Geochemical variations in aeolian mineral particles from the Sahara-Sahel Dust Corridor

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Abstract

The Sahara-Sahel Dust Corridor runs from Chad to Mauritania and expels huge amounts of mineral aerosols into the Atlantic Ocean. Data on samples collected from Algeria, Chad, Niger and Western Sahara illustrate how corridor dust mineralogy and chemistry relate to geological source and weathering/transport history. Dusts sourced directly from igneous and metamorphic massifs are geochemically immature, retaining soluble cations (e.g. K, Na, Rb, Sr) and accessory minerals containing HFSE (e.g. Zr, Hf, U, Th) and REE. In contrast, silicate dust chemistry in desert basins (e.g. Bodélé Depression) is influenced by a longer history of transport, physical winnowing (e.g. loss of Zr, Hf, Th), chemical leaching (e.g. loss of Na, K, Rb), and mixing with intrabasinal materials such as diatoms and evaporitic salts. Mineral aerosols blown along the corridor by the winter Harmattan winds mix these basinal and basement materials. Dusts blown into the corridor from sub-Saharan Africa during the summer monsoon source from deeply chemically weathered terrains and are therefore likely

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to be more kaolinitic and stripped of mobile elements (e.g. Na, K, Mg, Ca, LILE), but retain immobile and resistant elements (e.g. Zr, Hf, REE). Finally, dusts blown southwestwards into the corridor from along the Atlantic Coastal Basin will be enriched in carbonate from Mesozoic-Cenozoic marine limestones, depleted in Th, Nb and Ta, and locally contaminated by uranium-bearing phosphate deposits.

Keywords: Mineral aerosols, Sahara-Sahel dust, dust and soil geochemistry.

1. Introduction

Aeolian processes in North Africa annually transfer vast amounts of desert mineral particulates westwards to the Atlantic, Caribbean and America, and northward into the Canary Islands and mainland Europe (Prospero et al., 1981; Karyampudi et al., 1999; Zhang and Pennington, 2004; Moreno et al., 2005). Most particles travel to the African coast via the Sahara-Sahel Dust Corridor (SSDC), a huge zone lying between latitudes 12°N and 28°N and running 4000 km east-west from Chad to Mauritania (Fig. 1). This area is the World's largest aeolian dust source, and there is consequently considerable interest in its influence on global climate, ecosystems and health (Tegen et al., 1996; Goudie and Middleton, 2001; Karyampudi and Pierce, 2002; Prospero et al., 2002; Dunion and Velden, 2004; Zhang and Pennington, 2004). Not all dust outbreaks from the Sahara have the same source, and there has been a considerable amount of work in characterising African aerosol chemistry, mainly using major element variations (e.g. Chester et al., 1971; Chiappello et al., 1997), and on chemical variations linked to particle size fractionation (Schütz and Rahn, 1982). In this paper we combine several geochemical techniques (XRD, SEM-FESEM, ICP-AES and ICP-MS) to contribute to the understanding of how particle geochemistry, particularly with respect to trace elements, can vary depending on the geological source.

Samples of SSDC desert soil, road, and aeolian dusts (Fig. 1) were collected during quiet conditions from: 1. The Algerian Hoggar Massif in the area of Tamanrasset (soil samples - **HM1**: 22°55'N, 05°29'E and **HM2**: 22°47'N, 05°32'E) where the local geology is dominated by Precambrian igneous and metamorphic rocks; 2. The Bodélé Depression in Chad, the world's largest natural source of silicate dust which emanates mostly from dessicated diatomaceous lake deposits (**CB1** soil sample: 17°45'N, 17°48'E and **CB2**: resuspended dust in car air filter after journey from the Bodélé Depression to Lake Chad); 3. Aeolian dusts deposited in SW Niger (13°23'N, 2°28'E) by northeasterly winter Harmattan winds (**HAR**) and southwesterly summer monsoon winds (**MON**); 4. Western Sahara soil samples from the Atlantic coastal strip, a carbonate-rich geological depocentre initiated by Atlantic opening in Triassic times and now scoured by trade winds (Fig. 1) which regularly supply mineral particulates to the Atlantic (**WS1**: 27°50'N, 12°53'W; **WS2**: 27°02'N, 13°05'W; **WS3**: 26°37'N, 13°03'W: near the Bukraa opencast phosphate mine).

