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Three North African Dust Source Areas and their Geochemical Fingerprint

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Abstract

North Africa produces more than half of the world's atmospheric dust load. Once entrained into the atmosphere, this dust poses a human health hazard locally. It also modifies the radiative budget regionally, and supplies nutrients that fuel primary productivity across the North Atlantic Ocean and as far afield as the Amazonian Basin. Dust accumulation in deep sea and lacustrine sediments also provides a means to study changes in palaeoclimate, particularly those associated with rainfall climate change. Systematic analysis of satellite imagery has greatly improved our understanding of the trajectories of long-range North African dust plumes, but our knowledge of the dust-producing source regions and our ability to fingerprint their contribution to these export routes is surprisingly limited. Here we report new radiogenic isotope (Sr and Nd) data for sediment samples from known dust-producing substrates (dried river and lakes beds), integrate them with published isotope data and weight them for dust source activation. We define three isotopically distinct preferential dust source areas (PSAs): a Western, a Central and an Eastern North African PSA. More data are needed, particularly from the Western PSA, but our results show a change in PSA dust source composition to more radiogenic Nd- and less radiogenic Sr-isotope values from west to east, in line with the overall decreasing age of the underlying bedrock. Our data reveal extreme isotopic heterogeneity within the Chadian region of the Central PSA, including an extremely distinctive geochemical fingerprint feeding the Bodélé Depression, the most active dust source on Earth. Our new analysis significantly improves the reliability by which windblown dust deposits can be geochemically fingerprinted to their distant source regions.

Keywords:

North Africa, dust source, Bodélé Depression, radiogenic isotopes, εNd, 87Sr/86Sr

1. Introduction

Atmospheric dust is a key component of Earth's climate system, influencing the global radiative budget both directly by controlling absorption and scattering of solar radiation, and indirectly by stimulating cloud condensation and cover (Thompson et al., 2019). Today, North Africa exports more dust to the atmosphere than any region on Earth with an estimated annual production of 170 to 1600 Tg yr^{-1} (Engelstaedter et al., 2006), over half the total global flux (Ginoux et al., 2012). In some regions, these loadings constitute a serious risk to human health; high concentrations of atmospheric particulate matter with an aerodynamic diameter of less than 2.5 µm ($PM_{2.5}$) are estimated to have led to between 194,000 and 709,000 infant deaths in North Africa in 2015 alone (Heft-Neal et al., 2018). Once lofted into the atmosphere, North African dust can be carried thousands of kilometres, primarily to the west, in plumes that are visible from space and are suggested to transport Fe and other fertilizing micronutrients to sites of primary production in the surface waters of the North Atlantic Ocean (Jickells, 2005) and the rainforests of the Amazonian Basin (Koren et al., 2006; Yu et al., 2015). The accumulation of this dust in marine or lacustrine sediments also provides a way to investigate past changes in climate variability on geological timescales (Cole et al., 2009; Grousset et al., 1998; Tiedemann et al., 1994).

Systematic analysis of satellite imagery has greatly improved our understanding of the generation and export of North African dust. Key developments include providing the spatial coverage to map the locations of highest dust generation (Engelstaedter et al., 2006;

Schepanski et al., 2012) and establishing that there are two main transport pathways westwards, a “northern route” towards the Caribbean which dominates during boreal summer, and a “southern route” towards the Amazon during winter (Engelstaedter et al., 2006; Meng et al., 2017). Yet despite these recent advances, major uncertainties remain in our knowledge of North African dust-producing source regions and our ability to fingerprint their contribution to these export routes (Bakker et al., 2019; Formenti et al., 2011; Scheuvens et al., 2013). This limited knowledge base hinders the development of a comprehensive source-to-sink understanding and adds uncertainty to climatic interpretations that rely on the accumulation of dust in geological archives.

