Technical Note: Mineralogical, chemical, morphological, and optical interrelationships of mineral dust re-suspensions

Johann P. Engelbrecht[1], Hans Moosmüller[1], Samuel Pincock[1], Radhakrishna Murty Jayanty[2], Traci Lersch[3] and Gary Casuccio[3]

[1]Desert Research Institute (DRI), Reno, Nevada 89512-1095, U.S.A.
[2]RTI International, Raleigh, North Carolina 27675-9000, U.S.A.
[3]RJ Lee Group, Inc., Monroeville, Pennsylvania 15146, U.S.A.

Correspondence to: Johann P. Engelbrecht (johann@dri.edu)

Abstract This paper promotes an understanding of the mineralogical, chemical, and physical interrelationships of re-suspended mineral dusts collected as grab samples from global dust sources. Surface soils were collected from arid regions, including the southwestern USA, Mali, Chad, Morocco, Canary Islands, Cape Verde, Djibouti, Afghanistan, Iraq, Kuwait, Qatar, UAE, Serbia, China, Namibia, Botswana, Australia, and Chile. The < 38 μm sieved fraction of each sample was re-suspended in a chamber, from which the airborne mineral dust could be extracted, sampled, and analyzed. Instruments integrated into the entrainment facility included two PM_{10} and two $PM_{2.5}$ filter samplers, a beta attenuation gauge for the continuous measurement of PM_{10} and $PM_{2.5}$ particulate mass fractions, an aerodynamic particle size (APS) analyzer, and a three wavelength (405, 532, 781 nm) photoacoustic instrument with integrating reciprocal nephelometer for monitoring absorption and scattering coefficients during the dust re-suspension process. Filter sampling media included Teflon® membrane and quartz fiber filters for chemical analysis, and Nuclepore® filters for individual particle analysis by Scanning Electron Microscopy (SEM). The < 38 μm sieved fractions were also analyzed by X-ray diffraction for their mineral content while the > 75 μm, < 125 μm soil fractions were mineralogically assessed by optical microscopy.

Presented here are results of the optical measurements, showing the interdependencies of single scattering albedos (SSA) at three different wavelengths and chemical as well as mineralogical content of the entrained dust samples. Relationships between the SSA of airborne dusts, and iron (Fe) in hematite, goethite, and clay minerals (montmorillonite, illite, palygorskite) are demonstrated. Differences in clay mineralogy between samples from Mali and those from other localities are highlighted.

Results from this study can be integrated into a database of mineral dust properties, for applications in climate modeling, remote sensing, visibility, health (medical geology), ocean fertilization, and impact on equipment.

Keywords. Dust sources; Re-suspension chamber; Dust mineralogy; Particle size distributions; Single Scattering Albedo (SSA); SEM based particle analysis

1 Introduction

Globally, airborne mineral dust aerosol has the largest mass emission rate, average column mass burden, and average optical depth, of all aerosol types (Satheesh and Moorthy, 2005). More than half (≈ 62%) of the global airborne mineral dust mass is emitted in North Africa (Engelstaedter et al., 2006), with lesser fractions being from Asia (≈ 15%) and the Arabian Peninsula (≈ 11%) (Tanaka and Chiba, 2006). Minerals are among the largest atmospheric aerosol particles, ranging from 0.1 μm to more than 100 μm in diameter. Generally, only particles less than ≈ 10 μm in diameter survive long-range transport (Zender et al., 2003;Maring et al.,

2003), although larger quartz particles (up to ≈ 160 μm) ascribed to Saharan dust storms, were reported from deposits on La Graciosa Island (Menéndez et al., 2014), about 160 km off the west coast of Africa.

The mineralogy of dust from different geological domains differ from each other (Guthrie and Mossman, 1993). How these relate to variations in optical properties influencing radiative effects (e.g., Satheesh and Moorthy, 2005), visibility impairment (e.g.,
5   Watson et al., 1999), and their potential health effects (Fubini and Fenoglio, 2007), is not clearly understood. An interdisciplinary approach, to characterizing mineral dust from different geological source regions provides important information for modeling of radiative effects and climate change (e.g., Satheesh and Moorthy, 2005), remote sensing (e.g., Ginoux et al., 2012), visibility impairment (e.g., Watson, 2002), as well as impact on solar energy production (e.g., Sayyah et al., 2014), ecosystems (e.g., Field et al., 2010), and human health (Middleton et al., 2008;Norboo et al., 1991;Wiggs et al., 2003). The prime objective of this
10 publication is to provide speciated information on mineral dust aerosol optical properties, mineralogy, chemistry, as well as particle morphology and size distribution, information required for assessments of regional and global impacts of mineral dust.

Related previous publications provided geographically limited information on mineral dust optical properties (e.g., Moosmüller et al., 2012;Müller et al., 2009;Petzold et al., 2009;Schladitz et al., 2009;Von Hoyningen-Huene et al., 2009), chemical composition, mineralogy, and particle size distribution (e.g., Al-Dousari and Al-Awadhi, 2012;Avila et al., 1997;Kahlaf et al., 1985;Kandler et
15 al., 2007;Kandler et al., 2009;Molinaroli, 1996); this publication expands on these previous contributions by presenting properties of a global set of re-suspended dust samples applying similar sample preparation and analysis methods.

2 Sampling and analysis

2.1 Sample collection and preparation

Surface soil samples had previously been collected at more than 100 sites worldwide, of which 65 were selected for re-suspension,
20 monitoring, and analysis (Figure 1, Supplement S1). Included are samples from the Middle East [Iraq (6), Kuwait (5), Qatar (1), UAE (1), Afghanistan (3), Djibouti (1)], U.S.A. [Arizona (4), California (2), Nevada (3), Colorado (1), Utah (2)], China [5], Spain [Canary Is. (8)], Morocco [1], Mali [3], Cape Verde Is. [1], Chad [3], Serbia [3], Australia [3], Botswana [4], Namibia [3], Chile [1], as well as one sample of hematite (Fe_2O_3) (Figure 1).

The selected samples, although of variable mineral and chemical compositions, are representative of many major dust regions.
25 However, surface soils from dust sources in parts of North Africa, Middle East, India, Central Asia, South America, Australia, and southern Africa have either not been collected and therefore not included in the sample set, or for reasons related to the samples themselves, not analyzed in this study.

The grab soil samples collected in the field were generally sieved to < 1 mm to remove larger pebbles, plant material, and other unwanted artifacts. Where necessary, they were air-dried in the laboratory, before being labelled, catalogued, and stored in capped
30 plastic bottles. Particle size distributions were performed on the < 600 μm sieved sample sets. Sub-sets of these samples were screened to < 38 μm for mineral analysis by X-ray powder diffraction (XRD), and for re-suspension purposes. Further sub-sets were sieved to > 75, < 125 μm for mineralogical investigation by optical microscopy.

The < 38 μm sieved size fraction of each sample was visually color coded by comparing to the Munsell color charts (Supplement S3.1). The Munsell system specifies the color of soils based on hue, lightness, and color purity (chroma) and has been adopted for
35 soil research by the US Department of Agriculture. Various red or brown hues of the dusts can generally be ascribed to the presence of iron and manganese oxides, and iron bearing clay minerals in the soils.

2.2 Petrographic assessment

Mineral grain mounts for optical microscopy were prepared of the > 75, < 125 µm sieved soil fractions. The mineral grains were mounted in epoxy on a glass slide and ground to a thickness of approximately 30 µm for transmitted light optical microscopy. Petrographic microscopy is particularly suited to the optical identification of mineral grains larger than about 10 µm. Mineral

5   properties such as texture, color, pleochroism, birefringence, refractive index, and twinning, were used to identify transparent silicate minerals. Minerals readily identified in the dust grain mounts in this study include quartz, various feldspars (plagioclases, orthoclase, microcline), amphiboles (hornblende), pyroxenes (augite), micas (muscovite, biotite), chlorite, carbonates (calcite, dolomite), and oxides (Supplement S2.1).

2.3 Mineral analysis by X-ray diffraction (XRD)

10   XRD is the single most important non-destructive technique for characterization of minerals such as quartz, feldspars, calcite, dolomite, clay minerals, and iron oxides in fine dust. A Bruker D8 XRD system with sample stacker and spinner was used for measurement of mineral abundances. The Bruker Topas® software and the relative intensity ratio procedure were applied for semi-quantitative XRD analyses of the < 38 µm screened dust samples (Rietveld, 1969;Chung, 1974;Esteve et al., 1997;Sturges et al., 1989;Caquineau et al., 1997) (Supplement S2.2).

15   ### 2.4 Particle size analysis

Laser particle size analysis (LPSA) was performed on each of the soil samples. The system measures the size-class fractions of a soil or sediment sample in an aqueous suspension, based on the principle that light scatters at angles inversely proportional to, and with intensity directly proportional to particle size (Gee and Or, 2002). The grab samples were sieved to < 600 µm before being introduced to the laser analyzer (Micromeritics Saturn DigiSizer 5200®) in an aqueous medium of 0.005% surfactant (Na

20   metaphosphate). They were then internally dispersed by applying ultra-sonication and circulated through the path of the laser light beam. The measured size-class fractions were grouped as clay (< 2 µm), silt (2 - 62.5 µm) and sand (62.5 - 600 µm) (Engelbrecht et al., 2012) (Supplement S2.3).

2.5 Dust entrainment

A dust entrainment facility was configured with an array of instrumentation, including (i) three-wavelength photoacoustic

25   instrument, (ii) beta attenuation gauge, (iii) aerodynamic particle size analyzer, (iv) desktop computer and (v) PM filter samplers (http://www.dri.edu/das-laboratories/4185-dust-entrainment-and-characterization-facility).