2. Particle characterisation and bulk sample major element geochemistry

1450 SEM (JEOL5900LV) individual chemical analyses (Fig. 2) on sub-angular to sub-rounded silt-sized silicate particles show them to be mostly silica (quartz and diatoms), aluminous clay minerals (kaolinite and illite-sericite), and AlSi silicates (feldspars and dioctahedral smectitic clays). Analyses were performed manually on carbon coated samples using an energy dispersive X-ray microanalysis system (EDX) with a spectrum acquisition time of 30 s live time, all particles present in several randomly chosen areas of the sample were analysed in its centre. Microscope working distance was 25 mm, accelerating voltage 20 kV, and beam current was approximately 1.00 μ A. These results were compared with XRD analyses of the samples performed in a Bruker D-5005 diffractometer. Results were obtained using CuK_α radiation, a wavelength of $\lambda=1.5405$ and a secondary graphite monochromator. Analytical conditions were step size of 0.05° (with 3 s timing per step), 40 kV and 30 mA.

The Hoggar Massif soil samples (HM1&2: Fig. 3a, b) show the greatest chemical variation and obvious derivation from the local “hard rock” geology (e.g. abundant hornblende and alkali feldspar), with XRD data confirming the presence of kaolinite, illite, montmorillonitic smectite with minor calcite and palygorskite. The Bodélé Depression lake sediment soil sample (CB1) comprises mostly quartz, clay minerals (illite, kaolinite and abundant smectite) and freshwater *Aulacoseira* diatoms (Fig. 3c), the latter commonly combined with clay to form armoured clay-diatom agglomerations (Fig. 3d). The resuspended road dust (CB2; Fig. 3e) and Harmattan aeolian dust (HAR) are mineralogically similar to, but finer grained than, CB1, although HAR has a detectable component of “hard rock” minerals such as hornblende. In contrast, the monsoon (MON) aeolian dust is depleted in diatoms and enriched in fine kaolinite, consistent with its southerly derivation because illite/kaolinite ratios in windblown dust are known to increase southwards towards the tropics (e.g. Goudie and Middleton, 2001). Finally, all three Western Sahara soil samples (WS1, 2 & 3) are greatly enriched in detrital carbonate grains (both calcite and dolomite) (Fig. 3f), and all contain palygorskite-sepiolite mafic clays (commonly found associated with carbonates in desert environments, Khademi and Mermut, (1999)).

Whole rock major element and trace elements were determined by means of Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). A weight of 0.07 g of each sample was digested using 65% pure HNO₃ and 40% pure HF for 4 h at 90 °C. Then solutions were left to evaporate at 230 °C after adding 60% pure HClO₄ (acid ratios being HNO₃: 3 HF: HClO₄). The resulting solid was then dissolved adding HNO₃ and dd water (Mili-Q) obtaining a 5% HNO₃ solution that can be analysed in both ICP-AES and MS. Digestion of international reference materials (NIST-SRM 1633b) and acid blanks were prepared following the same procedure. Analytical errors were estimated at <5% for most of the elements, and about 10% for Cd, Mo and P. High insoluble elements, such as Si, were digested with a different method. A weight of 0.05 g of

each sample was attacked with HNO_3 and HCl (1:3) and warmed at 130°C for 45 min. Solutions were then left to cool and after other 45 min mixed with a 8% diluted H_3BO_3 solution previously prepared.

