Authors' Response to Co-Editor Decision and Comments

Co-Editor Decision: Publish subject to technical corrections (08 Aug 2016) by Dr. Daniel Knopf

Comments to the Author:

5 Dear Authors,

I am happy to accept your manuscript for publication in Atmospheric Chemistry and Physics.

I just have minor technical corrections given below.

With kindest regards,

Daniel Knopf

Authors' response:

Dear Dr. Knopf

Thank you very much for your reviews, and for accepting our manuscript "Technical Note: Mineralogical, chemical,

morphological, and optical interrelationships of mineral dust re-suspensions" for publication in ACP.

15 Regards

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Technical corrections:

Inconsistent nomenclature for goethite:

30 p. 4, l. 22: p. 12, l. 37; p. 15, l. 8; Fig. 20: "goethite (FeO.OH)"

p. 13, l. 36: "goethite (FeO(OH))"

p. 15, l. 12: "goethite (FeO.OH))"

I thought it should be "goethite (FeO(OH))"?

Authors' response:

35 For the sake of consistency we replaced all by "goethite (FeO(OH))", as suggested.

p. 4, l. 2: Typo after "(Figure 2)".

Authors' response:

40 Typo was corrected by deleting the redundant "n"

All changes are marked in red below

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

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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 interdependency of single scattering albedos (SSA) at three different wavelengths, and mineralogical content of the entrained dust samples. To explain the elevated concentrations of iron (Fe) and Fe/Al ratios in the soil re-suspensions, we propose that dust particles are to a large extent composed of nano-size particles of micas, clays, metal oxides, and ions of potassium (K^+), calcium (Ca^{2+}) and sodium (Na^+) evenly dispersed as a colloid, or adsorbed in amorphous clay-like material. Also shown are differences in SSA of the kaolinite/hematite/goethite samples from Mali and those from colloidal soils elsewhere. 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 entrainment chamber; Dust mineralogy; Particle size distributions; Single Scattering Albedo (SSA); SEM based particle analysis; Dust Fe/Al ratios, Colloids in amorphous clays

1 Introduction

Globally, airborne mineral dust 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.

In the past only particles less than ≈ 10 μm in diameter were considered to survive long-range transport in the atmosphere (Maring et al., 2003;Zender et al., 2003). However, multiple studies have shown larger mineral dust particles to have been transported over distances of hundreds to thousands of kilometers (Alastuey et al., 2005;Betzer et al., 1988;Lawrence and Neff, 2009;McTainsh et al., 1997;Neff et al., 2013;Menéndez et al., 2014). For example, 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. Samples of airborne dust collected off the west African coast (Stuut et al., 2005) had mean size ranges from 8-42 μm, with particles of up to 200 μm being identified. A study on Asian dust transported to Korea over a distance of 2000 km showed a substantial proportion of giant mineral particles, up to 60 μm in diameter (Jeong et al., 2014) . These particles were found to be comprised of clay aggregates as well as clay coated quartz, feldspar and mica grains. Complex aggregates (iberulites) of up to 100 μm in diameter, were postulated to have formed in the atmosphere during long range transport of mineral dust from Saharan and Sahel to southern Spain (Díaz-Hernández, 2008). Furthermore, these aggregates were found to be composed largely of quartz, feldspars, and carbonates, in a matrix of various clay minerals (Cuadros et al., 2015).

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, visibility impairment and their potential health effects, 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;Fubini and Fenoglio, 2007;Ghio et al., 2014). The prime objective of this publication is to provide speciated information on mineral dust aerosol optical properties, mineralogy, chemistry, as well as particle morphology and size distribution, all 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., 2012a;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 al., 2007;Kandler et al., 2009;Molinaroli, 1996). This paper expands on these previous contributions by presenting properties of a global set of re-suspended dust samples applying similar sample collection, 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, 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] (Figure 1), as well as one sample of hematite (Fe_2O_3). The selected samples, although of variable mineral and chemical compositions, are representative of many major dust regions. 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 investigated 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 artefacts. Where necessary, they were air-dried in the laboratory, before being labelled, catalogued, and stored in capped plastic bottles. Particle size distributions were performed on the < 600 μm sieved sample sets. Sub-sets of these samples were sieved to < 38 μm for mineral analysis by X-ray powder diffraction (XRD), and for re-suspension and subsequent chemical and SEM-based analysis. Further sub-sets were sieved to > 75, < 125 μm for mineral mounts and investigation by optical microscopy. All samples were dry sieved, so as to retain their mineralogical and physical integrity as close as possible to the surface soils from which they were sampled. Wet sieving of samples was avoided since that would dissolve evaporite minerals such as chlorides and sulfates contained in several samples, and also disaggregate clay clusters into smaller fragments, thereby creating a bias towards the finer particle sizes (Choate et al., 2006). The re-suspension of dry sieved samples in a chamber would approximate the mechanism for generating dust, such as particle bombardment and fragmentation of dust aggregates (Kok, 2011;Shao, 2001).

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 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 properties such as texture, color, pleochroism, birefringence, refractive index, and twinning, were used to identify silicate minerals. Minerals readily identified in the grain mounts 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)

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 system with sample stacker and spinner was used to collect the XRD spectra while Bruker Topas® software and the relative intensity ratio (RIR) procedure was applied for semi-quantitative analyses of the XRD spectra of the dust samples (Rietveld, 1969;Chung, 1974;Esteve et al., 1997;Sturges et al., 1989;Caquineau et al., 1997) (Supplement S2.2).

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 dry sieved to < 600 μm before being introduced to the laser analyzer (Micromeritics Saturn DigiSizer 5200®) in an aqueous medium of 0.005% surfactant (Na metaphosphate). The particles 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 (Figure 2) was configured with an array of instrumentation, including (i) three-wavelength photoacoustic 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).

5 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, particulate mass concentration, and particle size measurements (Moosmüller et al., 2012a). Dust measurements were focused on the size fraction associated with long-range transport, typically less than 9 µm in physical 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)

10 and $PM_{2.5}$ (≤ 2.5 µm aerodynamic diameter) were separated. The procedure was to place about one gram of the < 38 µm sieved 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 there the dust aerosol was drawn through size selective inlets, either PM_{10} or $PM_{2.5}$, and directed to the optical, gravimetric, and particle size monitors, as well as to the four PM filter samplers.

The most important optical properties (Bergstrom et al., 2007;Chýlek and Wong, 1995;Hassan et al., 2015) for calculating aerosol

15 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

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

The (Photoacoustic Instrument [PA]); (Arnott et al., 1999;Arnott et al., 2000;Lewis et al., 2008) with an integrating reciprocal

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

25 The 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 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

30 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

Teflon® membrane and quartz fiber filters were conditioned and weighed prior to and after sampling, for subsequent mass and chemical composition determinations of the dust samples.

35 In the course of this study, about 130 Teflon® filters were analyzed by Energy Dispersive X-ray Fluorescence Spectrometry (EDXRF) (Watson et al., 1999;US EPA, 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 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^-),

5   phosphate (PO_4^{3-}), and the ammonium (NH_4^+) cation. Further 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 abundances 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

10   between the single scattering albedos (SSA) and the major chemical components are graphically presented.

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) using automated particle analysis software, and the second, high resolution manual SEM examination to

15   document particle morphology.

CCSEM was performed using a combination of Backscattered Electron (BSE) and Secondary Electron (SE) imagery along with Energy Dispersive Spectroscopy (EDS). A large number (often more than 1,000) of individual particles were analyzed for chemical composition, particle size and shape, by this method. 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

20   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 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.

25   Individual particle morphology including aspect rations were measured by CCSEM. Although labour intensive, manual SEM analysis allows for collection of superior quality, high resolution SE images. 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

30   **3.1 Sample descriptions**

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 shown on each sample

35   sheet in Supplement S3.2 are beta attenuation gauge mass measurements for PM_{10} and $PM_{2.5}$ collected in parallel with the absorption and scattering coefficients, particle size distributions and statistics, mass measurements for PM_{10} and $PM_{2.5}$ on Teflon filters and $PM_{2.5}/PM_{10}$ mass ratios, collected for each sample. Statistics on the SEM measured aspect ratios are tabulated on the

individual 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 expressed as oxides are shown in Supplement S4.4, the relationships between chemical compositions and SSA in Supplement S4.5, and ratios of selected major components tabulated in Supplement S4.6. The scanning

5   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 individual particles are tabulated in Supplement S5.4 and shown on Supplement S3.2. Supplement S5.5 shows the energy dispersive (EDS) spectra and associated secondary electron images (SEI's) of selected individual particles from

10   each re-suspended dust sample.

Descriptions of samples from each geographic region are summarized below.

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

15   consumption by the city of Los Angeles (Wilkerson et al., 2007). The coarser sand and silt fractions contain plagioclase, calcite, quartz, biotite mica, and hornblende, with halite, kaolinite, and trona in the finer fractions. These minerals 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 resulted in ongoing mitigation steps to contain alkaline dust being blown off the playa. (www.ladwp.com/OwensLake).