A dust plume was generated by puffing the dust into the re-suspension chamber with compressed air, from which particles were sampled onto filters, together with continuous optical, concentration, and particle size measurements (Moosmüller et al., 2012). Dust measurements were focused on the size fraction associated with long-range transport, typically less than 9 µm in physical

30   diameter (Pye, 1987). All samples were screened through a 38 µm pore size stainless steel sieve prior to re-suspension. The < 38 µm sieved fractions served as source material from which fractions of interest, i.e., PM_{10} (≤10 µm aerodynamic diameter) and $PM_{2.5}$ (≤2.5µm aerodynamic diameter) were separated. The procedure was to place a portion of the < 38 µm dust sample in a conical glass flask, into which a jet of filtered air was injected, transporting the dust plume into the plexiglass-fronted re-suspension chamber. From here the dust aerosol was drawn through size selective inlets, either PM_{10} or $PM_{2.5}$, and directed to the optical,

35   gravimetric, and particle size monitors, as well as to the four PM filter samplers (Figure 2).

The most important optical properties (Bergstrom et al., 2007;Chýlek and Wong, 1995;Hassan et al., 2015) for calculating aerosol radiative impacts are *scattering* (β_{sca}) and *absorption* (β_{abs}) coefficients, from which the *extinction* (β_{ext}) coefficients are calculated, (*scattering* (β_{sca}) + *absorption* (β_{abs}) = *extinction* (β_{ext}),

and

5 $Single\ Scattering\ Albedo\ (SSA) = \frac{\beta sca}{\beta sca + \beta abs} = \frac{\beta sca}{\beta ext},$ (1)

The (Photoacoustic Instrument [PA]); (Arnott et al., 1999;Arnott et al., 2000;Lewis et al., 2008) with an integrated nephelometer for optical characterization of re-suspended mineral dusts provided scattering and absorption coefficients, from which extinction coefficient and single-scattering albedo were calculated. Real-time measurements of aerosol light absorption, due predominantly to light-absorbing minerals (e.g., hematite, Fe_2O_3; goethite, FeO.OH; Fe-bearing clays) in most dusts were accomplished with the

10 integrated PA/nephelometer. Scattering and absorption coefficients were measured for $PM_{2.5}$ dust size fractions at 405, 532, and 781nm wavelengths (Supplements S3.1, S3.2).

These optical coefficients have dimensions of inverse distance (Mm^{-1}), and it is useful to ratio them to aerosol mass concentration ($\mu g/m^3$), yielding aerosol scattering (absorption) mass efficiencies (m^2/g). Optical mass efficiencies can be used in conjunction with modeling of atmospheric mass loadings and concentrations to calculate optical coefficients of the ambient atmosphere, or in

15 reverse, to calculate ambient mass concentrations from optical measurements (e.g., from satellite-based optical remote sensing instruments). Such measurements are most important in the green wavelength (500–550 nm) where the solar spectral power density is greatest. Additional measurements covering the shortwave radiatively important spectrum are significant for aerosol light absorption, because of spectrally varying absorption of some mineral dust particles (Moosmüller et al., 2009).

2.6 Filter analysis

20 Teflon® membrane and quartz fiber filters were conditioned and weighed prior to and after sampling, to measure the particulate mass sampled in the re-suspension chamber during each sampling period.

In the course of this study, about 130 Teflon® filters were analyzed by Energy Dispersive X-ray Fluorescence Spectrometry (EDXRF) (Watson et al., 1999) for chemical elements, including magnesium (Mg), aluminum (Al), silicon (Si), potassium (K), calcium (Ca), titanium (Ti), and iron (Fe). After completion and validation of the EDXRF results, the same 130 Teflon® filters

25 were dissolved in nitric/hydrochloric acid and the solutions analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) for 12 trace metals: antimony (Sb), arsenic (As), beryllium (Be), cadmium (Cd), chromium (Cr), lead (Pb), manganese (Mn), nickel (Ni), vanadium (V), zinc (Zn), mercury (Hg), and strontium (Sr). Water extractions were performed on one half of each of the 130 quartz fiber filters. Aliquots of the extractions were analyzed by Ion Chromatography (IC) (Chow and Watson, 1999) for water soluble anions: sulfate (SO_4^{2-}), nitrate (NO_3^-), chloride (Cl^-), phosphate (PO_4^{3-}), and the ammonium (NH_4^+) cation. Further

30 aliquots of the extractions were analyzed by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES), for water-soluble cations: sodium (Na^+), potassium (K^+), calcium (Ca^{2+}), and magnesium (Mg^{2+}).

 Chemical concentrations for PM_{10} and $PM_{2.5}$ size fractions were combined into normative mineral abundances and also presented graphically. Scatter plots of selected major chemical species were compiled, and elemental ratios tabulated. The relationships between the single scattering albedos (SSA) and the major chemical components are shown as plots.

2.7 Scanning electron microscopic (SEM) based individual particle analysis

SEM-based individual particle analysis was performed on 65 $PM_{2.5}$ filters selected from approximately 130 sampled Nuclepore® filters. The samples were analyzed by two complementary methods, the first being Computer Controlled Scanning Electron Microscopy (CCSEM), and the second high magnification SEM to obtain secondary electron images (SEI).

5   CCSEM is a combination of Backscattered Electron Imagery (BEI) and Energy Dispersive Spectroscopy (EDS) which automatically analyses a large number (more than 1,000) of individual particles, for chemical composition, particle size, and shape. The measurements on the particles are grouped into "bins" by chemical composition and particle size. The CCSEM results were routinely grouped into 29 chemical categories or "bins". Following on preliminary statistical evaluation of the chemical data and *a priori* information of the mineralogical composition (from XRD, optical microscopy, and particle morphology) the number of

10   chemical „bins" were reduced to 15. Mineral names were assigned to each of the chemical categories, e.g., Si rich category as quartz, Ca rich category as calcite, Ca, Mg category as dolomite, Ca, S category as gypsum, Fe rich category as iron oxides, Ca, Al, Mg category as clay, Ca, Al, Si category as Ca feldspar, Na, Al, Si category as Na feldspar, K, Al, Si category as K feldspar, etc. Summary plots of the inferred mineral compositions were compiled. Individual particle morphology including aspect rations were measured by CCSEM. Although labour intensive, the field emission electron source allows for high magnifications and sharp

15   secondary electron images (SEI). This technique further allows for the study of particle shape, surface coatings, and chemical composition. Approximately five SEIs with energy dispersive spectra (EDS) for each of the 65 $PM_{2.5}$ filter samples were collected in this fashion.

3 Results

3.1 Sample descriptions

20   All results are compiled in Supplementary material to this paper, with the sampling sites listed in Supplement S1, mineralogy by optical microscopy in Supplement S2.1, mineralogy by XRD in Supplement S2.2, particle size fractions (volume %) distributions of the < 600 µm sieved particle fractions in Supplement S2.3. Munsell color indices and single scattering albedos (SSA) are listed in Supplement S3.1, and together with particle size statistics and SSA statistics in Supplement S3.2. Also tabulated on each sample sheet in Supplement S3.2 are beta attenuation gauge mass measurements for PM_{10} and $PM_{2.5}$ collected in parallel with the SSA

25   and particle size analysis. Mass measurements for PM_{10} and $PM_{2.5}$ on Teflon filters and PM_{10}/$PM_{2.5}$ mass ratios were also collected as controls on the same day. Where measured, statistics on the SEM based aspect ratios are tabulated on the same sheets. The chemical results of the PM_{10} and $PM_{2.5}$ dust samples on filters are tabulated in Supplement S4.1, normative mineral calculations from the chemical results tabulated in Supplement S4.2 and plots therefrom shown in Supplement S4.3. Scatter plots of the major chemical species as oxides are shown in Supplement S4.4, the relationships between chemical compositions and SSA in

30   Supplement S4.5, and ratios of selected major components in Supplement S4.6. The scanning electron microscopic based individual particle results collected on $PM_{2.5}$ Nuclepore® filters are shown in Supplements S5.1 – S5.4. The individual particle results tabulated in 29 chemical bins are given in Supplement S5.1, their mineral interpretations in 15 mineral bins in Supplement S5.2 with associated plots of the last mentioned in Supplement S5.3. Statistics on the SEM based aspect ratios measured on most samples are tabulated in Supplement S5.4 and shown on Supplement S3.2. Supplement S5.5 shows the energy dispersive (EDS)

35   spectra and associated secondary electron images (SEI's) of selected individual particles from each re-suspended dust sample. Descriptions of samples from each geographic region can be summarized as follows.

3.1.1 Southwestern U.S.A. – Owens Lake, CA (S1019)

The dried lakebed contains minerals washed in from the adjoining Sierra and Inyo Mountains by the Owens River as well as small amounts of evaporite deposits. Since about 1926 the lake remained dry as a result of overuse of its water for agriculture, and human consumption by the city of Los Angeles (Wilkerson et al., 2007). The coarser sand and silt fractions contain plagioclase, calcite,

5   quartz, biotite mica, and hornblende, with halite, kaolinite, and trona in the finer fractions. These are abundant in local aerosols generated during dust events, thereby creating a major health issue along the Owens Valley and poor visibility at the nearby China Lake Naval Air Station (Reheis, 1997;Wilkerson et al., 2007). This led to ongoing mitigation steps to contain alkaline dust being blown off the playa. (www.ladwp.com/OwensLake).

3.1.2 Southwestern U.S.A. – Black Rock, NV (S1045)

10   The Black Rock playa about 150 km north of Reno is known for its very flat lakebed, with approximately one meter altitude variation over an area of 300 km^2. The lakebed is normally wet during the winter and spring, from water and fine sediment fed from the Quinn and Mud Meadows Creek rivers. During the other seasons the dry playa is used for multiple recreational activities, including the annual Burning Man (http://www.burningman.com/) event each August. As a result, the dry lakebed is continually being disturbed by human and vehicular traffic, creating intense dust palls during windy conditions (Adams and Sada, 2010). The

15   surface sediment of the playa is fine silt and clay, illite, with small amounts of calcite, quartz, halite, kaolinite, feldspars, hornblende, and sepiolite.