Finally they were made up to 100 ml with H_2O MQ. These analyses (Table 1) also demonstrate the calcic nature of the Western Sahara group (WS1, 2 & 3), placing them between the composition of limestone and shale (Fig. 4a). The Chad (CB1&2) and Harmattan (HAR) samples cluster about a weathering trend from unweathered shale, and the Hoggar Massif samples (HM1&2), which contain the highest amounts of K and Na, plot closest to the basaltic-to-granitic silicate fractionation trend of unweathered igneous rocks (Fig. 4a). The kaolinitic monsoonal dust (MON) is both Al-rich and depleted in the soluble bases Na, K, Ca (as well as Mg), consistent with its more equatorial source where warmer and wetter conditions promote chemical weathering. In this sense the monsoon sample (MON) is the antithesis of those collected from the Hoggar Massif (HM1&2) where dry conditions and local derivation have favoured retention of the alkali elements (compare 1.92 wt% Na_2O in HM2 with 0.35 wt% Na_2O in MON).

Figure 4b estimates and subtracts the amount of carbonate and phosphate in the samples in order to obtain a clearer idea of the amount of chemical weathering that has affected the silicates in each sample. The three samples from Western Sahara (WS1, 2 & 3), with their carbonate content removed, now also plot in a similar area to those from the Hoggar Massif (HM1&2, $\text{CIA}=58\text{-}67$), suggesting the presence of a siliciclastic component of igneous/metamorphic detritus eroded from bordering West African Craton/Panafrican basement rocks. In contrast, the samples from the Chad Basin (CB1&2) and Harmattan winds (HAR) have a $\text{CIA}=76\text{-}80$, rising to 84% for the most chemically weathered dust derived from the monsoon winds (MON).

3. Bulk sample trace element geochemistry

ICP-MS and ICP-AES data (Table 1) show the highest overall concentrations of trace elements to reside in the Hoggar Massif samples (HM1&2) and the phosphatic Bukraa sediment (WS3). More specifically, HM1 and HM2 are richest in Li, V, Co, Ni, Cu, Ga, Rb, Y, Zr, Ba, Hf and Th, the samples from Western Sahara (WS) show higher Sr and As, MON is richest in Zn, and CB1 in Sn (there are Sn deposits in the Tibesti Mountains north of the Bodélé Depression). The suggestion made previously that the Harmattan sample (HAM) may contain wind-transported “hard rock” particles is supported by their relatively high Ni, Cr and Ba content (similar to or greater than HM1&2).

3.1. Large Ion Lithophile Elements (LILE).

The incompatible LILE such as Rb, Sr, Cs and Ba concentrate in the late stage products of igneous differentiation, finally entering minerals such as alkali feldspar and mica. LILE concentrations in our samples show relative depletion when compared to concentrations in average continental crust (UCC) (Fig. 5a) due to their mobility and susceptibility to chemical weathering. Thus the monsoon wind sample (MON) is especially depleted in LILE whereas both Hoggar Massif samples (HM1&2) have relatively high concentrations of Rb (higher than UCC), Sr, Cs, and Li (as well as K and Na), further emphasising the likelihood of a granitoid source yielding particles of micas, feldspar and clays derived from them. Strongly positive correlations between Rb and Ba (0.86) and Rb and K (0.98) in all six siliciclastic samples indicate similar geochemical behaviour and a distribution for these elements mainly controlled by illite (Fig. 5b). In contrast, the Sr enrichment in all three of the calcareous samples from Western Sahara (WS1-3 Fig. 5b) is explained by the chemical preference of this trace element for Ca (Deer et al., 1966).

Figure 5c plots Rb, Sr and Ba against CIA values to illustrate changes in LILE contents with increasing degrees of chemical weathering in the six siliciclastic samples. The plot again shows the Rb+Sr enrichment of the Hoggar Massif samples (HM1&2) and the depletion of

the monsoon sample (MON). The diatomaceous silt from the Bodélé Depression (CB1), despite its low SiO₂ content, has less than half the Rb content of the Hoggar samples (HM1&2) which, with the accompanying loss of Ba and K, is attributed to the relative lack of illite and K-feldspar indicated by the XRD and SEM data. In contrast, CB1 shows less depletion in Sr (and Ca and Na), consistent with the dominance of smectite over other clay minerals and the presence of evaporitic sulphates. The quartz-diluted and chemically weathered monsoon wind sample (MON) has, by contrast, very low values of both Rb and Sr, with no preferential loss of Rb.