The first studies employing satellite-derived data in North Africa inferred dust sources from high atmospheric dust loads using AI (Aerosol Index) or AOT (Aerosol Optical Thickness) data from the Total Ozone Mapping Spectrometer (TOMS) or Ozone Monitoring Instrument (OMI) (Engelstaedter et al., 2006; Israelevich et al., 2002; Middleton and Goudie, 2001; Prospero et al., 2002) (Figure 1A). These analyses provided an invaluable first-look at the problem but were limited by a daily temporal resolution that conflated dust emission and dust transport (Schepanski et al., 2012). More recent work has employed the thermal infra-red (IR) radiances dataset from the Meteosat Second Generation (MSG) Spinning Enhanced Visible and InfraRed Imager (SEVIRI) which benefits from a much higher temporal resolution (every 15 minutes versus every 24 hours for TOMS and OMI). This higher temporal resolution approach allows dust source activation events to be identified at hourly resolution and geo-located by manually tracking dust plumes back to their precise origin (Schepanski et al., 2012, 2007). The resulting dust source activation frequency (DSAF) maps therefore effectively remove transport bias altogether (Figure 1B).

Once defined geographically by remote sensing analysis, preferential source areas of dust generation (PSAs) are characterised geochemically to permit dust provenance studies (e.g., Figure 1C & 1D). The primary tools that have been used in this endeavour are the radiogenic isotope analysis (mainly ε_{Nd} and $^{87}Sr/^{86}Sr$) of bedrock and associated lithogenic sediments. Yet, there are two main problems associated with existing interpretations of this kind. First, previous landmark studies noted the disparity in dust sources identified by the different remote sensing techniques outlined above and combined the results of the these approaches to identify six PSAs in North Africa (Fig. 1C, Scheuvens et al., 2013). However, this approach incorporates residual transport bias in dust emission estimates (Schepanski et al., 2012, 2007). Second, a large fraction of the Sr and Nd isotope data set currently available does not come from active deflating dust source regions (Abouchami et al., 2013; Blanchet, 2019; Gross et al., 2016; Scheuvens et al., 2013; Zhao et al., 2018). Dust in North Africa is mostly derived from distinct palaeolake- and alluvial- deposits, which are located in the foothills of Saharan mountain ranges (Bakker et al., 2019; Schepanski et al., 2009). Surrounding bedrock and sand sediments contain very limited fine-grained material, which inhibits dust production from these surfaces (Bullard et al., 2011). While samples from bedrock and sand deposits in the vicinity of dust sources are useful for investigating local sediment dynamics, they are not well-suited to fingerprinting the signature of dust transported over thousands of kilometres. Moreover, existing radiogenic isotope data have been generated on differing grain size fractions, ranging from bulk sediment measurements to < 2 µm. While the Nd isotope composition of aeolian dust is considered insensitive to grain size, Sr isotope data are widely suggested to be susceptible to a substantial grain-size dependent fractionation (with an increase in $^{87}Sr/^{86}Sr$ of ~ 0.01 in the < 2 µm fraction compared to the >50 µm fraction, (Feng et al., 2009). Here we address these issues in identifying the geochemical signature of exported dust by presenting new PSAs, geographically defined based on the dust source activation frequency (DSAF) map of Schepanski et al. (2012) and isotopically characterised using our new Sr and Nd data combined with existing datasets from known dust sources. Our overarching aim is to improve the reliability by which dust deposits can be geochemically fingerprinted to their distant source regions.

2. Materials and Methods

2.1 Geographical definition of PSAs using remotely sensed data

To identify dominant dust source areas in North Africa and to guide sampling, we used the DSAF map of Schepanski et al. (2012), generated using MSG SEVIRI thermal IR radiances (Figure 1B & Figure 2A). It is based on highly resolved data (temporal resolution of 1-hour, spatial resolution of 3 km x 3 km) collected from the geostationary MSG satellite, allowing dust source activation events to be identified and traced back to their point of origin (see Schepanski et al., (2007, 2012) for their methodological details). We used their 1 ° x 1 ° annual DSAF map of the region between 5 °N; 20 °W and 40 °N; 40 °E for from March 2006 to February 2010 (Schepanski et al., 2012) to select samples for geochemical characterisation of PSAs.