3.1.3 Southwestern U.S.A. – NW Reno, NV (S1052, S1053)

Two sedimentary samples were collected along the Peavine Mtn. foothills in northwest Reno. The first (S1052) is an off-white diatomaceous lakebed deposit (Smedman, 1969), containing diatoms (Figure 3), plagioclase, quartz, and lesser amounts of clay

20   (kaolinite, sepiolite, illite, montmorillonite), stilbite, and biotite. The second (S1053) is a yellowish brown soil, the color being ascribed to iron oxides (goethite), in addition containing quartz and plagioclase and smaller amounts of hornblende, biotite, muscovite, and clays (palygorskite, montmorillonite, kaolinite).

3.1.4 Southwestern U.S.A. – Carbondale, CA (S1065)

Carbondale Red Clay (CRC) is a commercially available fireclay, considered to be the equivalent of Martian dust (White et al.,

25   1997). The material is composed of very fine quartz and kaolinite with a high percentage of hematite (Figure 4).

3.1.5 Southwestern U.S.A. – Yuma, AZ (S3003, S3004)

The two grab samples were collected from improved artillery firing pads at the Yuma Proving Ground (YPG) (Gillies et al., 2007). The samples contained major amounts of quartz and calcite, together with lesser amounts of dolomite, mica (biotite, muscovite), feldspars (plagioclase, microcline), hornblende, gypsum, and clays. To compact the soil and contain the dust during the gun firing

30   activities, brines containing potassium sulfate and other salts were sprayed onto the firing pads. Trace amounts of copper, bismuth, and lead from the spent ordnance are present in the finer dust fractions (Engelbrecht et al., 2012).

3.1.6 Southwestern U.S.A. – Yuma, AZ (S3008)

This surface soil sample was collected during a field campaign measuring dust emissions created by rotary-winged aircraft (Gillies et al., 2010). The sample contains rounded quartz grains together with calcite, feldspars (plagioclase, microcline), and mica (biotite,

35   muscovite) and small amounts of hornblende, clay (kaolinite), dolomite, gypsum, and apatite (Engelbrecht et al., 2012).

3.1.7 Southwestern U.S.A. – Ft. Carson, CO (S3011)

This sample contains quartz, plagioclase, and calcite with lesser amounts of microcline, hornblende, biotite, muscovite, dolomite, gypsum, and kaolinite (Engelbrecht et al., 2012).

3.1.8 Southwestern U.S.A. – Dugway, UT (S3016, S3017)

5   The two samples from Utah are very similar to each other, both taken along a dirt road. They contain major amounts of quartz and calcite (Figure 5), and lesser amounts of dolomite, aragonite, plagioclase, with traces of biotite, muscovite, illite, and orthoclase.

3.1.9 Middle East – Djibouti (S2001)

Djibouti lies in the Afar triangle, which is part of the African continental rift zone, underlain by basaltic lava flows and associated sediments. Minerals in the screened dust sample including pyroxenes, amphiboles, olivines, plagioclases, ilmenites, and magnetites

10   are also contained by the surrounding rock outcrops. The nearly 10% calcite in the sample could be from raised coral reefs in the surrounding Red Sea (Engelbrecht et al., 2009b;Engelbrecht et al., 2008).

3.1.10 Middle East – Afghanistan (S2002, S2003, S2016)

The first two sites are in northeastern Afghanistan in the mountainous region close to the Pakistan border, while the third is in the Helmand province. Dust storms originating in the southwestern part of the country move in a counter-clockwise direction,

15   funneling wind-blown dust toward Kabul, Bagram, and Khowst. The Bagram site is underlain by loess (wind deposited silt), sand, and clay, and the Khowst site by alluvium containing gravel, sand, silt, and clay. These two samples from Afghanistan are mineralogically similar, both containing major amounts of quartz, calcite, and clays (kaolinite, illite, palygorskite), with smaller amounts of feldspar, mica (biotite, muscovite), chlorite, and amphibole (Engelbrecht et al., 2009b;Engelbrecht et al., 2008). The sample from Helmand Province (S2016) contains quartz (Figure 6) and calcite with small amounts of clays, gypsum, and silicates.

20   **3.1.11 Middle East – Qatar (S2004) and UAE (S2005)**

Qatar and United Arab Emirates (UAE) are impacted largely by windblown dust transported from the Arabian Peninsula, Iraq, and Kuwait. Samples from both sites contain large amounts of calcite, dolomite, quartz, and feldspar and lesser amounts of chlorite, clay minerals, gypsum, anhydrite, and feldspar. The Qatar sample also contains small amounts of hornblende and ilmenite, probably transported from weathered volcanics in Saudi Arabia to the west of the sampling site (Engelbrecht et al.,

25   2009b;Engelbrecht et al., 2008).

3.1.12 Middle East – Iraq (S2006, S2007, S2008, S2009, S2010, S2011)

Satellite imagery shows dust storms, blowing across Iraq and Kuwait from salt pans and wadis along the border with Turkey and Syria (Engelbrecht et al., 2009b), being major sources of windblown dust in the region. The Baghdad site, as well as the three sites further north and close to the Tigris River (Taji, Balad, and Tikrit), contain major amounts of quartz, calcite, and feldspar, plus

30   smaller amounts of gypsum, micas, and clays. The Tallil sample, collected further to the south and close to the Tigris–Euphrates river confluence also contains the mineral gypsum. This sampling site is underlain by poorly drained river-basin soils, and the gypsum could be wind-entrained from the gypsiferous desert immediately to the southwest of this site. Al Asad, closer to the Euphrates River and 200 km to the west of Baghdad, differs from the other five Iraqi sites, in that its soil contains largely dolomite, with small amounts of gypsum, and quartz. This region is underlain by soils containing gypsum, limestone, and sandstone (Buringh,

1960). XRD analysis of the sieved dust samples confirms that the limestone in this region is largely dolomite, instead of calcite identified at the other five Iraq sampling sites (Engelbrecht et al., 2009b;Engelbrecht et al., 2008).

3.1.13 Middle East – Kuwait (S2012, S2013, S2014, S2015, S2017)

The samples from Kuwait contain higher fractions of silt than those from adjacent Iraq, with major amounts of quartz and calcite.

5   Samples from all five sites also contain a significant amount of feldspar, mostly plagioclase and potassium feldspar (Figure 7) as well as micas (muscovite, biotite) and clay minerals. Previous studies on fallout dust in Kuwait (Al-Awadhi, 2005) identified the clay minerals as palygorskite, together with illite-montmorillonite and a smaller proportion of kaolinite. Al-Awadi (2005) and Kahlaf et al. (1985) suggest that most of the dust in Kuwait is transported from the dry Mesopotamian floodplain in southern Iraq and other parts of the Arabian peninsula (Engelbrecht et al., 2009b;Engelbrecht et al., 2008).

10 **3.1.14 Northern Africa – Canary Islands (Las Canarias) [Lanzarote (1005, S1006, S1007, S1008), Gran Canaria (S1027), Fuerteventura (S1033, S1034, S1035)]**

The Canary Island archipelago lies along the westward pathway of Saharan dust blowing off the west coast of North Africa, the islands being impacted by severe dust events in that region (Alastuey et al., 2005;Bergametti et al., 1989;Criado and Dorta, 2003;Gelado-Caballero et al., 2005;Maring et al., 2000;Menéndez et al., 2009;Pérez-Marrero et al., 2002;Viana et al.,

15 2002;Kandler et al., 2007;Engelbrecht et al., 2014;Coudé-Gaussen et al., 1987;Mizota and Matsuhisa, 1995). It was shown that fractions of transported Saharan dust had been deposited on the islands, contributing to the formation of local soils (Jahn and Stahr, 1996;Menéndez et al., 2014;Mizota and Matsuhisa, 1995;Muhs et al., 2010;von Suchodoletz et al., 2013;Zarei, 1989;Menéndez et al., 2007;von Suchodoletz et al., 2009;Williamson et al., 2004;Moreno et al., 2001). It can be assumed that samples of soils collected on these islands contain Saharan dust in variable proportions. Two samples from Fuerteventura are from dust traps in

20 lava flows, on the eastern side of the island, approximately 100 km off the coast of Morocco. The sieved dust samples from the Canary Islands generally contain quartz as the major component, with minor amounts of calcite, feldspars, amphibole, pyroxenes, micas, clays, iron oxides, and volcanic glass.

3.1.15 Northern Africa – Cape Verde (S1013)

The Il de Sal Island is part of the Cape Verde (Cabo Verde) archipelago, approximately 700 km off the west coast of Africa, and

25 considered to be within the Sahel geographic zone. As with the Canary Islands, the Cape Verde islands are impacted by dust blowing off the west coast of Africa. The soil sample collected on the north side of Sal Island is assumed to contain trapped Sahara and Sahel dust. The sample contains plagioclase and pyroxene in the coarse fraction, possibly from local weathered volcanic rocks, together with quartz, illite clay, calcite, carbonate apatite, halite, and kaolinite in the fine fraction. The carbonate apatite (francolite) (Figure 8) is possibly of local biogenetic origin.

30 **3.1.16 Northern Africa – Morocco (S1025)**

Morocco is considered a major source of dust blowing across the Mediterranean into Europe. Several studies were performed on airborne mineral dust from this region, some as part of the Sahara Mineral Dust Experiment (SAMUM) (Heintzenberg, 2009;Kandler et al., 2009;Schladitz et al., 2009;Müller et al., 2009;Petzold et al., 2009;Von Hoyningen-Huene et al., 2009;Scheuvens et al., 2013;Molinaroli, 1996;Molinaroli et al., 1993;Blanco et al., 2003;Avila et al., 1997;Jahn and Stahr, 1996).

35 The major minerals in this sample are quartz and illite, with smaller amounts of calcite, feldspars (plagioclase, orthoclase), hornblende, dolomite palygorskite (Figure 9), and kaolinite.

3.1.17 Northern Africa – Mali (S1009, S1010, S1011)

The three grab samples were collected in a field approximately 5 km to the northwest of Bamako in Mali, all within 200 m of each other. The sampling was from soils immediately to the south of the Sahel, the latter being defined as that zone adjoining the southern Sahara which receives on average between ~200 and ~600 mm rainfall per year. The samples are from within a tropical

5   region where lateritic soils are developed, containing high amounts of hematite, goethite, and kaolinite (Figure 10) (Roquin et al., 1989;Roquin et al., 1990).