3.2. High Field Strength Elements (HFSE).

The HFSE (e.g. Zr, Hf, Nb, Ta, Th and U) concentrate in relatively rare accessory mineral phases such as zircon, apatite, monazite, allanite, titanite and rutile. Most of these minerals occur as very small grains that are relatively resistant, especially when physical weathering processes dominate over chemical breakdown. Thus our six siliciclastic desert samples are strongly enriched in HFSE compared with UCC values (Fig. 5a). The samples from Western Sahara (WS1-3) in contrast have generally rather low HFSE contents, especially Ta (Fig. 5d) and Nb (≤ 1 ppm and 6 ppm respectively). This difference is attributed primarily to the abundance of carbonate in the Atlantic Coastal Basin (Piqué, 2001), with the consequent dilution of accessory mineral-rich detritus derived from the erosion of siliciclastic rocks. The dilution effect of adding marine carbonate particles will be enhanced in those HFSE, such as Th, which are depleted in seawater and not metabolised by marine organisms. A low concentration of Ti in all three Western Sahara samples accords with the depletion in Ta and Nb, both of which are associated with Ti-bearing accessory minerals such as rutile and titanite, and all three of which are also very depleted in seawater. A notable exception to the general pattern of HFSE depletion in the Atlantic Coastal Basin samples is provided by enriched levels of Th (16 ppm) and, especially, U (8 ppm) in the phosphatic sample WS3

from Bukraa (Fig. 5a, d). U is preferentially concentrated in phosphate ores (typically ranging from 50-200 ppm), and Th is always present at least in background amounts (Schmidt and Küppers, 1995).

The samples collected from the Hoggar Massif (HM1&2) are the richest in HFSE, with U, Th, Zr and Hf being over double UCC values in both HM1 and HM2 (Fig. 5a). Zircon is clearly an important HFSE-bearing phase, controlling Zr/Hf values which, for the six carbonate-poor samples, cluster within the narrow range of 35-43, a value strongly indicative of zircons derived from a granitoid source (Deer et al., 1966). The samples from Western Sahara (WS1-3) show a similar, but slightly broader range in Hf/Zr values. Despite the evidence of strong chemical weathering having affected the monsoon dust sample (MON), and its high silica content, it retains relatively high HFSE concentrations (Fig. 5e), interpreted as reflecting the highly resistant nature of accessory mineral particles such as zircon and monazite at source and during transport. In contrast, Zr, Hf and Th are relatively depleted in the Bodélé Depression sample CB1 irrespective of its low CIA value (Fig. 5e) and SiO₂ content. We suggest that the explanation for this lies in the physical winnowing out of heavy mineral particles such as zircon and monazite during the transport history of accessory minerals into the basin.

3.3. Rare Earth Elements (REE).

REE are usually concentrated in accessory minerals such as bastnasite, monazite, xenotime, allanite, apatite, titanite, and zircon, although some clays also are REE-enriched. These REE-bearing phases tend to be not only stable and resistant to decay, but small enough to be incorporated into dusts, so that REE content in desert particulates is primarily controlled by source rocks and soils. Normalising our REE data to average UCC all samples are relatively enriched, with the exception of those most diluted by silica (CB2) and by carbonate (WS2). Our samples show LREE fractionation, a negative Eu anomaly, and nearly flat HREE,

