavailabilities for marine and terrestrial ecosystems.

### 2.3 Dust composition on the single particle scale

For observations regarding aerosol, the single particle perspective can be of particular interest, as many properties may considerably differ on a particle-to-particle scale. E.g., if an aerosol contains 20% quartz and

80% clay minerals, it usually does not mean that every particle consists proportionally of the according materials. Usually, it does also not mean that there are 20% quartz particles, but instead, there will most probably be a distribution of internally and externally mixed particles with different abundances.

Different approaches have been used for measuring single particles, namely single particle mass spectrometry (e.g., ATOFMS = aerosol time-of-flight mass spectrometry [67]), optical and electron microscopy (including SEM = scanning electron microscopy and TEM = transmission electron microscopy), X-ray microscopy, etc., all of which have different strengths and weaknesses (see [68]). As electron microscopy appears to be the most wide spread approach, we will focus here on this technique (see also [43]).

When particle composition is measured, commonly an image of the particle and sets of chemical and morphological information become available. After manual or automated analysis of the particles, data are often generalized by classifying particles according to their properties. However, to date there is no standard approach, so a comparison between different studies remains restricted by the use of different generalization techniques.

Generally, individual-particle studies confirm the results of bulk analytical techniques such as X-ray diffraction of bulk samples. They reveal that mineral dusts mainly consists of Si-rich particles (e.g. quartz), different Al- and Si-rich particles (mainly phyllosilicates and feldspar minerals), and Ca- and CaMg-rich particles (calcite and dolomite, respectively) [42]. For example, in Eastern China dust storm samples were reported to consist of 50-58% aluminosilicates, of which around 45-50% were clay minerals and amphiboles and 5% were feldspars, 5-10% quartz, 5-60% carbonates, 15% sulfate-phosphate-(silicate) mixtures, and 3-5% iron-rich particles [50, 69]. A similar composition was found for Central China [70] with with 60-70% aluminosilicates, 10-20% carbonates, 2-7% iron-rich (the remainder consisting of Na, Cl, S, K), as well as in Western China (between Gobi and Taklamakan) with 45-57% phyllosilicates, 7-10% quartz, 5% feldspars, 15% amphiboles, 18-25% carbonates, 4% iron-rich, and 1% Ti-rich [50, 71]. It becomes obvious that other mineral classes apart from the silicates and carbonates as S-rich, Cl-rich, C-rich, Fe-rich, or Ti-rich particles are either low in abundance or are enriched owing on one side to external and internal mixing of mineral dust with particles of other sources (for example sea-salt, bio-mass burning; see below). On the other side, individual-particle studies from eastern Asia observed that enrichment of S-, Cl- or P-rich particles can occur due to a primary source sediment derived origin [69, 72].

However, this classification of particles into distinct groups might hide the fact that there exist many particles with transient compositions. An evaluation of the chemical data with for example simple point plots in most cases don't exhibit sharp boundaries between different classes but rather smeared point clouds [46, 55]. This shows that mineralogical 'end-member

particles' are rather rare, and that most particles represent (complex) internal mixtures between different phases. For example, Fe-rich particles as hematite or goethite are mainly associated with aluminosilicates [47, 73, 74] and are often positioned at the surface of the particle with important consequences for the refractive index of these phase aggregates.

In addition, external and internal mixing of mineral dust with other aerosol types (altered sea-salt, secondary aerosol, biomass-burning) occurs sometimes rather fast and is observed even in freshly emitted dusts (for northern African dust [55, 73], for Asian dust lower internal mixings are reported [71, 75]). On their way downwind and depending on ambient conditions such as the presence of different acid trace gases, the surface of mineral dust particles will be modified by the generation of coatings of C-, S-, N- or P-rich components, and therefore are reported frequently [76-84]. Also, mixtures of dust and biological material are found [85]. Especially the carbonate particles may react partially or completely into new phases like calcium sulfate [50], calcium nitrate [35], or calcium chloride [86] during transport. Commonly, apart from an existing mixture already in source soils [72, 87], cloud processing is assumed to be the reason for the internal mixture, which is also reported for African dust [35, 88]. Less frequently, turbulent processes are assumed to mix the particle species [84, 89, 90]. While it is hypothesized that coagulation is less efficient than cloud processes [90], dust-laden airmasses are frequently transported under dry conditions. As a result, governing processes are most likely variable and depend on transport conditions [89-91]. For example, for particles originating in Africa after a cross-ocean long-range transport toward the Caribbean, only around 5-20% of mixtures were observed [89, 92, 93]. In contrast, commonly higher and highly variable numbers are reported for dust observed in coastal China and Japan [83, 84, 90, 91]. This difference corresponds to different transport patterns, dry transport in a layer aloft [94], versus potentially humid transport close to the ocean surface.