It was shown (Oldfield et al., 2014) that the magnetic properties of dusts and their hematite/goethite contents could be applied to distinguishing dust sources from North Africa impacting the Caribbean region. Moosmüller et al. (2012) showed that the optical properties such as single scattering albedo were controlled by the iron (Fe) content of airborne dust. Roquin et al. (1989) was able

10   to correlate the mineralogical and chemical information from Dagadamou region in southern Mali, with the SPOT satellite imagery.

3.1.18 Northern Africa – Chad (S1049, S1050, S1051)

The three dry lakebed deposits were collected from the Bodélé depression in the southern Sahara region of North Africa. Bodélé is considered the largest single source of global dust (Chappell et al., 2008;Goudie and Middleton, 2001;Tegen et al., 2006;Todd et al., 2007;Washington and Todd, 2005;Middleton and Goudie, 2001), contributing to the fertilization of the Atlantic Ocean, and

15   land as far as the Amazon basin (Ben-Ami et al., 2010;Koren et al., 2006;Chadwick et al., 1999;Formenti, 2001;Bristow et al., 2010).

The samples contain largely amorphous silica (diatoms) (Figure 11), quartz, and calcite with small amounts of plagioclase, potassium feldspar, kaolinite, illite, and halite.

3.1.19 China – Northwest (S1014, S1016, S1017, S1018)

20   The four samples are from far northwestern China, two (S1017, S1018) being from a locality referred to as Daemon City, named after the unusual wind eroded sandstone formations. The two samples are composed of major amounts of quartz and calcite, with plagioclase, kaolinite, mica (biotite, muscovite), and montmorillonite. The two samples further north (S1014, S1016) from the Daemon City area contain quartz and biotite, with lesser amounts of plagioclase, calcite (Figure 12), hornblende, kaolinite, illite, sepiolite, and halite.

25   ### 3.1.20 China – Loess Plateau (S1055)

This region in China contains the greatest thickness of loess in the world. It is an un-stratified silt deposit formed by wind-blown fluvio-glacial deposits related to the geologically recent ice ages (Derbyshire et al., 1998;Kohfeld and Harrison, 2003;Sun et al., 2000;Lewis et al., 1984). This silt sample contains quartz with smaller amounts of calcite, plagioclase, potassium feldspar, biotite, hornblende, clay (kaolinite, illite), and mica (muscovite, biotite).

30   ### 3.1.21 Southern Africa – Botswana (S1038, S1039, S1040, S1041)

The samples are from the dry lakebed of Makgadikgadi Pan in central Botswana. The samples contain largely calcite, quartz, the evaporate minerals trona and halite, and smaller amounts of sepiolite, illite, dolomite, plagioclase, orthoclase, muscovite, hornblende, and dolomite. During the dry winter months, the Makgadikgadi Pan is a major source of dust in southern Africa.

3.1.22 Southern Africa – Namibia (S1022, S1023, S1024)

Three soil samples were collected along the southern and western edge of Etosha Pan in northern Namibia. They all contain major amounts of the evaporate minerals halite, thenardite, and calcite, with sepiolite and small amounts of dolomite, plagioclase, muscovite, and amphibole (Buch and Rose, 1996). Etosha Pan, especially during the dry winter season, is a major source of dust

5   in Namibia, exacerbated by disturbance of the lakebed being from the passage of large herds of game.

3.1.23 South America – Chile (S1042)

The sample was collected at the Yungay (Rock Garden) site in the Atacama Desert of Chile, considered to be one of the most arid regions of the world (McKay et al., 2003), also described as Mars-like (Navarro-González et al., November 2003). The sample contains quartz as its main constituent, together with clay (illite, montmorillonite), gypsum, anhydrite, plagioclase, calcite,

10 orthoclase, biotite, and amphibole.

3.1.24 South Australia (1056, S1057, S1058)

The three samples are from riverbed or lakebed deposits in the Lake Eyre and Lake Frome regions of South Australia (Marx et al., 2009;McGowan and Clark, 2008.;Moreno et al., 2009), considered to be a major source of dust in the southern hemisphere, being deposited on the snow as far as the New Zealand Alps (McGowan et al., 2005;Marx et al., 2009). Evidence of dust from this region

15 was also found in East Antarctica (Revel-Rolland et al., 2006). The samples are composed largely of quartz grains (Figure 13) coated with clays, together with small particles of clay minerals (illite, kaolinite, palygorskite), gypsum, calcite, feldspar (plagioclase, orthoclase), and dolomite.

3.1.25 Europe – Serbia (S1060, S1062, S1064)

The loess samples were collected along the Danube valley in Serbia, representing loosely bound wind-blown silt deposits, related

20 to the ice periods of the recent geological past (Marković et al., 2009;Marković et al., 2011). The three samples are similar, being composed mainly of rounded particles of quartz (Figure 14), with muscovite, biotite, clay (illite, kaolinite, montmorillonite), plagioclase, dolomite, and calcite.

3.2 Discussion

3.2.1 Particle size analysis

25 Particle size analysis on the < 600 μm sieved samples are tabulated in Supplement S2.3.

From field and laboratory measurements on dust from the western U.S.A., Engelbrecht et al. (2012) found dust emissions to be in part dependant on the particle size distributions of the soils being disturbed. Surface soils with silt contents greater than 50% and less than about 10% clay, had the greatest likelihood of being sources of substantial dust emissions. On the USGS soils ternary plot (Figure 15), soils with the greatest potential to generate dust when disturbed are those that plot in the bottom one third of the

30 "silt loam" field. Included are two previously characterized dust sources from Yakima Test Center (Engelbrecht et al., 2012), three from the Bodélé Depression (S1049-S1051) (Washington and Todd, 2005;Koren et al., 2006), three loess samples from the Danube River valley (S1060, S1062, S1064) (Marković et al., 2011;Marković et al., 2009), one sample from Kuwait (S2017) (Al-Dousari and Al-Awadhi, 2012;Draxler et al., 2001;Kahlaf et al., 1985), and two loess samples from China (S1017, S1055) (Burbank and Jijun, 1985;Derbyshire et al., 1998). It also includes four silt samples collected on Fuerteventura (S1033, S1034), Lanzarote

35 (S1007), and Gran Canaria (S1027) in the Las Canarias archipelago, assumed to consist largely of dust from the western Sahara

(Menéndez et al., 2014), as well as one diatomaceous silt sample from Reno, USA (S1052). This particle size criterion provides an important measure for whether a site or region has the potential to be a significant dust source.

Optical properties such as SSA are also dependent on the particle size distributions (van Staveren et al., 1991,Mourant, 1997 #73;Moosmüller and Arnott, 2009) as well as particle shape (Alexander et al., 2013) and mineralogy. For this project we measured
5   scattering and absorption coefficients for PM$_{2.5}$, thereby confining the particle size distributions.

3.2.2 Chemistry - mineral associations

Dust samples are composed of mineral mixtures in variable proportions, either individual minerals, intergrown with each other, coatings, or composite mineral clusters. Their chemical compositions can in many instances be related to their mineral constituents, as verified by XRD and SEM-based individual particle analysis.

10   The correlation matrix (Table 1) provides a general assessment of chemical species associated with specific minerals in the suite of samples. The high correlation of 0.91 between water soluble sodium (Na$^+$) and sulphate (SO$_4^{2-}$)is ascribed to the mineral thenardite (Na$_2$SO$_4$) identified by XRD in two samples (S1023, S1024) from Etosha Pan, a dry lake bed in Namibia. Some sodium (Na$^+$) is also correlated to the chloride (Cl$^-$), pointing to the evaporite mineral halite (NaCl), often found in dry lakebeds.

Aluminum (Al) is highly correlated with iron (Fe), as well as titanium (Ti) and manganese (Mn), pointing to these elements
15   occurring together in clay minerals such as montmorillonite and palygorskite. Also, the correlation of silicon (Si) with aluminum (Al) and potassium (K) points to the clay mineral illite or possibly potassium feldspar. The correlation (0.42) between calcium (Ca) and magnesium (Mg) is ascribed to these species occurring together in the carbonate mineral dolomite (CaMg[CO$_3$]$_2$). There is also a distinct negative correlation between aluminum (Al) on the one side, and calcium (Ca) and magnesium (Mg) on the other, pointing to an inverse relationship between the clay minerals and the carbonates (calcite, dolomite) in the aerosol samples, i.e.,
20   the higher the clay content in the sample, the lower the carbonates, and *vice versa*. The inverse relationship between clays and carbonates shown by some samples is also evident in the plot of calcium (expressed as CaO) against aluminum (expressed as Al$_2$O$_3$) (Figure 16, Supplements S4.4, S4.6).

The scatter of CaO values shown in Figure 16 is ascribed to the fact that calcium not only occurs in calcite (CaCO$_3$), but also in dolomite (CaMg[CO$_3$]$_2$) and to a lesser extent in plagioclase feldspars (NaAlSi$_3$O$_8$ - CaAl$_2$Si$_2$O$_8$), gypsum (CaSO$_4$.2H$_2$O), and salts
25   occurring in evaporite deposits.

For all samples, the reasonably strong correlation (0.79) between iron (Fe) and aluminum (Al) is shown in Table 1, with most (136) samples falling into one oblong shaped cluster (Figure 17) with a linear correlation of 0.76. The median Fe/Al ratio for this group is 0.86, varying between a Fe/Al ratio of 0.41 and 3.78. This brackets the range found for re-suspended PM$_{10}$ (Fe/Al = 0.87 − 1.38) and PM$_{2.5}$ (Fe/Al = 0.74 − 1.10) samples from the Middle East (Engelbrecht et al., 2009b), and is similar to ambient PM$_{2.5}$ samples
30   (Fe/Al = 0.77 − 1.11) previously collected in the same region (Engelbrecht et al., 2008;Engelbrecht et al., 2009a). These are not unlike the Fe/Al = 0.95 − 1.85 measured by Jaafar et al. (2014) in the course of dust events in Beirut, Lebanon. Chemical analysis of five total suspended particle (TSP) dust samples from western Iran (Najafi et al., 2014) yielded Fe/Al ratios in the range of 0.64 − 0.81. Al-Awadhi (2005) found the Fe/Al mass ratio from fallout dust in Kuwait to vary on average from 0.23 for the light mineral fractions to 0.97 for the heavy mineral fractions.