(Fig. 5a) consistent with a felsic provenance for the REE-bearing host minerals. Figure 5a also shows MREE (such as Gd) and HREE (such as Yb and Lu) have been preferentially fractionated over LREE (such as La), which suggests the presence of relatively LREE-depleted detrital minerals such as zircon and xenotime rather than monazite and bastnasite (both LREE-enriched). As with most other trace elements, the two samples from the Hoggar Massif (HM1&2) have high Σ REE values (>260 ppm), although the Western Sahara sample WS3 shows slightly higher concentrations (Σ REE=299; Fig. 5f), an observation once again presumably linked to the enhanced phosphate content at Bukraa. With only minor exceptions, REE behave similarly in all samples (including the three from Western Sahara) and there is no REE fractionation between them. It may be concluded from this that whatever the differences in weathering and transportation history of these samples, superficial processes have not acted to change REE concentrations relative to each other. As with LILE and HFSE, sample CB1 (Σ REE=109) is relatively depleted in REE, whereas MON (Σ REE=199) is enriched despite its high CIA and SiO_2 (Fig. 5g). Such REE enrichment of the most chemically weathered sample may owe its origin to the relative durability of REE-bearing accessory phases, and/or enhancement of REE content during deep weathering of soils in the source area (c.f. Nesbitt and Markovics, 1997).

3.4. *La-Th-Sc*.

Comparisons between the LREE Lanthanum, the HFSE Thorium, and the compatible transition element Scandium offer further insight into the geochemical character of these particulate samples (Fig. 6). The lack of a significant contribution of mafic rocks is indicated for our samples which all plot far from the 100%Sc apex, in a region characteristic of chemically evolved, granitoid compositions within a continental margin setting (e.g. Nyakairu and Koeberl, 2001; Cullers, 2002). A granitoid source is also likely when considering Th/Sc, Th/Cr and Eu/Eu* ratios (Table 1: compare Cullers, 2002, Fig. 7). Figure 6 indicates a trend

away from Th (and, to a lesser extent, La) from the Hoggar Massif samples (HM1&2) to those from Chad (CB1&2). This trend is not attributable to igneous fractionation, which produces a line moving directly away from Sc. Neither is it a function of chemical weathering, as illustrated by the intermediate position of the high CIA sample MON. One explanation for this would be the geochemical effect of physically removing Th by the loss of heavy minerals such as monazite and zircon during sedimentary transport. The anomalously high levels of La and Th in the phosphatic Western Sahara (WS3) have displaced this sample from the trend shown by the other samples.

4. Discussion

The following major geological source areas will supply mineral particles to the SSDC: (i) the old Precambrian and Palaeozoic massifs; (ii) the young siliciclastic basins running from Chad to Mauritania; (iii) the Atlantic Coastal Basin; (iv) the deeply weathered ancient basement of the sub-Saharan lands to the south. Within this context we place our samples within five geochemical groups of particulates each of which will contribute to the mineral aerosol loading of the Sahara-Sahel Dust Corridor.

1. Atlantic margin-type particulates: These have a geochemistry strongly influenced by the weathering and erosion of Mesozoic-Cenozoic Atlantic margin marine limestones and marls: abundant carbonate (and therefore Ca, Mg and Sr), the presence of hematite and mafic clays such as palygorskite, and a depletion in Ti, Nb, Ta and Rb. HFSE and REE contents are normally low, except where associated with localised phosphate deposits such as at Bukraa. Background silicate content, diluted by the carbonate particles, was probably sourced from relatively weakly chemically weathered granitoid basement rocks of the Saharan desert hinterland. Many of these mixed carbonate-silicate “Atlantic Margin” dusts will be blown out over the ocean by the trade winds and will add their calcareous signature to the export of other more siliciclastic particulates from Africa into the eastern Atlantic and beyond.

2. Saharan cratonic basement-type particulates: these are relatively rich in the more soluble major elements (Na, K, Ca and Mg) and LILE, and contain abundant REE and HFSE in unaltered host accessory minerals locally derived from granitoid protoliths in West African cratonic basement massifs. Such particulate material is an important source of the “new dust” of Evans et al. (2004): recently formed particles derived from the weathering and erosion of desert rock exposures. With time, much of this material will be washed by ephemeral desert rains southwards and westwards into the Iulemmeden and Taoudeni basins. Some will be picked up en route by duststorms and lofted into the atmosphere, potentially adding its still-discernably trace element-rich granitoid geochemical signature to the Saharan Air Layer.