20   **3.1.2 Southwestern U.S.A. – Black Rock, NV (S1045)**

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 transported 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, for a week each in August each year. As a

25   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 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

30   diatomaceous lakebed deposit (Smedman, 1969), containing diatoms (Figure 3), plagioclase, quartz, and lesser amounts of clay (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)

35   Carbondale Red Clay (CRC) is a commercially available fireclay, considered to be the equivalent of Martian dust (White et al., 1997). The material is composed of very finely ground 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 contain 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 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, 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)

Fort Carson is located in central Colorado at the foot of the Rocky Mountains and to the south of Colorado Springs. The area is underlain largely by fluvial and alluvial clays, silts and sands. The surface soil sample was collected from an unpaved road on the military base. It contains major amounts of quartz and plagioclase, with lesser amounts of biotite, potassium feldspars, calcite, hornblende, muscovite, dolomite, gypsum, and clays (Engelbrecht et al., 2012).

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

The two samples from Utah are similar to each other, both collected along built-up dirt roads on the military base. The road gravel material had been excavated from a local quarry. Both samples 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 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, 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 last mentioned 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 largely quartz (Figure 6) and calcite with small amounts of clays, gypsum, and silicates.

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, and anhydrite. 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., 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 flats 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 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 evaporite 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 Tallil. 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. Samples from all five sites also contain a significant amount of feldspar, both 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 to have been transported from the dry Mesopotamian floodplain in southern Iraq and other parts of the Arabian peninsula (Engelbrecht et al., 2009b;Engelbrecht et al., 2008).

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., 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 safely assumed that samples of soils collected on these islands contain Saharan dust in variable proportions. Two samples from Fuerteventura are from dust traps in 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 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 some 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.

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). 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 samples are 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 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. (2012a) 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 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 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)

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.

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).

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 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, 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 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 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

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 approximately 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 "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 (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 (Moosmüller et al., 2012b). For this project we measured 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 mineral grains, intergrowths, coatings on other particles, or composite mineral clusters. Their chemical compositions can in many instances be related to their mineral constituents, as verified by optical microscopy, XRD, and SEM-based individual particle analysis.

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_2SO_4) identified by XRD in two samples (S1023, S1024) from Etosha Pan, a dry lakebed 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 occurring together or attached to 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., 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_2O_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 ($NaAlSi_3O_8$ - $CaAl_2Si_2O_8$), gypsum ($CaSO_4.2H_2O$), and other evaporite deposits.

There is a reasonably strong linear correlation of 0.79 between iron (Fe) and aluminum (Al) for the complete set of 165 samples (Table 1), with most (122) samples falling in one oblong shaped cluster with an average slope of 0.76 (Figure 17). This subset of samples has an average Fe_2O_3 content of 4.76% for PM_{10} and 5.22% for $PM_{2.5}$, with respective Fe/Al ratios of 0.87 and 1.01 (Table 2). The Fe/Al ratios for these two subsets of samples vary from 0.47 - 3.38 for PM_{10}, and 0.41 - 3.78 for $PM_{2.5}$. 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), similar to ambient $PM_{2.5}$ samples (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 ratios of 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.

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. Dust samples from Niger, Cape Verde, Tunisia, and China, suspended on polycarbonate filters, and analyzed by XRF give concentrations of 6.2 − 8.7 % Fe_2O_3, and Fe/Al ratios of 0.43 − 6.0 (Lafon et al., 2006). We suggest that differences in aluminum (Al) and iron (Fe) concentrations, and subsequent range in Fe/Al ratios found by investigators are not only due to chemical differences amongst the samples, but can also in part be ascribed to different sampling and analytical procedures.

The high correlation between iron (Fe) and aluminum (Al) is attributed to the co-existence of oxide minerals (hematite, goethite, magnetite) with various clay minerals, chlorite, and other silicates, in varying proportions, depending on factors of provenance, chemical weathering, and pedogenesis. Comparisons of chemical results of re-suspended soils from this study, and those published on biotites (Deer et al., 1962) and clays (Weaver and Pollard, 1973) are shown in Table 2. Clay minerals identified by XRD and SEM include illite, palygorskite and kaolinite, with montmorillonite in few samples. The average aluminum (Al) concentrations of the re-suspended soils (3.58% for PM_{10}, 4.64% for $PM_{2.5}$) (Table 2(a), 2(b)) are substantially lower than those of chlorite (22.06%) (Table 2(d)), illite (13.94%) (Table 2(e), montmorillonite (11.6%) (Table 2(f) or palygorskite (6.73%) (Table 2(g)). The iron (Fe) contents of the re-suspended soils (ave. 3.33% for PM_{10}, 3.65% for $PM_{2.5}$) are less or similar to those of chlorite (2.21%), illite (3.47%), montmorillonite (2.79%), or palygorskite (2.26%). The resultant Fe/Al ratios for the re-suspensions are proportionally greater (ave. 0.87 for PM_{10}, 0.1.01 for $PM_{2.5}$) than those of chlorite (0.07), illite (0.25), montmorillonite (0.24) or palygorskite (0.27) (Table 2 (d)-(g)). The greater Fe/Al ratios for the re-suspended soil samples point to the presence of Fe-bearing minerals in the particles, other than clays. It is evident that although some iron (Fe) is contained in the crystal structures of the chlorite, illite, montmorillonite, palygorskite and other clay minerals, the remaining iron (Fe) is contained in hematite (Fe_2O_3), goethite (FeO(OH)) or magnetite (Fe_3O_4), as well as iron bearing silicates such as biotite (Table 2(b), amphibole, and pyroxene, all minerals identified in the sieved soil samples by optical microscopy and XRD. Also, the grouping of the majority (122) of the samples (Figure 17) and the high correlation coefficient of 0.75 between iron (Fe) and aluminum (Al) (Table 1), point to a strong mineralogical interrelationship amongst the Fe-bearing clays, Fe-oxides (goethite, hematite, magnetite), and other silicates such as

micas (biotite, muscovite) and chlorite. Recent studies (Reynolds et al., 2014a;Reynolds et al., 2014b;Kars et al., 2015), applying a combination of Mössbauer spectroscopy, magnetic measurements, reflectance spectroscopy and scanning electron microscopy (SEM), provide evidence of nanometer size (< 20 nm) particles of hematite, goethite and magnetite, associated with clay particles. Similarly, from magnetic measurements on the Callovian-Oxfordian claystones in the Paris Basin, Kars et al. (2015) suggest

5   nanoparticles of goethite to be dispersed in the clay-like matrix. Fordham (1990), from their investigations by electron microscopy and XRD concluded that biotite underwent chemical weathering at particle edges and along cleavage plains, to form finely divided interstratified flakes of illite and iron oxides, in the clay-like matrix, which Borchard et al. (1971) had identified as colloidal coatings of amorphous clay-like particles of volcanic ash in soils.

To explain the elevated concentrations of iron (Fe), titanium (Ti) calcium (Ca), potassium (K) and Fe/Al ratios in the soil re-

10 suspensions, compared to those in crystalline clay minerals (illite, montmorillonite and palygorskite) (Table 2), we propose that particles of PM_{10} and $PM_{2.5}$ are to a large extent composed of nano-size particles of micas, clays, metal oxides, and ions of potassium (K^+), calcium (Ca^{2+}) and sodium (Na^+) evenly dispersed as a colloid, or adsorbed in amorphous clay-like material. These composite particles are suggested to be weathered products of biotite and other ferro-magnesian particles are suggested to have formed by chemical weathering and pedogenesis, forming particle coatings, individual particles, or interstitial material with quartz,

15 feldspars and other silicates.

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) 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

20 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 the samples to an average geometric mean diameter of 1.19 μm, with a standard deviation 0.11 μm (Supplement S3.2). The aspect ratios of particles in the samples varied in the geometric mean range of 1.37 to 1.70 (Supplement S5.4). The 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).

25 As was previously shown (Moosmüller et al., 2012a), there is a direct relationship between the iron (expressed as 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 microscopy (Supplement S2.1), XRD (Supplement S2.2), and SEM based individual particle analysis (Supplements S5.1, S5.2), the sample chemical 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 from the amorphous iron (Fe) bearing clays (illite,

30 montmorillonite, palygorskite), and the other from Mali where the iron (Fe) is predominantly in the hematite or goethite with kaolinite (not containing 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 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)

35 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 nm (also 532 nm) is stronger for hematite and goethite, than for iron

(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 samples, Carbondale Red Clay has the lowest SSA at 405 nm (SSA = 0.629) and at 532 nm

5   (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 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

10 0.999 (781 nm).

4 Conclusions

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 the globe.