35   A set of 28 Saharan dust samples collected from the Bodélé Depression in Chad (Bristow et al., 2010) were analyzed for a number of chemical species and have a Fe/Al mass ratio in the range of 0.53 − 1.71. In contrast, soil samples collected along the southern Sahel region in Mali (Figure 17) were different (Roquin et al., 1990) with Fe/Al mass ratios for the ferricretes ranging from 2.95 to 3.43. Prospero (2001) found an average Fe/Al mass ratio of 0.55 for PM$_{10}$ and PM$_{2.5}$ samples collected in south-central Florida,

having been transported across the Atlantic Ocean from Saharan Africa. Surface soils from Mongolia and China analyzed by Xuan (Xuan, 2005) were also in the lower range, with Fe/Al = 0.33 – 0.50.

3.2.3 Optical (SSA) – mineral relationships

SSA for each of the re-suspended $PM_{2.5}$ samples measured at three wavelengths: blue (405nm), green (532nm) and red (781nm)

5   on re-suspended $PM_{2.5}$ dust samples are given in Supplements S3.1 and S3.2. The SSA of mineral dust aerosols is determined by size distribution, morphology, and complex refractive index (Jeong and Nousiainen, 2014;Moosmüller and Arnott, 2009;Sokolik et al., 2001). In our case, constraints were placed on the size distributions by the use of a $PM_{2.5}$ size selective inlet, limiting the particle size for all samples to an average geometric mean diameter of 1.19 μm and a standard deviation 0.11 μm (Supplement S3.2). The aspect ratios of particles in the samples varied in the geometric mean range of 1.41 to 1.70 (Supplement S5.4). The

10   relationships between SSA, iron (as Fe_2O_3), and aluminum (as Al_2O_3) mass fractions at 405nm are shown in Figures 18(a) and (b) (also Supplement S4.5).

As was previously shown (Moosmüller et al., 2012), there is a direct relationship between the iron (Fe_2O_3) content of the samples and the resultant SSA, most evident at 405nm (Figure 18(a)). From the mineralogical information gathered from the optical (Supplement S2.1), XRD (Supplement S2.2), and SEM based individual particle analysis (Supplements S5.1, S5.2), the chemical

15   compositions are evidently related to specific mineral groups. Shown in Figures 18(a) and (b) are one large and three much smaller mineral groups, the main one being the iron (Fe) bearing clays (illite, montmorillonite, palygorskite), and the other from Mali where the iron (Fe) is predominantly in the hematite or goethite with kaolinite (without Fe) as the dominant clay mineral (Roquin et al., 1990). The Carbondale sample was found to contain the iron mineral hematite and the clay mineral kaolinite while the Hematite group contained nearly pure hematite (Fe_2O_3) (Figure 18(a)). From the differences in the distribution patterns, it is evident

20   that the SSA is not only determined by the iron (Fe) content alone, but by the minerals in which iron (Fe) occurs. In the primary clay group (illite, montmorillonite, palygorskite) the iron (Fe) is contained in the crystal structure (Figure 19), but may possibly occur as nanometer-size interlayers of hematite or goethite. In the case of the samples from Mali, the iron (Fe) is contained in variable proportions of hematite (Fe_2O_3) or goethite (FeO(OH)) particles (Figure 20), since no iron *per se* is contained in the kaolinite crystal structure. The Carbondale sample contains hematite as the only iron (Fe) bearing mineral. Absorption in the 404

25   nm (also 532 nm) is stronger for hematite and goethite, than for Fe-bearing clay minerals, with the resultant lower SSA for the dust samples where hematite and goethite were identified as individual mineral grains.

It is evident that pure hematite (S1000) has the lowest SSA (0.407) at 405 nm, this also holds true at 532 nm (SSA = 0.487) and 781 nm (SSA = 0.976). Among dust aerosols, Carbondale Red Clay has the lowest SSA at 405 nm (SSA = 0.629) and at 532 nm (SSA =0.864). However, at 781 nm, the Carbondale sample has a slightly higher SSA (0.991) than two of the Mali samples (S1010: SSA = 0.981 and S1011: SSA = 0.988). The three Mali samples (S1009, S1010, S1011) also have the next lowest SSA values at

30   405 nm (see Figure 18) and at 532 nm. The highest SSA values were reported for the Etosha, Fischer Pan sample S1022 from Namibia with SSA = 0.991 (405 nm), 0.997 (532 nm), and 0.999 (781 nm) closely followed by those of two Bodélé Depression samples (S1049, S1050) and one from Peavine Mountain with identical values of SSA = 0.982 (405 nm), 0.995 (532 nm), and 0.999 (781 nm).

4 Conclusions

A holistic approach (Smuts, 1986) to the analysis of mineral dusts provided us with a comprehensive understanding of their optical, physical, mineralogical and chemical properties and useful information that can be applied in remote sensing, modeling, global climate, health, and other studies.

5 The suite of surface soil samples investigated here are representative of many recognized dust source regions of the globe. At the time of this study, we did not have samples from all dust regions of North Africa, the Middle East, Asia, Australia and South America. The purpose of our study was to provide an understanding of the optical, physical, mineralogical, and chemical interrelationships of a suite of re-suspended dust samples. It can be argued that soil samples re-suspended in the laboratory may be representative of surface wind generated dust, part of which (e.g., $PM_{2.5}$) could be lofted and transported over large distances across

10 the globe. However, it is recognized that dusts may be modified as they mix with other aerosols, and age under changing meteorological conditions and over time.

The individual analytical procedures each provided an important component to an understanding of mineral dust properties. Laser based particle size analysis of the $< 600\mu m$ sieved sample provided information on the silt content of the soils, and the likelihood of dust being suspended when disturbed. Petrographic microscopy on the $> 75, < 125\mu m$ sieved soil fractions readily identified the

15 minerals such as quartz and feldspars which make up the coarse fractions of dust. XRD provided quantitative information on the mineral content of the $< 38\mu m$ sieved soil fractions. The chemical results (XRF, ICP-MS, ICP-OE, IC) of the PM_{10} and $PM_{2.5}$ size fractions were related to their mineralogical contents as well as to better characterizing dust sources and other pollutants. Qualitative information on the mineralogy of the dusts, obtained from optical microscopy, XRD, and SEM, allows for their chemical compositions to be calculated to normative mineral compositions. Such normative mineral compositions are useful when

20 comparing samples to each other. SEM provided information on the individual particle size, shape, and chemical composition which in most cases can be related to the mineralogy of the samples. Mineral coatings of clay and intergrowths often mask the identity of the underlying minerals and need to be considered when interpreting optical measurements.

Two major groups of iron bearing minerals were identified, the largest being the clay mineral group (illite, montmorillonite, palygorskite) which contains variable amounts of iron in the crystal structure, or as oxide or hydroxide interlayers. Iron can also

25 occur as nano-size iron oxides or hydroxides in the clay particles, although no evidence of this was found. The other smaller group includes samples from the subtropical region south of the Sahel, where the clay mineral in the laterites was found to be largely kaolinite, with the iron as individual oxide (hydroxide) minerals hematite or goethite. The SSA measured on the $PM_{2.5}$ size fraction samples, more obvious in the blue wavelength (i.e., 405 nm), varied not only with the iron (Fe) content but also with the mineralogy, i.e., whether the iron was contained in a clay mineral (illite, montmorillonite, palygorskite) or as iron oxide (hematite) and

30 hydroxide (goethite).

Understanding the dust mineralogy, especially of the clay minerals, is of prime importance to our understanding of the optical properties of airborne mineral dusts. This paper greatly expands on our previous results from a limited sample set, showing a linear relationship between SSA and iron content (Moosmüller et al., 2009) showing that this relationship is also controlled by sample mineralogy and mineralogical interrelationships.

35 **Data availability**

Mineralogical, chemical, physical, and optical results are presented in tables and figures in the Supplement to this paper. Requests for data from this study can be directed to Johann Engelbrecht (johann@dri.edu) at the Desert Research Institute (DRI) in Reno, Nevada.

Author contribution

Johann Engelbrecht was the principal investigator and manager for the research project, responsible for the design of the program, configuring the hardware, experimental activities, data interpretation, compilation of results, and compiling this paper. Hans Moosmüller was the co-principal investigator for the project, formulating the problem, advising on the experimental design,

5 interpretation of results, and compiling parts of this paper. Samuel Pincock was responsible for a major part of the experimental work in the re-suspension laboratory, including the collection and validation of measurements. R.K.M. Jayanty from RTI International was responsible for the chemical analysis of filter samples. Gary Casuccio and Traci Lersch from RJ Lee Group were responsible for the SEM based individual particle analysis.