3. Saharan sedimentary basin-type particulates: these sediments lack primary mafic minerals and have a simple felsic composition of mostly clay minerals, quartz and diatoms. Weathered silicate particles from a mostly granitoid basement have been transported into the former freshwater lake system and mixed with intrabasinal clays, biological detritus and evaporitic salts. This is the “old dust” of Evans et al. (2004), with particles commonly having a polycyclic history involving repeated fluvial and aeolian transport and sedimentation going back millions of years. The major element geochemistry indicates a higher CIA than the Hoggar Massif (HM) samples, a bulk chemistry similar to “weathered shale”, and a depletion in trace elements, including most metals and especially Rb, Zr, Hf and Th, due to physical winnowing and chemical breakdown. The final product of these processes, winnowed from the desert surface of the Bodélé Depression and transported through the SSDC, provides an estimated 50% of all the dust that leaves the West African coast (Washington et al., 2003).

4. Harmattan dust: The Harmattan winds transport basinal diatomaceous dusts westwards along the SSDC, mixing them with lesser amount of particulates from the surrounding basement massifs, particular those to the north, and thus enriching them in “hard rock” minerals such as hornblende, and trace elements such as Ba, Rb, Zr, Hf, Σ REE and Th.

5. Summer monsoon dust: These kaolinitic dusts blow into the SSDC from sub-Saharan Africa, and will have a high CIA reflecting a derivation from deeply chemically weathered terrains. Such weathering has stripped these dusts of their mobile elements (Na, K, Mg, Ca, and LILE), whereas in contrast, the more immobile elements (notably Zr, Hf and REE) show relatively high concentrations, despite the effects of quartz dilution. This group of particulate materials thus has its own distinctive geochemical character that will add dust to the SSDC during the summer months.

To conclude, the regular dust intrusions that move westwards across the ocean from the African coast will derive from different sources and therefore potentially contain different mineral mixtures. Dry air outbreaks between latitudes 15°-25°N usually emanate either from the central Sahara-Sahel Dust Corridor (especially the Bodélé Depression) itself or originate from (and propagate along) the northwestern African Coast (Zhang and Pennington, 2004). These atmospheric outbreaks from the east and northeast merge into the Saharan Air Layer between latitudes 10°-25°, and commonly travel to the other side of the ocean, retaining their identity for over a week (Dunion and Velden, 2004; Zhang and Pennington, 2004). Atmospheric dust carried by these dry air outbreaks affects outgoing longwave radiation (Haywood et al., 2005), and some minerals (notable iron oxides) are more light absorptive than others (Lafon et al., 2004). Long range transport of dusts will inevitably tend to alter the bulk aerosol chemistry by mixing and size fractionation, thus obscuring geological signatures from specific source areas (e.g. Schütz and Seibert, 1987; Gullu et al., 1996; Claquin et al., 1999). It remains unproven, however, whether dust populations carried rapidly westwards by dry air outbreaks can at times retain a recognisable “geological” signature and, if so, whether such subtle compositional differences could influence physical interaction with the atmosphere over the ocean.

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Table caption:

Table 1. Major (wt% oxides) and trace (ppm) element analyses determined by means of ICP-AES and ICP-MS respectively. CIA: Chemical Index of Alteration. $Eu/Eu^* = Eu_{cn}/(Sm_{cn} \times Gd_{cn})^{1/2}$.

Figure captions:

Fig. 1. Maps of the Sahara-Sahel Dust Corridor showing (left) sample localities, outcrop of old basement rocks and young sedimentary basins, prevailing winds (inset), and (right) TOMS 13-year (1992-2005) averaged aerosol concentrations, highlighting the dust source hotspots of the Bodélé Depression and Taoudeni Basin (http://toms.gsfc.nasa.gov/aerosols/aerosols_v8.html; Bristow, 2005). MON: Monsoon, CB1&2: Chad Basin, HAR: Harmattan, HM1&2: Hoggar Massif, Algeria, WS1-3: Western Sahara.