15 However, it is recognized that dusts may be modified as they mix with other aerosols, and age under changing meteorological conditions and over time, in some cases forming aggregates of larger particles. In our study, SEM based secondary electron images of the re-suspended dust samples are evidence of such mineral clusters, as well as of coatings on quartz, feldspar and other mineral grains, similar to that reported on in ambient aerosols (Cuadros et al., 2015;Díaz-Hernández, 2008;Engelbrecht et al., 2009a;Jeong et al., 2014). The suite of surface soil samples investigated here are representative of many recognized dust source regions of the

20 globe. At the time of this study, we did not have samples from all dust regions of North Africa, the Middle East, Asia, Australia or South America.

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 related, and other studies.

25 The individual analytical procedures each provided an important component to an understanding of mineral dust properties. Laser based particle size analysis of the < 600 μm sieved sample provided information on the silt content of the soils, and the likelihood of dust being suspended when soil surfaces are disturbed. Petrographic microscopy on the > 75, < 125 μm sieved soil fractions readily identified the minerals such as quartz, feldspars, micas and carbonates, which make up the coarse fractions of dust. XRD provided semi-quantitative information on the mineral content of the < 38μm sieved soil fractions. The chemical analyses (XRF,

30 ICP-MS, ICP-OE, IC) of the PM_{10} and $PM_{2.5}$ size fractions could be related to their mineralogy, including their clay (illite, palygorskite, montmorillonite, kaolinite) and iron oxide (hematite, goethite) relationships, as well as to characterizing dust sources. Qualitative information on the mineralogy of the dusts, obtained from optical microscopy, XRD, and SEM, allowed for their chemical compositions to be calculated to normative mineral compositions. Such normative mineral compositions are useful when comparing samples to each other. SEM, furthermore, provided information on the individual particle size, shape, and chemical

35 composition which in many cases could be linked to the sample mineralogies. Surface coatings of clays, oxides and amorphous phases, often mask the identity of the underlying mineral grains of quartz and feldspars, a feature which needs to be considered when interpreting SEM and optical measurements.

Two mineralogically distinct phase assemblages are distinguished. The first and largest group of samples are described as colloids in amorphous clay (allophane) (Kars et al., 2015) clusters, surface coatings on particles, and interstitial soils, characterized here by an average Fe/Al ratio of 0.94 (range 0.41 – 3.78). The iron (Fe) and aluminum (Al) contents as well as the Fe/Al ratios are evidence of interstitial layers of iron (goethite) and aluminum (gibbsite), oxihydroxides (Birkeland, 1999), and possibly the presence of nano

5   size colloidal particles of oxides (hematite). The abundance of potassium (K^+) points to this element being adsorbed on the amorphous clay layers, or nano-size flakes of residual primary mica (biotite) or illite, retained in the amorphous colloid. The second and 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 (Fe) as individual oxide (hydroxide) mineral grains of hematite (Fe_2O_3) and goethite (FeO(OH)) (Roquin et al., 1990).

10   The SSA measured on the $PM_{2.5}$ samples, as clearly seen at the blue (i.e. 405 nm) wavelength, varied not only with the iron (Fe) content of the samples, but also with the mineralogy, i.e., whether the iron (Fe) was contained in a clay mineral (illite, montmorillonite, palygorskite), or as nano-size colloidal particles of hematite (Fe_2O_3) and goethite (FeO(OH)) in the amorphous clay-like matrix.

Understanding the dust mineralogy, especially of the clay minerals and iron oxides, is of prime importance to our understanding

15   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), here illustrating that this relationship is also controlled by sample mineralogy and mineralogical interrelationships.

Data availability

Mineralogical, chemical, physical, and optical results are presented in tables and figures in the Supplement to this paper. Requests

20   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

25   Moosmüller was the co-principal investigator for the project, formulating the problem, advising on the experimental design, 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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35   Engelbrecht for samples from U.S.A. (8), Canary Islands (8), Serbia (3), and Botswana (4); Henry Sun for a sample from Chile

(1); Ross Edwards for samples from China (4); Michał Skiba for a sample from Morocco (1); Davis Zhu for samples from U.S.A. (2); Steven Kohl for a sample from U.S.A. (1).

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8 Tables

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

Table 2. Average concentrations and elemental ratios for (a) PM$_{10}$ and (b) PM$_{2.5}$ re-suspensions from this study, (c) biotite, (Deer et al., 1962, Table 13), chlorite (Weaver and Pollard, 1973, Table XLII), Illite (ditto, Table III), montmorillonite (ditto,Table XXV), and palygorskite (ditto, Table LII).

	Soil re-suspensions		Biotite	Chlorite	Illite	Montmorillonite	Palygorskite
	(a)	(b)	(c)	(d)	(e)	(f)	(g)
N Obs.	61	61	14	7	24	101	15
	PM$_{10}$	PM$_{2.5}$					
	Ave. (range) mass %	Ave. (range) mass %	Ave. mass %	Ave. mass %	Ave. mass %	Ave. mass %	Ave. mass %
SiO$_2$	24.87 (3.70-56.75)	30.36 (6.07-56.12)	35.93	36.82	49.78	59.49	54.78
TiO$_2$	0.51 (0.03-1.71)	0.53 (0.03-1.74)	2.61	0.74	0.42	0.25	0.10
Al$_2$O$_3$	6.76 (0.38-12.77)	8.77 (0.33-15.23)	19.06	41.70	26.35	21.93	12.73
Fe$_2$O$_3$	4.76 (0.27-9.61)	5.22 (0.38-10.28)	2.52	2.74	4.30	3.77	2.73
FeO			16.40	0.38	0.61	0.20	0.45
MgO	1.81 (0.00-9.53)	2.80 (0.00-14.38)	9.04	4.39	2.75	3.55	0.41
CaO	9.98 (0.25-29.74)	9.53 (0.19-34.84)	0.27	0.88	0.32	1.18	1.00
Na$_2$O	1.87 (0.01-38.51)	1.83 (0.00-29.86)	0.62	0.17	0.25	0.82	8.68
K$_2$O	1.80 (0.22-3.33)	2.03 (0.32-4.02)	8.68	1.20	7.02	0.34	0.43
Si	11.62 (1.73-26.50)	14.18 (2.83-26.21)	16.78	17.19	23.25	27.78	25.58
Ti	0.30 (0.02-1.03)	0.32 (0.02-1.04)	1.57	0.44	0.25	0.15	0.06
Al	3.58 (0.20-6.75)	4.64 (0.18-8.06)	10.08	22.06	13.94	11.60	6.73
Fe^{III}	3.33 (0.19-6.72)	3.65 (0.27-7.18)	1.76	1.92	3.01	2.64	1.91
Fe^{II}	-	-	12.74	0.29	0.47	0.15	0.35
Mg	1.09 (0.00-5.75)	1.69 (0.00-8.67)	5.45	2.65	1.66	2.14	0.25
Ca	7.13 (0.18-21.26)	6.81 (0.13-24.91)	0.17	0.63	0.23	0.84	0.72
Na	1.39 (0.01-28.58)	1.36 (0.00-22.15)	0.46	0.13	0.18	0.61	6.44
K	1.49 (0.18-2.76)	1.68 (0.27-3.34)	7.20	0.99	5.83	0.28	0.35
Mass ratios							
Si/Al	5.51 (2.09-70.13)	5.49 (1.86-80.76)	1.67	0.81	1.67	2.39	4.04
Ti/Al	0.09 (0.05-0.29)	0.07 (0.03-0.29)	0.16	0.01	0.02	0.01	0.00
Fe/Al	1.01 (0.47-3.38)	0.87 (0.41-3.78)	1.44	0.07	0.25	0.24	0.27
Mg/Al	1.37 (0.0-28.38)	1.71 (0.0-48.98)	0.54	0.13	0.12	0.18	0.01
Na/Al	2.63 (0.0-83.46)	1.52 (0.00-34.1)	0.09	0.00	0.01	0.05	1.05
K/Al	0.58 (0.1-3.56)	0.51 (0.09-3.08)	0.71	0.03	0.42	0.02	0.01