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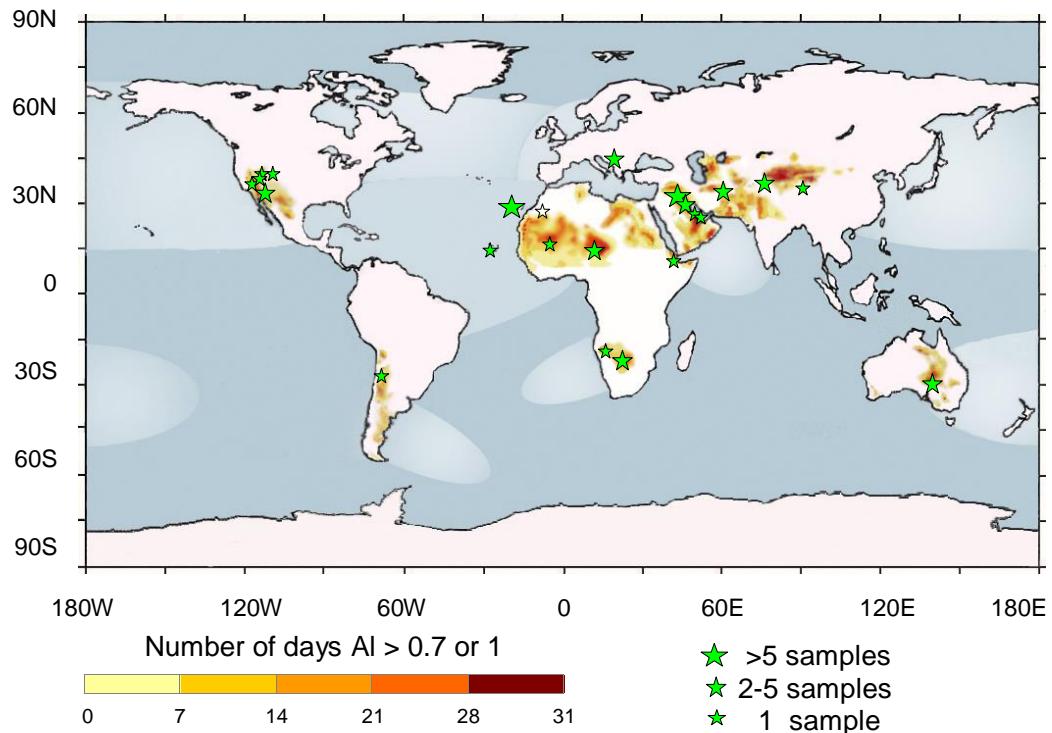
7 Figures

5   **Figure 1: Surface soil sampling regions shown as green stars on a global map of dust sources as identified from Total Ozone Mapping Spectrometer (TOMS) data (Prospero et al., 2002). The size of the marker is approximately proportional to the number of samples collected in the region.**

Figure 2: Integrated dust entrainment facility: Three-wavelength photoacoustic instrument (DMT PASS-3) on rack to the left, re-suspension chamber on trolley in the center, with a particle size analyzer and beta-attenuation gauge installed below the chamber, and an array of vacuum pumps on rack to the right.

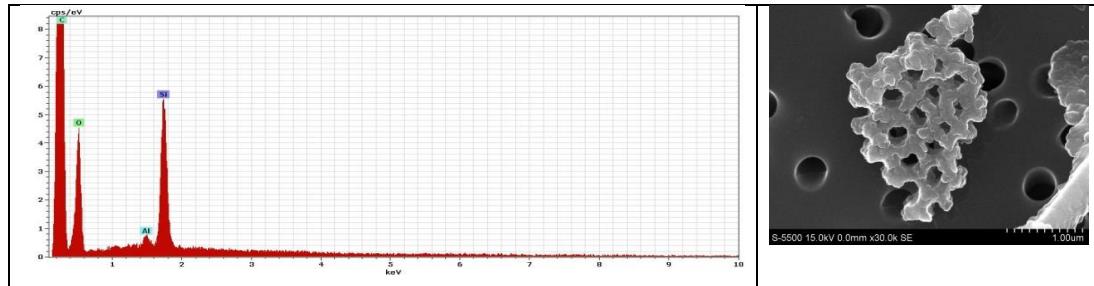



Figure 3: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1052 from Reno, Nevada: A fragment of a diatom containing a major component of silicon (Si), with trace amounts of aluminum (Al) pointing to a silica particle (amorphous?) with a minor amount (coating?) of clay, possibly kaolinite.

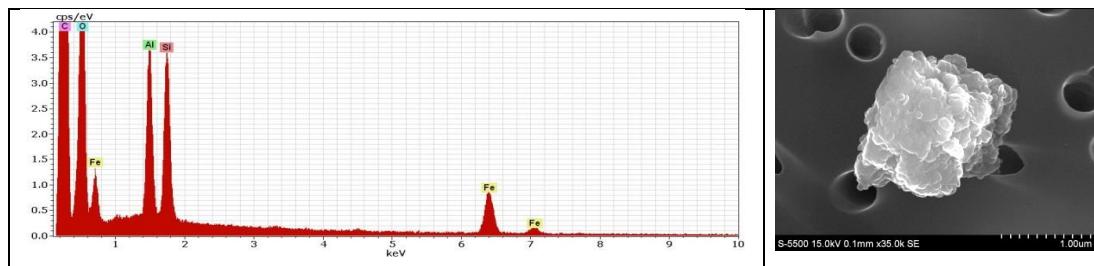



Figure 4: SEM based EDS spectrum and SEI of a particle from re-suspended Sample S1065 (red clay) from Carbondale, California. Composite particle containing major components of aluminum(Al) and silicon (Si) possibly the clay mineral kaolinite, as well as iron (Fe) in hematite or goethite.

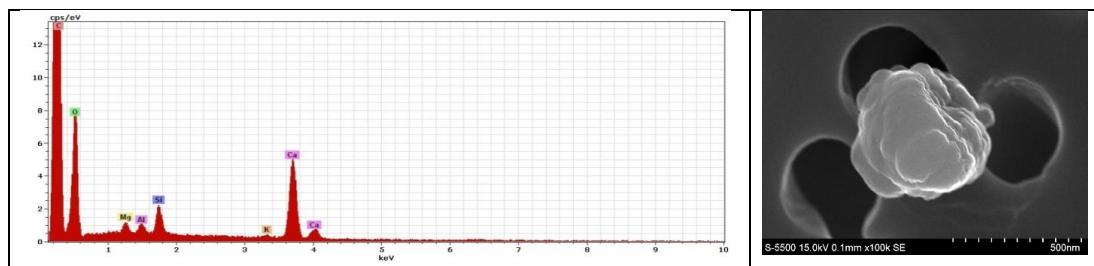



Figure 5: SEM based EDS spectrum and SEI of a particle from re-suspended sample S3017, from Dugway, Utah. Particle with major amount of calcium (Ca) from calcite, with aluminum (Al), silicon (Si), and magnesium (Mg) and potassium (K), evidence of a coating of clay, possibly illite.

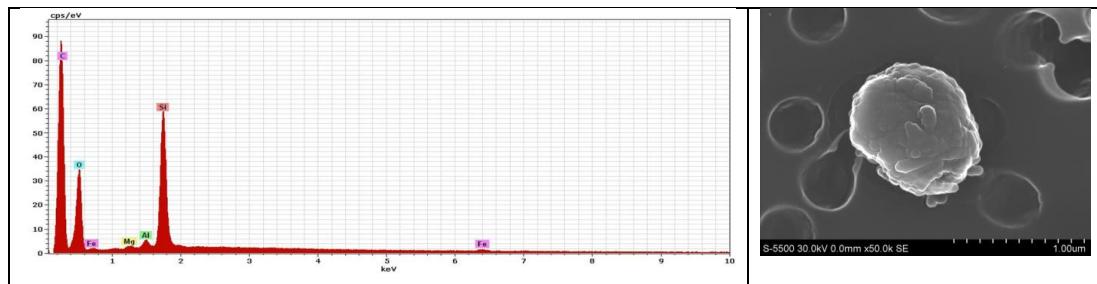



Figure 6: SEM based EDS spectrum and SEI of a particle from re-suspended sample S2016 from Camp Leatherneck, Helmand Province, Afghanistan. Well rounded grain of quartz (containing Si) with coating of finger shaped clay mineral containing aluminum (Al), magnesium (Mg) and iron (Fe), possibly palygorskite.

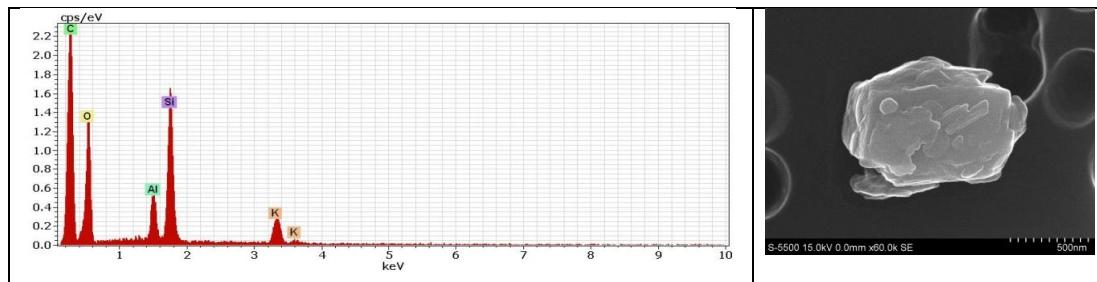



Figure 7: SEM based EDS spectrum and SEI of a particle from re-suspended sample S2017 from Kuwait. Orthoclase identified from its silicon (Si), aluminum (Al) and potassium (K) content as well as crystal morphology. Finger shaped particles on the feldspar surface may be palygorskite.

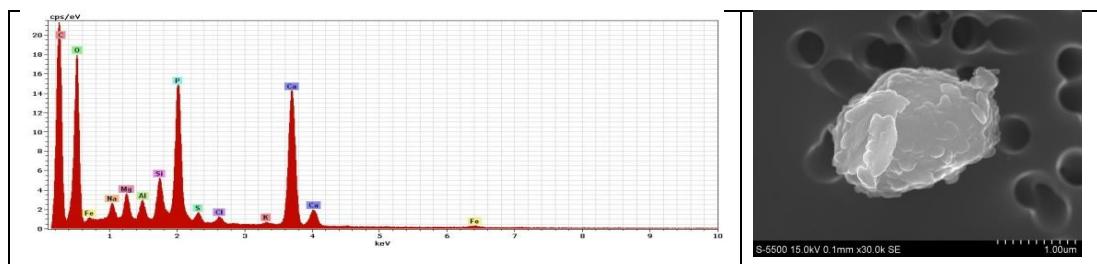



Figure 8: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1013 from Cape Verde Islands. Particle of apatite ($Ca_5(PO_4)_3(OH)$), identified from major components of phosphorous (P) and calcium (Ca). Small amounts of silicon (Si), aluminum (Al) and magnesium (Mg) can be attributed to a coating of clay. Small amounts of sulfur (S), sodium (Na) and chlorine (Cl) can be attributed to evaporites such as gypsum and halite attached to the particle.