### *2.3.1 Examples of Asian and African dust deposition*

The following section presents observations by the authors serving to illustrate the general findings above. Dust dry deposition was collected during the last years on a daily basis for periods ranging from one week to several years in different regions of the world. Deposition was collected with passive samplers on carbon adhesive and analyzed by scanning electron microscopy and X-ray fluorescence (for details refer to [89]). Measurements took place for African dust in Morocco near dust sources [95], at Cape Verde [55] and on Cyprus in boundary layer transport, at Tenerife, Canary Islands at the beginning of long-range transport [46], and on Barbados at the end of long-range transport [89]. Asian dust was sampled close to sources in Dushanbe, Tajikistan, in August 2016 and in Amakusa, Japan, in boundary layer transport. For contrasting the

African and Asian desert samples, Arctic dust was sampled on Svalbard in September 2017.

In general, a tendency to downwind-fining can be observed, with the largest particles close to the sources (Morocco, Tajikistan, Svalbard), whereas on the more distant sites the median diameters can be lower by half. No considerable change is observed over the long range transport from Tenerife to Barbados, which is related to the properties of the transport layer [96]. In all cases, quartz (mass) is quickly depleted from the aerosol, owing to its large particle size.

The composition in long-range transport at Barbados is constant over time. With respect to clay minerals, it shows low illite/kaolinite and chlorite/kaolinite ratios. Similar low ratios are found for Cape Verde and Tenerife. This reflects the humid weathering conditions for the source soils, yielding a high abundance of kaolinite. Measurement close to the arid sources in Morocco exhibit much higher illite and lower kaolinite contents. Particle samples in Japan show similar ratios as in Morocco, pointing to similarly arid sources. Even lower kaolinite contributions are found for Dushanbe and the Arctic dust, emphasizing the absence of warm and humid chemical weathering. For the Arctic dust, high abundances of chlorite are detected. The Eastern Mediterranean samples finally show a variable composition with high as well as low illite/kaolinite ratios, owing to a variation in source regions.

Carbonates are particularly abundant in Moroccan and Tajik measurements at an intermediate particle size (3-4  $\mu\text{m}$ ). While they are present as calcite and dolomite grains in Morocco, in Tajikistan they are frequently internally mixed with silicates, pointing to emission from evaporitic locations (e.g. dry lakebed). Interestingly, no gypsum formation is observed on these particles. At lower abundances, in Arctic dust carbonates are present as small particles (<2  $\mu\text{m}$ ). The more humid dust sources in comparison deliver much lower or no carbonates for African dust.

While for the Western African outflow, commonly less than 5% of feldspar particles are identified (even though they are present in slightly larger abundances in Morocco), at Dushanbe and Japan around 10% and more are found, consistent with previous measurements [42]. Also the Arctic dust is similarly rich in feldspar particles, but here they are concentrated at larger sizes (>4  $\mu\text{m}$ ). Looking more into detail, Na-feldspars contribute the majority in Tajikistan, while in African dust Na- and K-feldspars are equally abundant.

Fe oxide/hydroxide particles are found for African dust in approximately doubled abundances from the more humid dust sources, in comparison to the more arid ones. A similar trend can be observed for the Fe content of the silicate grains. In Dushanbe, a population of (probably) chlorite with a high Fe and Mg content is identified, increasing considerably the dust total Fe content.

Apart from the silicate, carbonate and oxide species, dust is internally and externally mixed at all places with different compounds, mainly different sulfates and sea salt. This mixture is moderately (Barbados) to highly (Cape Verde) variable for sea salt in the coastal sampling

locations, depending on meteorological conditions. For sulfate, the variability is higher for transported mineral dust than for source-near measurements in Morocco and Dushanbe. This indicates a source sulfate contribution for these more arid deserts, whereas this contribution is apparently masked by meteorological conditions and subsequent processing for the transported dust.

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