7 Figures

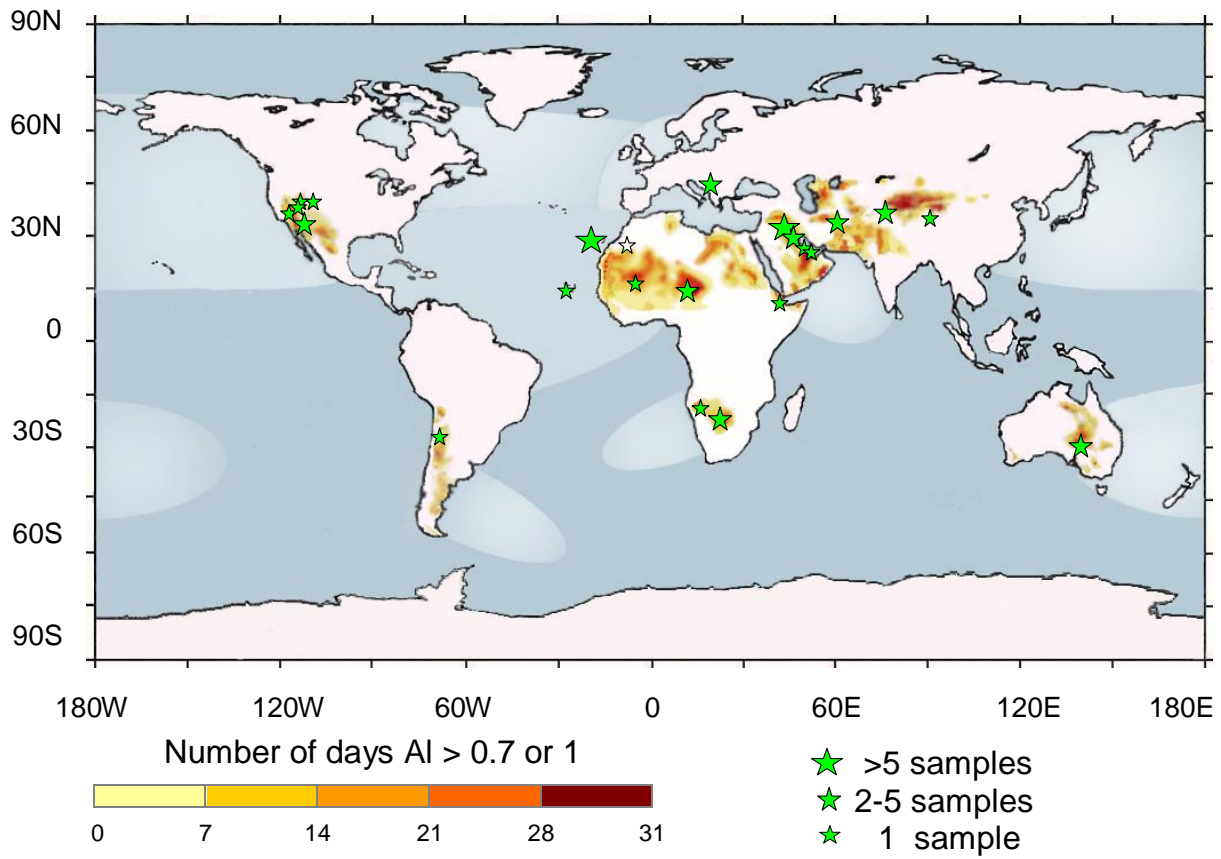



Figure 1: Surface soil sampling regions shown as green stars on a global map of dust sources as identified from Total Ozone Mapping Spectrometer (TOMS) data (Prospero et al., 2002). The size of the marker is approximately proportional to the number of samples collected in the region.

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.

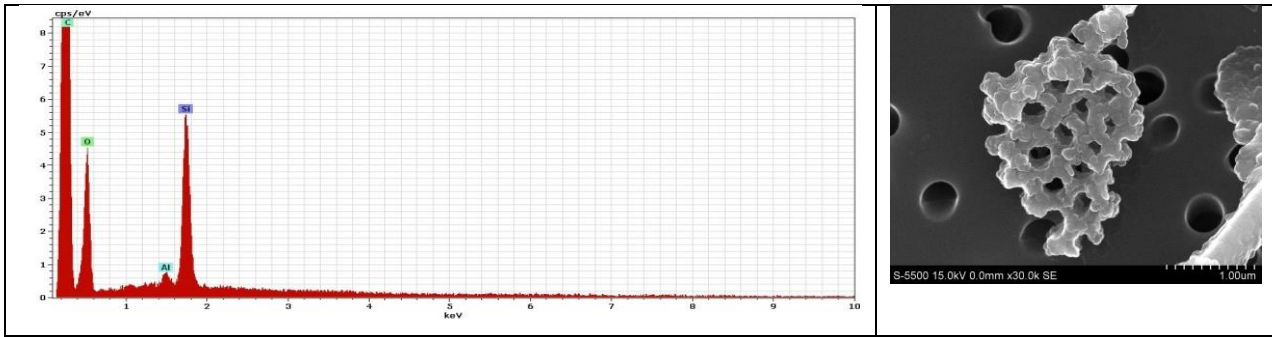



Figure 3: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1052 from Reno, Nevada: A fragment of a diatom containing a major component of silicon (Si), with trace amounts of aluminum (Al) pointing to a silica particle (amorphous?) with a minor amount (coating?) of clay, possibly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

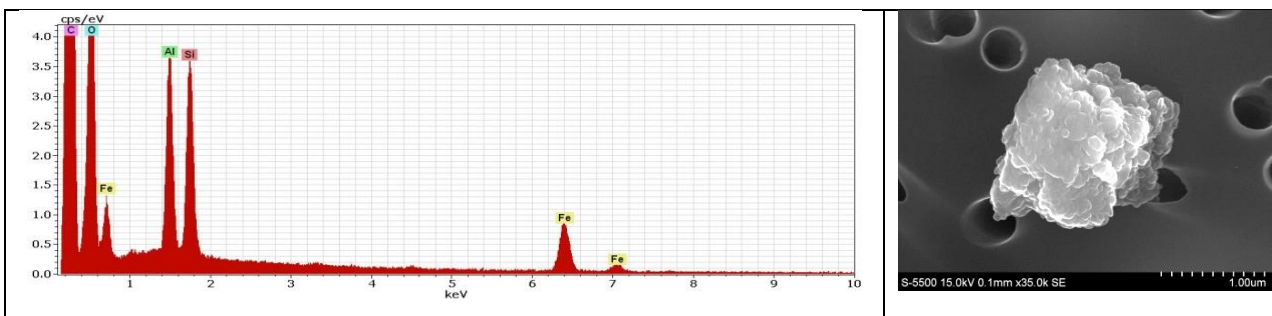



Figure 4: SEM based EDS spectrum and SEI of a particle from re-suspended Sample S1065 (red clay) from Carbondale, California. Composite particle containing major components of aluminum(Al) and silicon (Si) possibly the clay mineral kaolinite, as well as iron (Fe) in hematite or goethite.

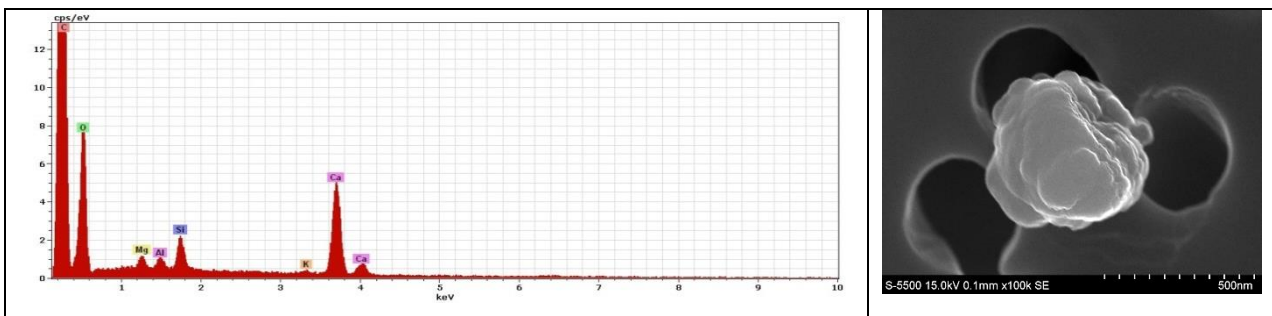



Figure 5: SEM based EDS spectrum and SEI of a particle from re-suspended sample S3017, from Dugway, Utah. Particle with major amount of calcium (Ca) from calcite, with aluminum (Al), silicon (Si), and magnesium (Mg) and potassium (K), evidence of a coating of clay, possibly illite.

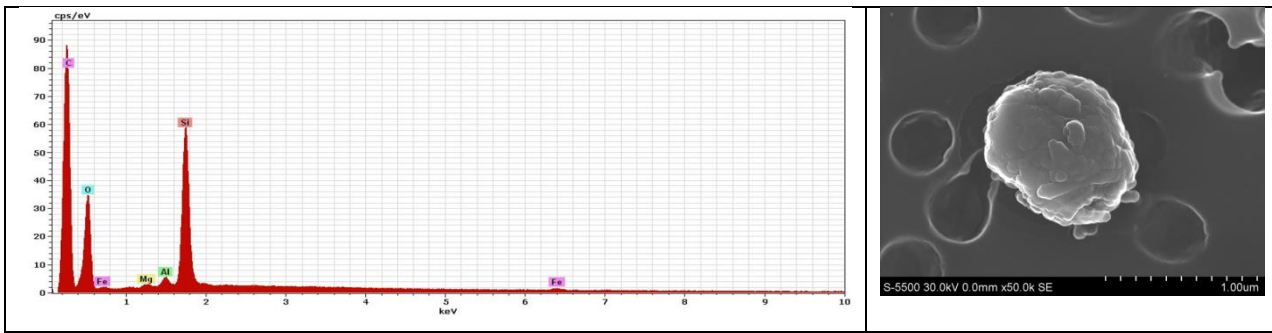



Figure 6: SEM based EDS spectrum and SEI of a particle from re-suspended sample S2016 from Camp Leatherneck, Helmand Province, Afghanistan. Well rounded grain of quartz (containing Si) with coating of finger shaped clay mineral containing aluminum (Al), magnesium (Mg) and iron (Fe), possibly palygorskite.