Fig. 2. Al/Si v (Mg+Fe+Al+Si) diagrams for all samples plotting SEM-EDX single particle analyses (see Moreno et al., 2003 for methodology). Si felsic: silica (quartz, diatomaceous hydrated amorphous silica), AlSi felsic: alkali and plagioclase feldspar, smectite clays, Al felsic: Al-clays (kaolinite, illite), muscovite, MgFe mafic: biotite, chlorite, mafic clays, Mg mafic: amphibole, pyroxene, talc, serpentine, Mg-clays, CC: carbonates (calcite and dolomite).

Fig. 3. FESEM images showing morphology and particle size of samples: a) Hoggar Massif (Algeria) desert soil sample HM2, b) Close up of a relatively fresh alkali feldspar in HM2; c) Resuspended road dust Chad Basin sample CB2: note mix of rock forming minerals and diatomaceous materials; d) Typical clay-diatom agglomerated clast in CB1; e) Detail of an *Aulacoseira* diatom from CB2 sample, f) dolomite from Western Sahara (WS) soil samples.

432

433 Fig. 4. a) Al_2O_3 -($\text{CaO}+\text{Na}_2\text{O}$)- K_2O triangular diagram (excluding $>30\%$ K_2O) illustrating bulk
434 compositional differences between sample groups, common rock types, and average upper
435 continental crust (UCC). Rock types and UCC data from Ronov, 1982; Gray and Murphy,
436 2002; Rudnick and Gao, 2003. b) Al_2O_3 -CN- K_2O diagram (Nesbitt and Young, 1984, 1989;
437 Nesbitt et al., 1996) produced by subtracting the amount of carbonate and phosphate in the
438 samples to allow estimation of the amount of chemical weathering that has affected the
439 silicates in each sample. $\text{CN} = \text{Na}_2\text{O}/62 + \text{Ca}^*$ where $\text{Ca}^* = [(\text{Na}_2\text{O} \times 0.35 \times 2)/62] - (3.3 \times$
440 $\text{P}_2\text{O}_5/142)$: Ca-Na ratio estimated as 1:3 on a molar basis. Note increasing chemical
441 weathering trend from Hoggar Massif samples (HM1&2) to the Monsoon sample (MON) (see
442 text for discussion). (MON: Monsoon, CB1&2: Chad Basin, HAR: Harmattan, HM1&2:
443 Hoggar Massif, Algeria, WS1-3: Western Sahara).

444

445 Fig. 5. a) Samples normalised values to upper continental crust. LILE: Large Ion Lithophile
446 Elements, HFSE: High Field Strength Elements, LREE and HREE: Light and Heavy Rare
447 Earth Elements; b, d, f) Relative abundances of selected LILE (b), HFSE (d), and REE (f) in
448 all samples compared to average upper continental crust (UCC); c, e, g) Relative abundances
449 of selected LILE (c), HFSE (e), and REE (g) plotted against Chemical Index of Alteration
450 (CIA) values. (MON: Monsoon, CB1&2: Chad Basin, HAR: Harmattan, HM1&2: Hoggar
451 Massif, Algeria, WS1-3: Western Sahara).

452

453 Fig. 6. La-Th-Sc triangular diagram (excluding $<50\%$ La) illustrating the granitoid signature
454 of our samples (rock types data from Nyakairu and Koeberl, 2001; Cullers, 2002). (MON:
455 Monsoon, CB1&2: Chad Basin, HAR: Harmattan, HM1&2: Hoggar Massif, Algeria, WS1-3:
456 Western Sahara).

Fig. 1

Fig. 2

Fig. 3

Fig 4

Fig 5

Fig 6