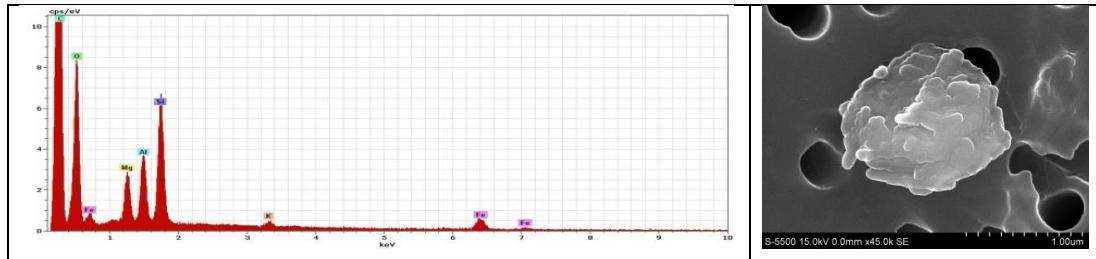



Figure 9: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1025 from Iriki, Morocco. From the relative intensities of the silicon (Si), aluminum (Al), magnesium, potassium (K), and iron (Fe) spectral peaks as well as the morphology shown by the SEI, it is concluded the particle to be a cluster illite (platy) and palygorskite (finger shaped).

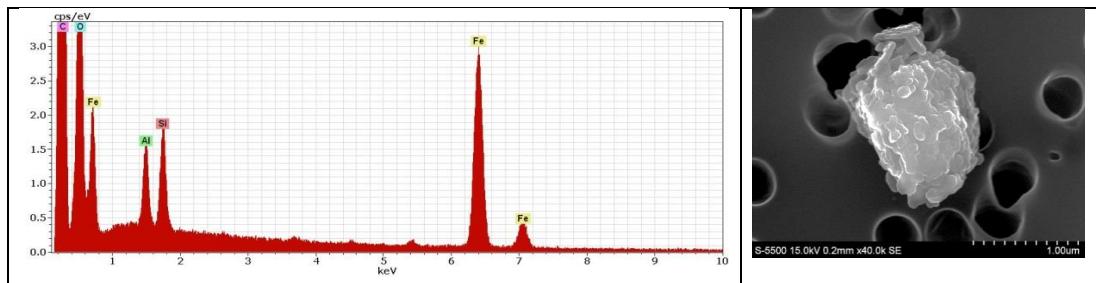



Figure 10: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1009 from Bamako, Mali. Composite grain composed of silicon (Si) and aluminum (Al) components of a clay mineral, possibly kaolinite with lesser amounts of palygorskite (finger shaped) and illite, together with iron (Fe) from hematite or goethite.

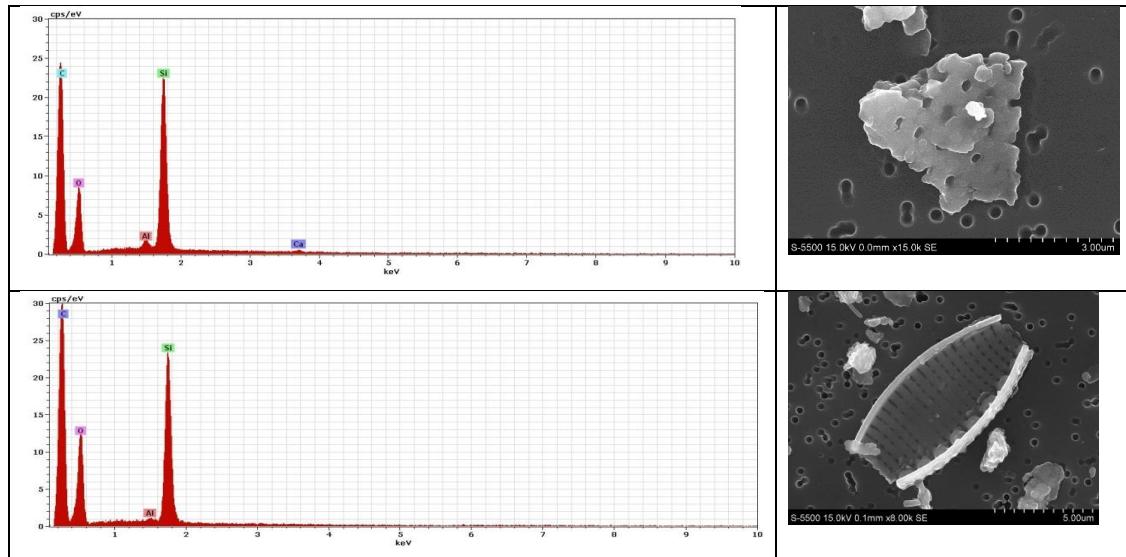



Figure 11: SEM based EDS spectra and SEI's of particles from re-suspended samples S1049 (above) and S1051 (below) from the Bodélé region of Chad. Two examples of diatom fragments, the particles contain largely silicon (Si), evidence of amorphous silica (SiO₂) or quartz, with small amounts of aluminum (Al) from a clay mineral coating, possible kaolinite and trace of calcium (Ca), possibly of calcite.

Figure 12: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1014 from northwestern China. Composite particle of calcite as seen from the high calcium (Ca) content together with a clay mineral (coating?), possibly illite as is evident from the silicon (Si), aluminum (Al), magnesium (Mg) and potassium (K) contents.

Figure 13: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1057 from Lake Eyre, Australia. Round quartz particle as evident from its silicon (Si) content, with small amounts of aluminum (Al), magnesium (Mg) and iron (Fe) from clay (coating), possibly illite or montmorillonite.

Figure 14: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1062 from Kostolac, Serbia. The silicon (Si) content points to a rounded quartz particle contained in the loess.

Figure 15: Size-class fractions grouped as clay (< 2 μm), silt (2 - 62.5 μm) and sand (62.5 – 600 μm). Samples from regions with the greatest potential to be sources of airborne dust lie in the "silt loam" and "silt" fields with less than about 10% clay, including samples
5 **from known dust regions in Chad , China, Afghanistan, Kuwait, northwesten Africa, southeastern Europe and parts of western U.S.A.**

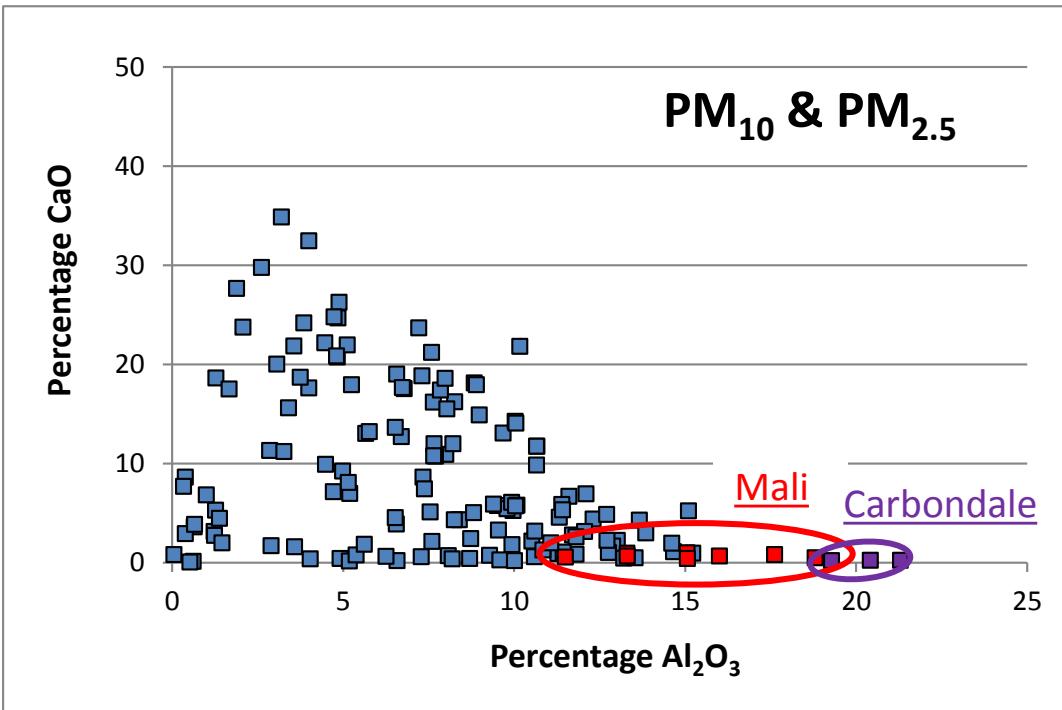



Figure 16: Scatter plot of CaO against Al₂O₃ showing an inverse relationship for some samples. This points in part to the substitution of
5 **calcite (dolomite) for clay minerals. The clay (kaolinite) bearing samples from Mali and Carbondale (circled) and several others lie along the X-axis, implying they contain no calcium bearing minerals such as calcite, dolomite, gypsum or calcium silicates.**