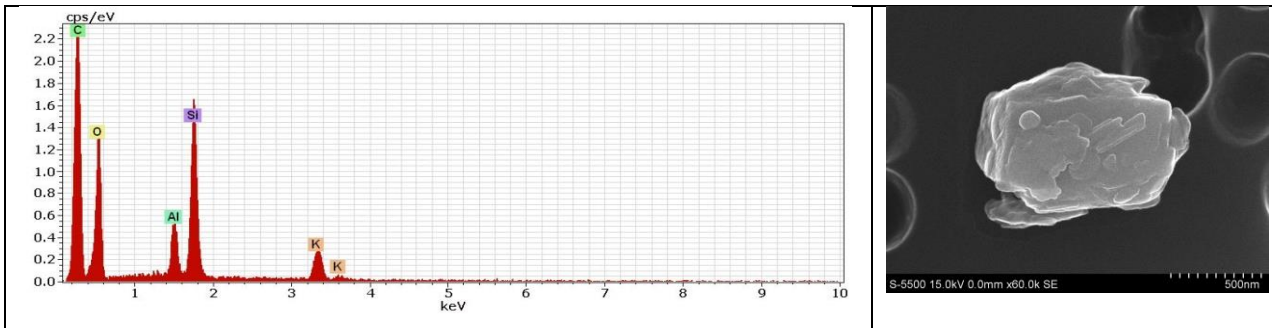



Figure 7: SEM based EDS spectrum and SEI of a particle from re-suspended sample S2017 from Kuwait. Orthoclase identified from its silicon (Si), aluminum (Al) and potassium (K) content as well as crystal morphology. Finger shaped particles on the feldspar surface may be palygorskite.

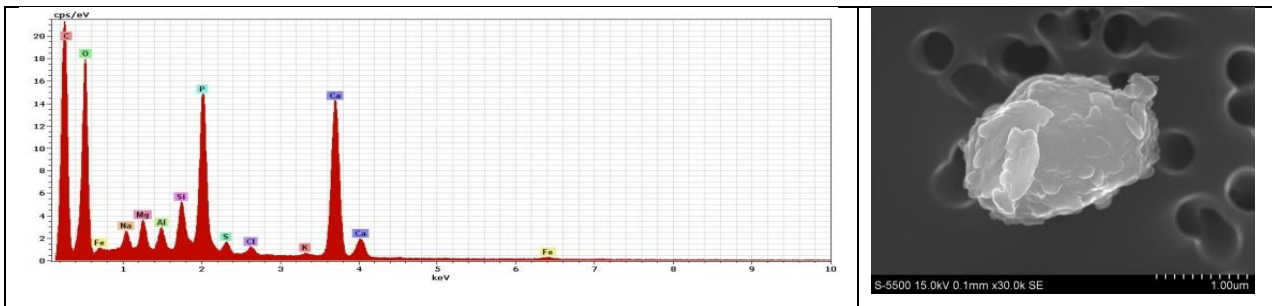



Figure 8: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1013 from Cape Verde Islands. Particle of apatite (Ca₅(PO₄)₃(OH)), identified from major components of phosphorous (P) and calcium (Ca). Small amounts of silicon (Si), aluminum (Al) and magnesium (Mg) can be attributed to 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.

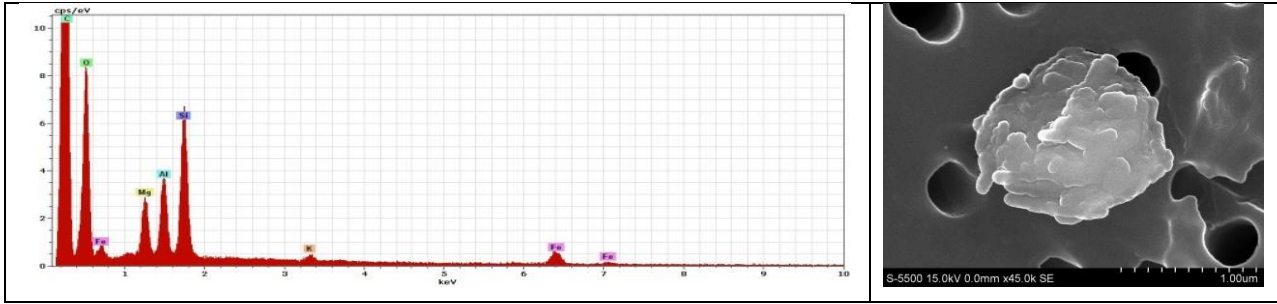



Figure 9: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1025 from Iriki, Morocco. From the relative intensities of the silicon (Si), aluminum (Al), magnesium, potassium (K), and iron (Fe) spectral peaks as well as the morphology shown by the SEI, it is concluded the particle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

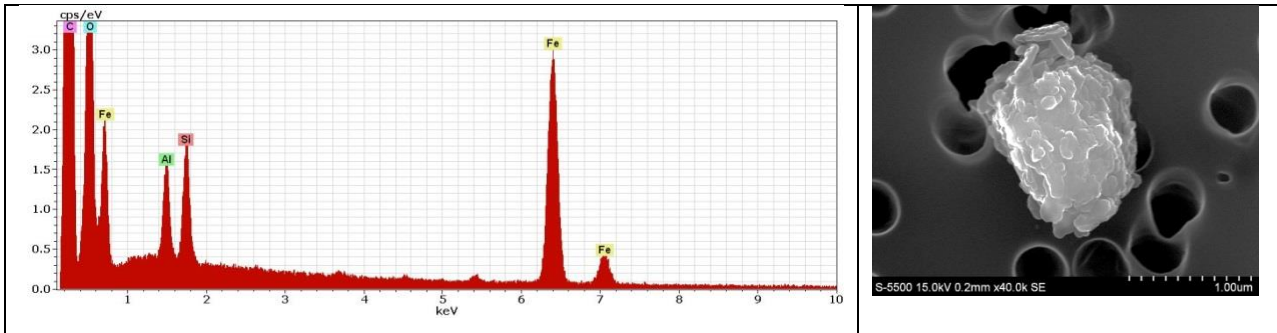



Figure 10: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1009 from Bamako, Mali. Composite grain composed of silicon (Si) and aluminum (Al) components of a clay mineral, possibly kaolinite with lesser amounts of palygorskite (finger shaped) and illite, together with iron (Fe)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

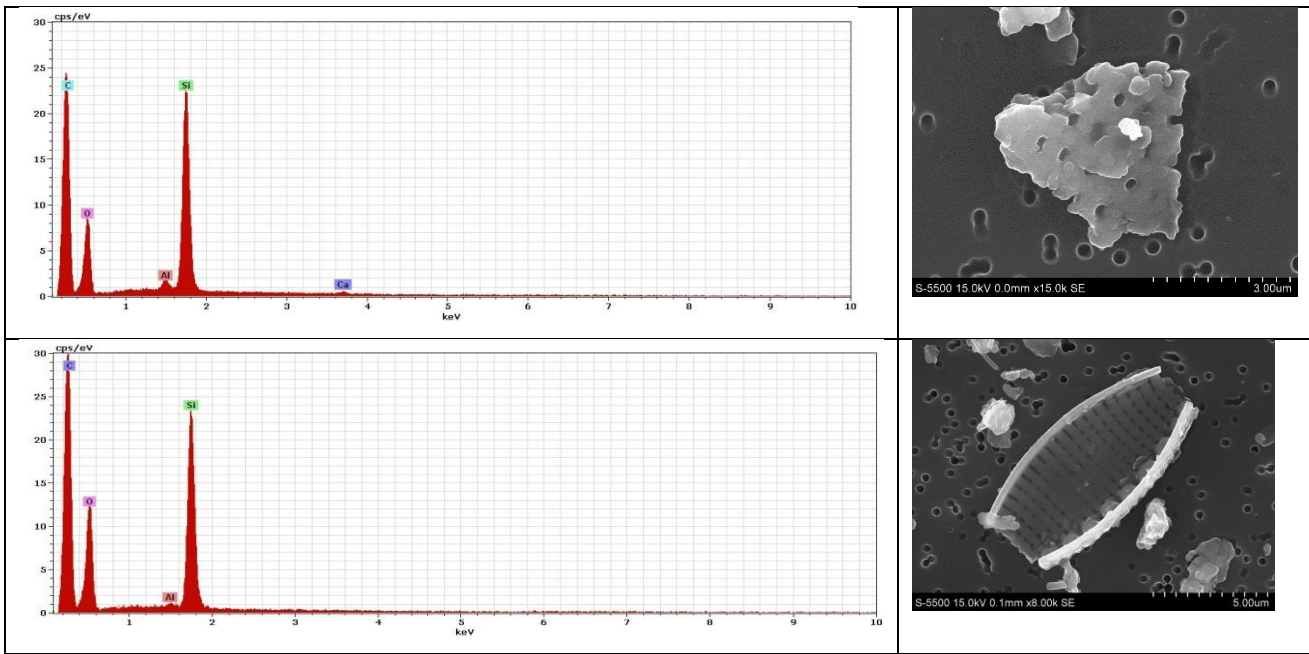



Figure 11: SEM based EDS spectra and SEI's of particles from re-suspended samples S1049 (above) and S1051 (below) from the Bodélé region of Chad. Two examples of diatom fragments, the particles contain largely silicon (Si), evidence of amorphous silica or quartz (SiO$_2$), **with small amounts of aluminum (Al) from a clay mineral coating, possible kaolinite and trace of calcium (Ca), possibly of calcite.**

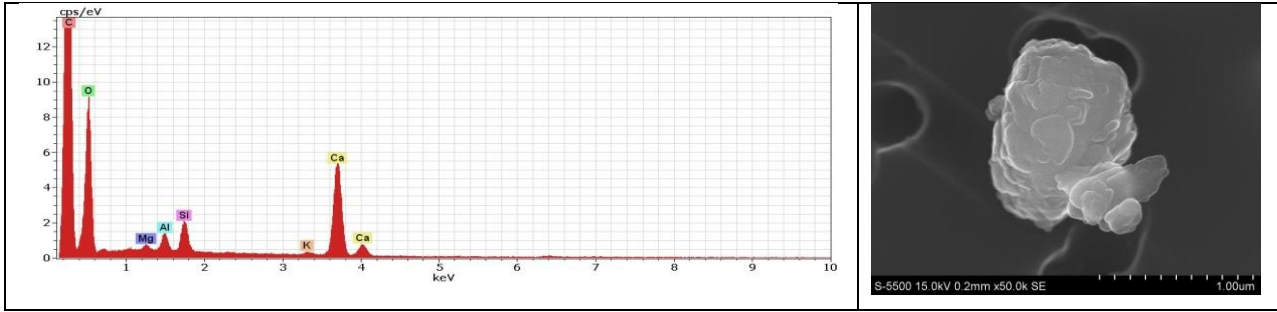



Figure 12: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1014 from northwestern China. Composite particle of calcite as seen from the high calcium (Ca) content together with a clay mineral (coating?), possibly illite as is evident from the silicon (Si), aluminum (Al), magnesium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

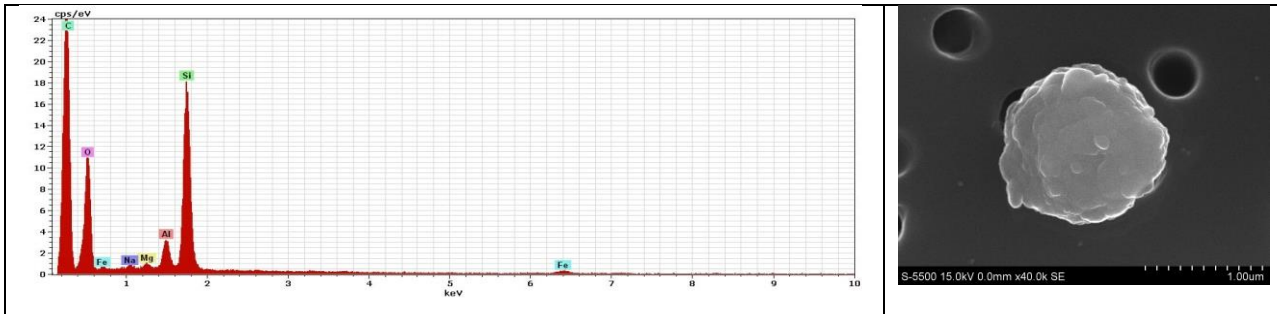



Figure 13: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1057 from Lake Eyre, Australia. Round quartz particle as evident from its silicon (Si) content, with small amounts of aluminum (Al), magnesium (Mg) and iron (Fe) from clay (coating), possibly illite or montmorillonite.

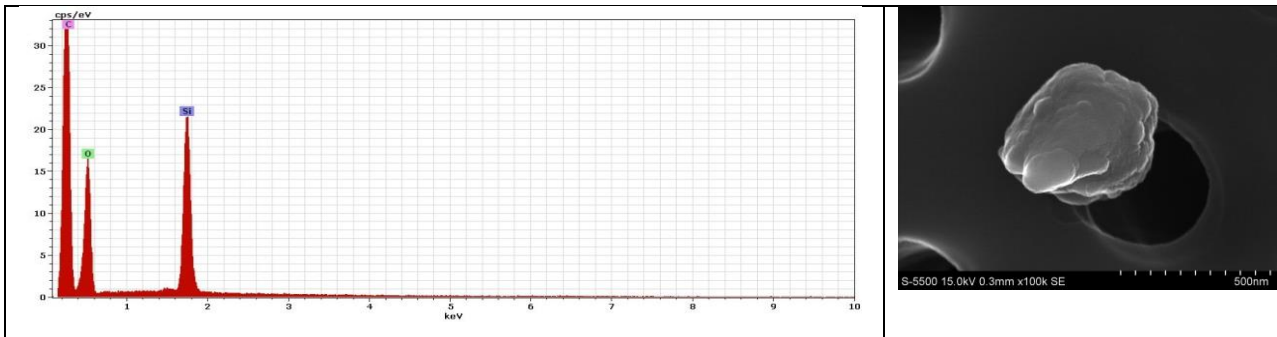



Figure 14: SEM based EDS spectrum and SEI of a particle from re-suspended sample S1062 from Kostolac, Serbia. The silicon (Si) content points to a rounded quartz particle contained in the loess.