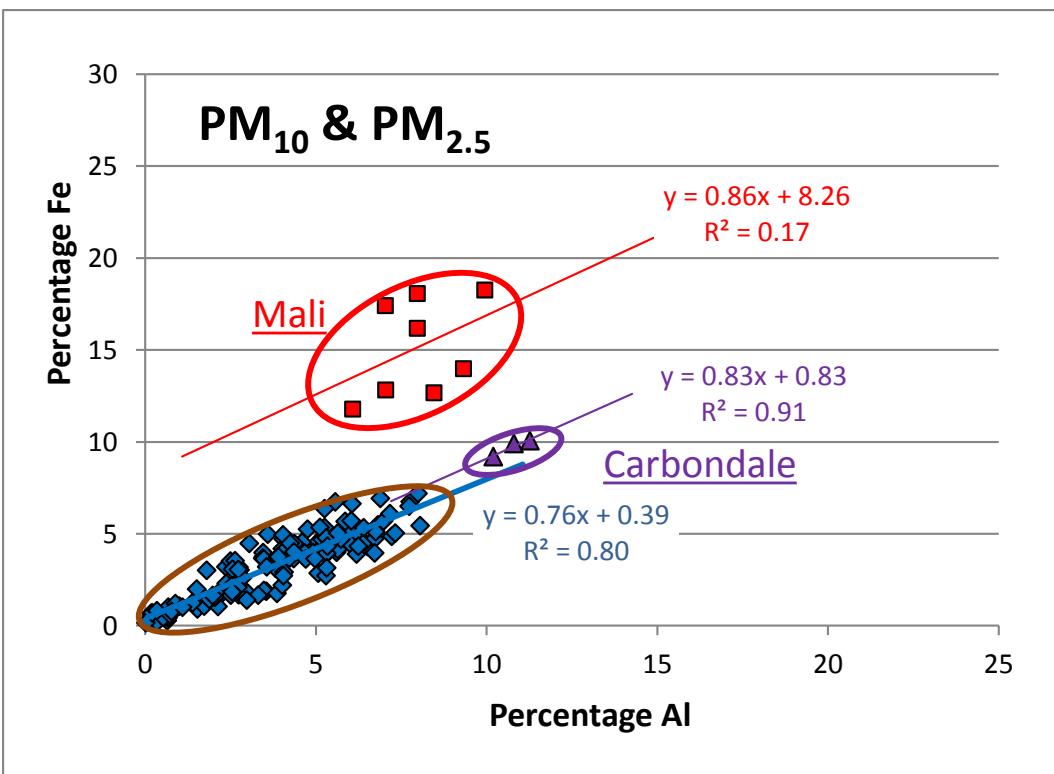



Figure 17: Plot showing the correlation between iron (Fe) and aluminum (Al) for all chemically analyzed samples. Three sample clusters can be identified with most samples fall in one field (brown oval) with an average Fe/Al slope of 0.76. Samples from Mali (red oval) and Carbondale (violet oval) have slightly higher slopes. Projected intercepts on the Fe axis point to those samples (from Mali) that contain
5 **part of their Fe in clay minerals and part in iron oxides such hematite or goethite. An intercept through the origin implies that all the iron is contained in the clay mineral structure.**

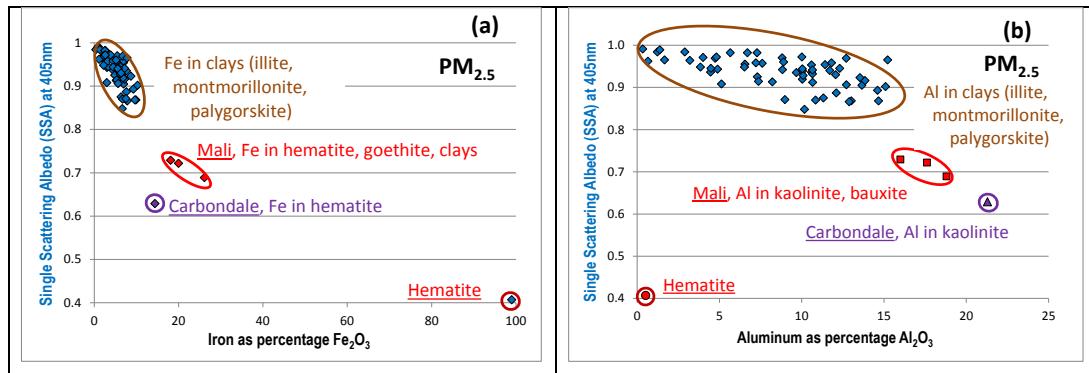



Figure 18: Single scattering albedo (SSA) at 405 nm in relation to (a) Fe_2O_3 and (b) Al_2O_3 content, illustrating the relationship to dust mineralogy, for re-suspended $PM_{2.5}$ dust. Most investigated samples contain iron bearing clays (illite, montmorillonite, palygorskite) with SSA greater than 0.85, while those with all or part of the iron as iron oxides (hematite, goethite) have SSA below 0.75.

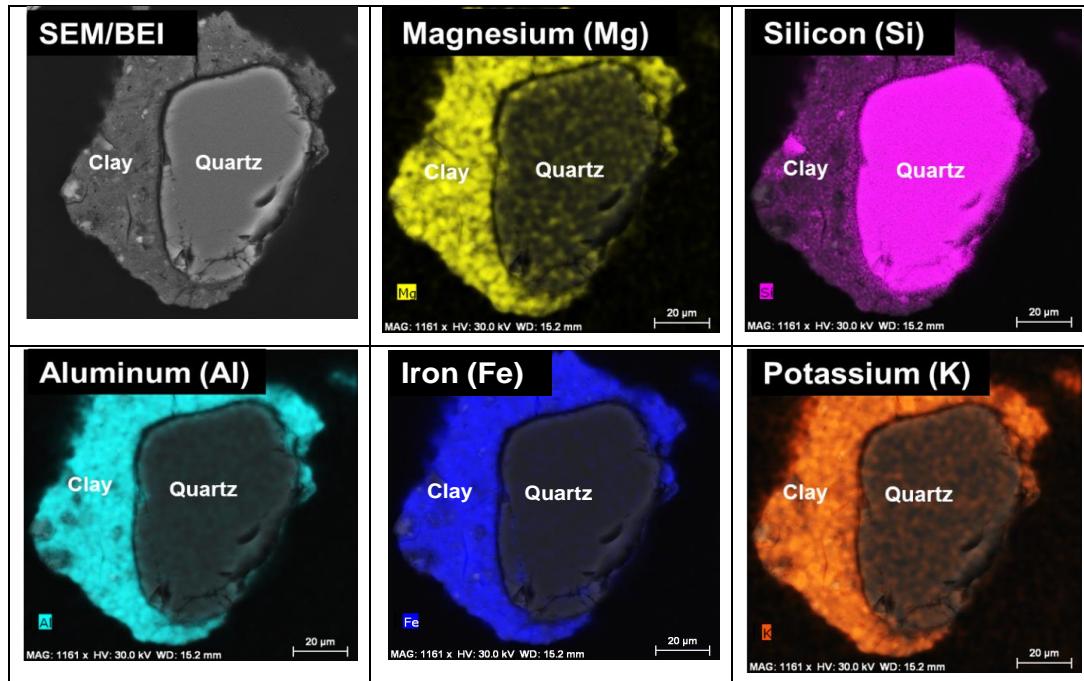
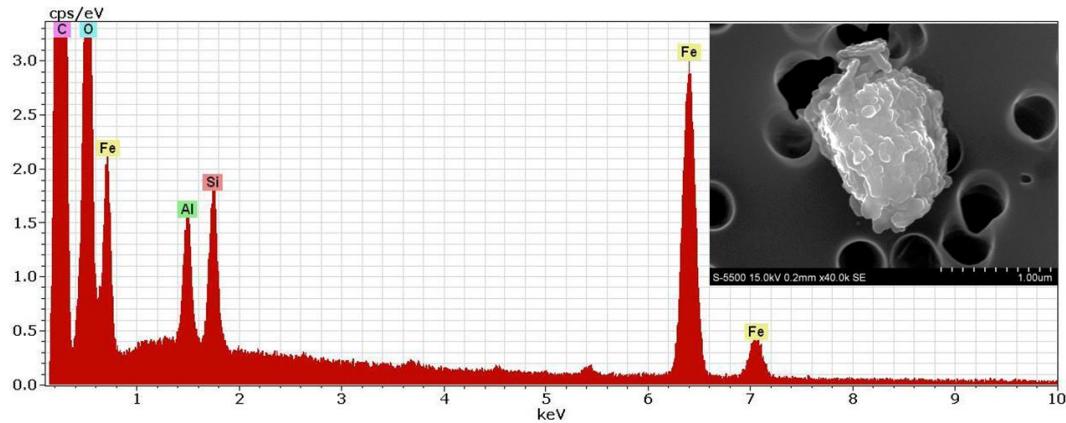



Figure 19: Sample S1056 from Lake Eyre, South Australia. SEM generated backscattered electron image (BEI) and chemical maps of a quartz grain with a prominent coating of clay. The uniform distribution of magnesium (Mg), silicon (Si), aluminum (Al), iron (Fe) and potassium (K) in the coating is evidence that all these elements are contained in in the structure of the clay mineral illite or nano-scale interlayers or particles of iron oxides.

5 **Figure 20: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1009 from Mali. The composite particle contains iron (Fe) as the mineral goethite (FeO.OH) and/or hematite (Fe$_2$O$_3$) together with silicon (Si) and aluminum (Al) in the clay mineral kaolinite.**

8 Table

Table 1: Linear correlation coefficients between pairs of chemical species, with pairs > 0.9 shown in green, 0.6 to 0.9 in yellow, 0.4 to 0.59 in blue, and -0.4 to -0.59 in red.

	Si	Ti	Al	Fe	Mn	Ca	Mg	K	Na$^+$	CL$^-$	SO$_4^{2-}$
Si	1										
Ti	0.37	1									
Al	0.47	0.75	1								
Fe	0.09	0.63	0.79	1							
Mn	0.32	0.60	0.41	0.21	1						
Ca	-0.48	-0.30	-0.40	-0.35	-0.06	1					
Mg	-0.18	-0.30	-0.41	-0.32	-0.01	0.424	1				
K	0.44	0.39	0.27	-0.04	0.58	-0.16	-0.05	1			
Na$^+$	-0.34	-0.36	-0.38	-0.29	-0.28	-0.14	0.06	-0.11	1		
CL$^-$	-0.15	-0.23	-0.22	-0.23	-0.11	-0.04	0.18	0.04	0.48	1	
SO$_4^{2-}$	-0.38	-0.32	-0.33	-0.26	-0.27	-0.04	0.06	-0.21	0.91	0.41	1