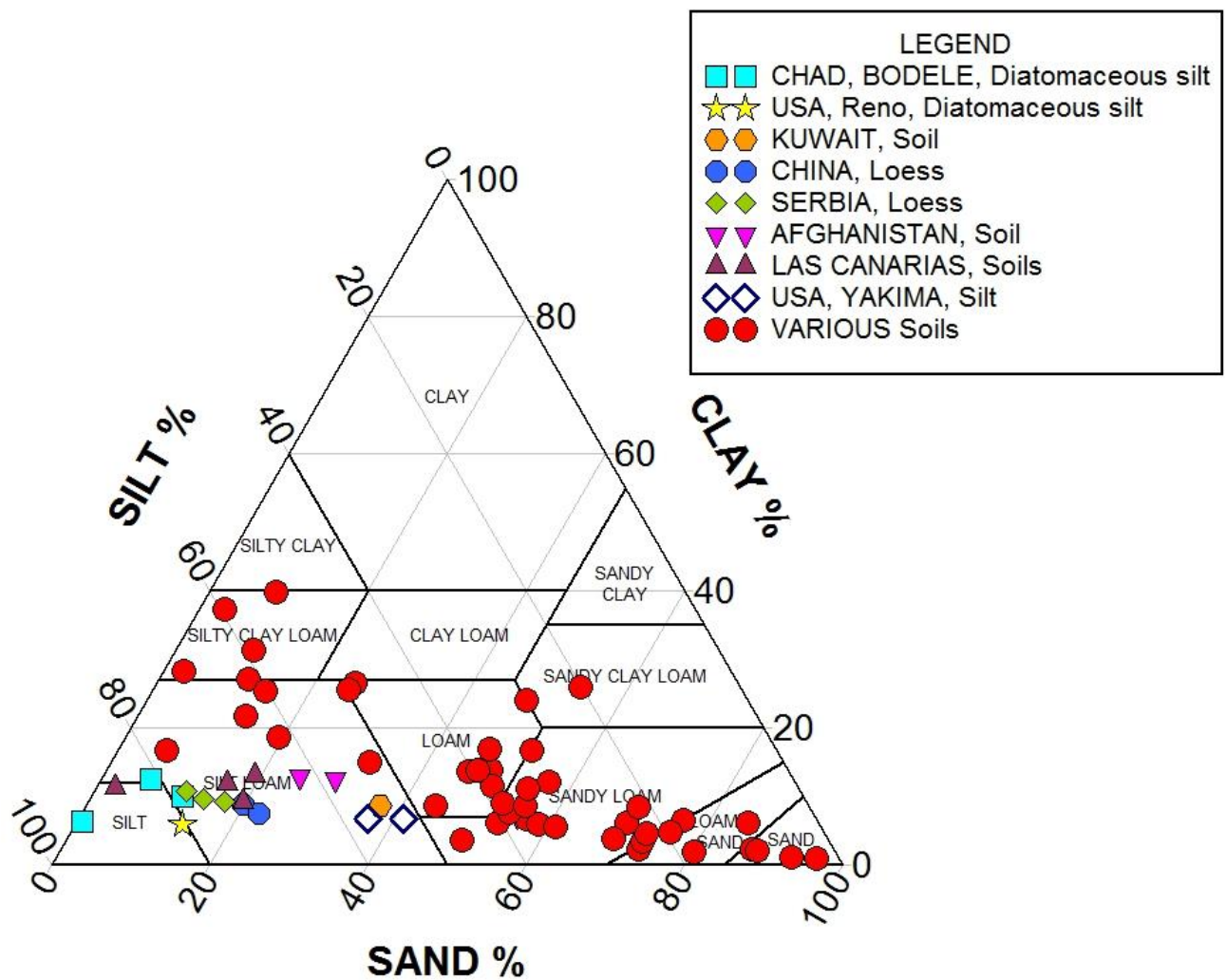



Figure 15: Size-class fractions grouped as clay (< 2 µm), silt (2 - 62.5 µm) and sand (62.5 – 600 µm). Samples from regions with the greatest potential to be sources of airborne dust lie in the "silt loam" and "silt" fields with less than about 10% clay, including samples from known dust regions in Chad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

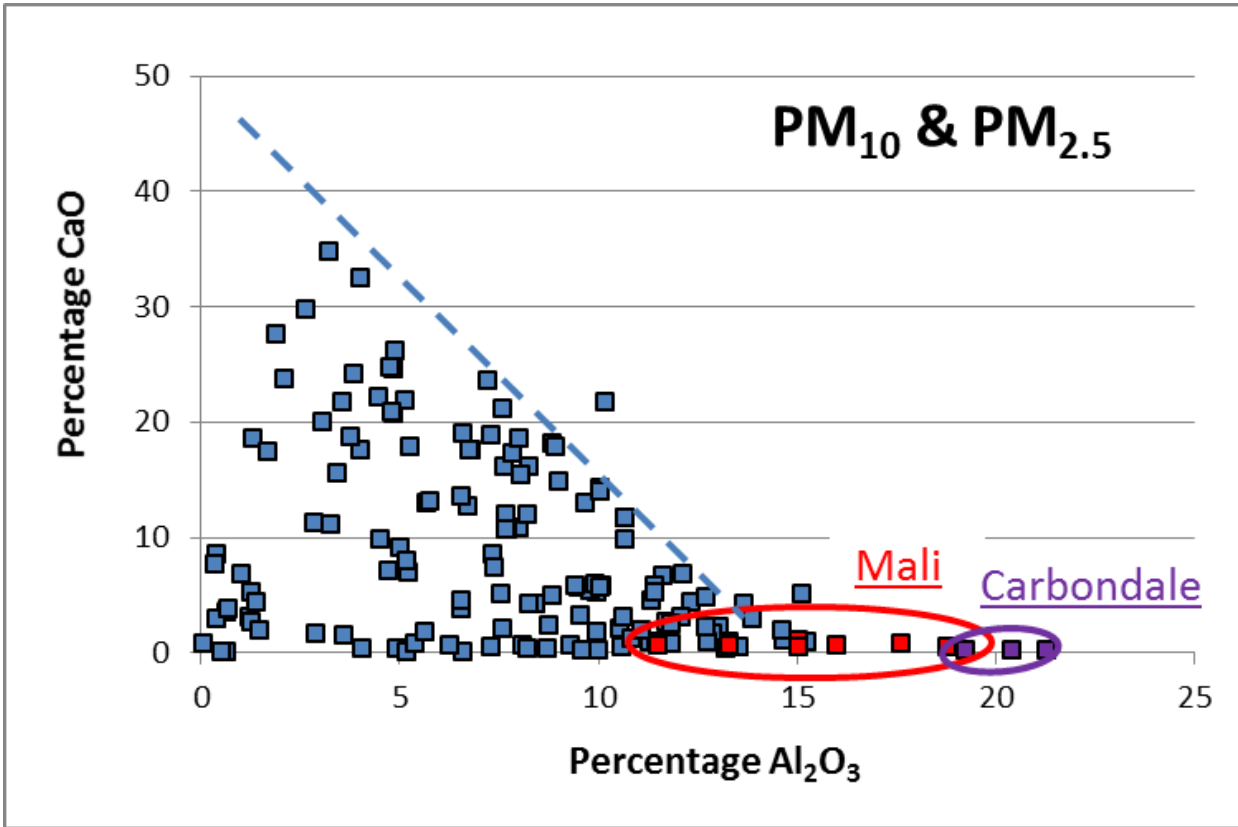



Figure 16: Triangular shaped field for the majority of the 130 dust re-suspensions, with a maximum for Al₂O₃ of about 15% and CaO of less than approximately 50%. The plot shows an inverse relationship between the two chemical species, with samples containing the highest Al₂O₃ also containing the lowest CaO, and *vice versa*. This points in part to the substitution of e.g. carbonates and gypsum for clay minerals or clay-like colloids. 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.

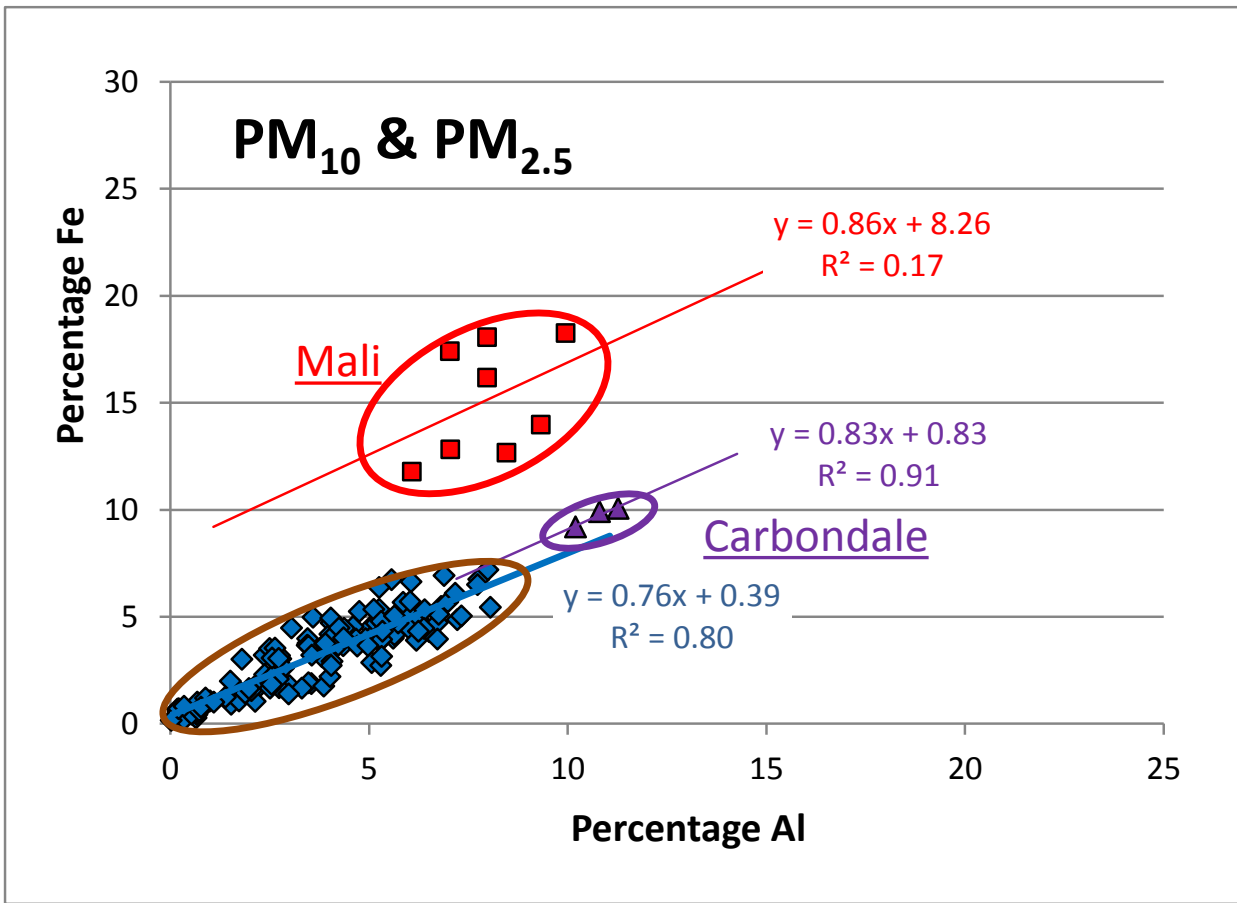



Figure 17: Plot showing the linear relationship between iron (Fe) and aluminum (Al) for 130 chemically analyzed soil re-suspensions. Three sample clusters can be identified with most (122) falling in one field (brown oval) with an average Fe/Al slope of 0.76. Samples from Mali (red oval) and Carbondale (violet oval) have slightly higher slopes. The high correlation and tight cluster of the largest (122 samples) group implies that the iron (Fe) is in part contained in the clay mineral structure or dispersed as colloidal oxihydroxide in the clay-like amorphous matrix.

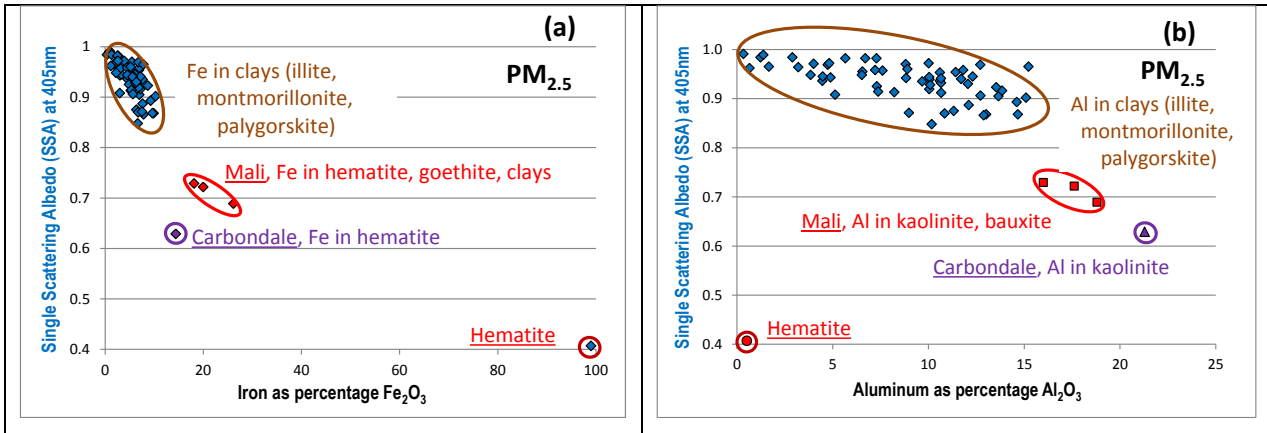



Figure 18: Single scattering albedo (SSA) at 405 nm for the PM$_{2.5}$ fractions, against (a) Fe$_2$O$_3$ and (b) Al$_2$O$_3$ content, illustrating the relationship to dust mineralogy. Most (122 samples) falling in the brown ellipse contain iron (Fe) partly in clays (illite, montmorillonite, palygorskite) and oxihydroxides (hematite, goethite), and as colloids in an amorphous clay like matrix. This group has SSA greater than 0.85, while those samples which contain all or part of the iron (Fe) as individual oxihydroxides (hematite, goethite), have SSA below 0.75.

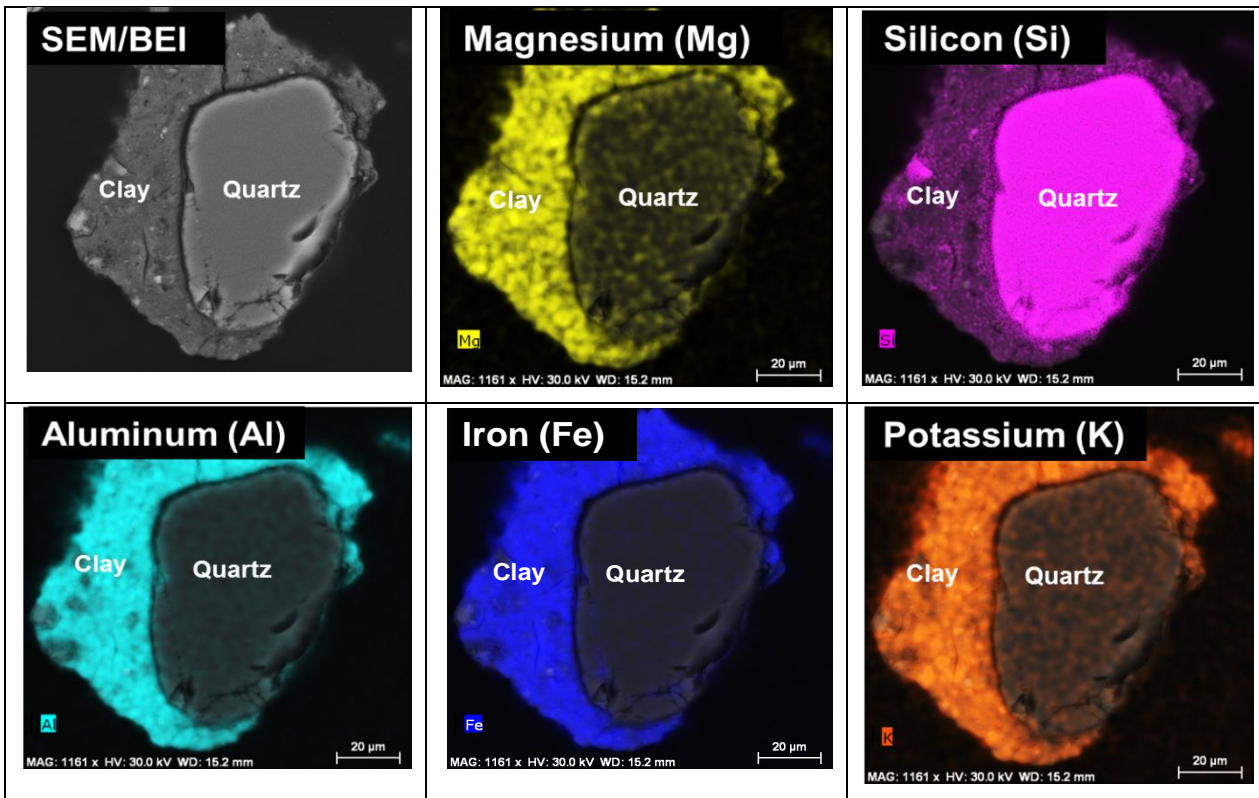
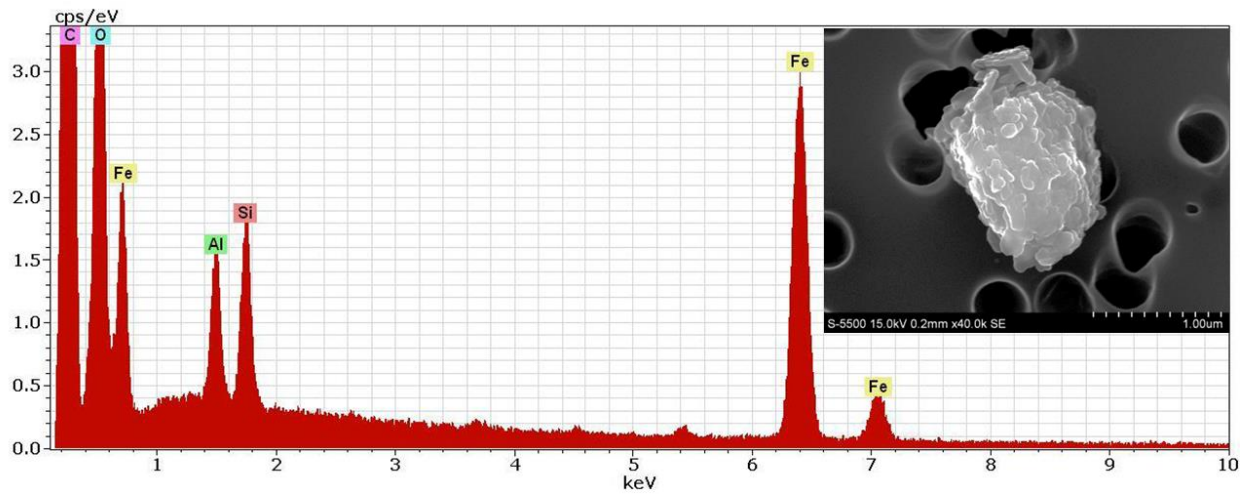



Figure 19: Sample S1056 from Lake Eyre, South Australia. SEM generated backscattered electron image (BEI) and chemical maps of a quartz grain with a prominent coating. The coating is composed of an amorphous clay matrix containing colloidal particles of sheet silicates (clay, mica), oxihydroxides (goethite, hematite) and adsorbed ions, explaining the uniform distribution of magnesium (Mg), silicon (Si), aluminum (Al), iron (Fe), and potassium (K).

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.**