2.2 Dust samples

Our sample set consists of sediments from dried lake and river beds from identified dust source regions in Chad (specifically the Bodélé Depression, Ennedi Mountains, the Bahr El Gazel and Lake Fitri), Morocco (Wadi Draa in the Zagora and M’Hamid regions), Sudan

(Nubian Desert) and Mauritania (Sebkhet Chemchan) (Figure 2A). Samples were collected during field campaigns between 2005 and 2019 and details are listed in Table 1.

2.3 Radiogenic Isotope analysis

Radiogenic isotope (Sr and Nd) data were generated on 42 sediment samples. Of these, 32 samples were dry sieved to isolate the < 32 µm fraction. To investigate the influence of changing sediment grain size on Sr and Nd isotope composition, five samples were further sieved to separate the 63 - 45 µm, 45 - 32 µm, and < 32 µm fractions, and then passed through a dust chamber to separate the < 10 µm fraction. Bulk measurements were obtained from nine samples from nearby the Bodélé Depression in Chad, and one from Mauritania. All samples were decarbonated in excess 10% (v/v) acetic acid overnight on a shaker table. The carbonate-free fraction was then rinsed three times with MQ water.

Radiogenic isotope analyses were carried out in a clean chemistry laboratory at the University of Southampton Waterfront Campus, NOCS. Approximately 100 mg of decarbonated sample was digested overnight using a HF-HNO_3-$HClO_4$ acid mixture at 130 °C. The digestion blanks (i.e. acid residue after full digestion procedure with no sample) were below detection level (level of detection was 0.051 ppm & 0.266 ppm for Nd and Sr respectively, calculated by $\bar{x}_{blank} + 3\sigma$). Nd and Sr were isolated using chromatographic column separation (adapted from Bayon et al. (2002)). Nd was isolated using a cation column (Bio-Rad AG-50W-X8 resin) followed by a reverse phase column (Ln-spec resin) (Pin and Zalduegui, 1997). Sr was isolated using Sr-spec resin. The total column blanks (i.e. when blank acid is run through the column procedure) were negligible (50 pg and 30 pg) compared to the total amounts analysed (1 µg and 200 ng) for Sr and Nd respectively.

Nd-isotope ratios were measured using a Thermo Scientific Neptune multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS). $^{143}Nd/^{144}Nd$ compositions were corrected following the method of Vance and Thirlwall (2002) through adjustment to a $^{146}Nd/^{144}Nd$ ratio of 0.7219 and a secondary normalisation to $^{142}Nd/^{144}Nd$ = 1.141876. Results for the JNdi-1 reference standard (Tanaka et al., 2000) measured as an unknown were 0.512115 with an external reproducibility of the ±0.000006 (2SD) across 6 analysis sessions over two years. For convenience $^{143}Nd/^{144}Nd$ is reported here in epsilon notation (ε_{Nd}), where $^{143}Nd/^{144}Nd_{CHUR}$ represents the Chondrite Uniform Reservoir value of 0.512638 (Jacobsen and Wasserburg, 1980):

$$\varepsilon_{Nd} = \left[\frac{{}^{143}Nd \big/ {}^{144}Nd_{sample}}{{}^{143}Nd \big/ {}^{144}Nd_{CHUR}} - 1 \right] \times 10^4$$

After column separation, the Sr fraction was dried down and loaded onto an outgassed tantalum filament with 1µl of a tantalum activator solution. The samples were analysed on a ThermoScientific Triton Plus Thermal Ionisation Mass Spectrometer (TIMS) using a multi-dynamic procedure with an ^{88}Sr intensity of 2V. Fractionation was corrected using an exponential correction normalised to $^{86}Sr/^{88}Sr$ = 0.1194. NIST987 (Yobregat et al., 2017) was run as a standard on each turret alongside our samples and was measured at 0.710241 ± 0.000013 (2SD) on 4 analyses. The long-term average for NIST987 on this instrument is

0.710243 ± 0.000020 (2SD) from 464 analyses. Rock standard JB-1a was run through the same digestion and chemical separation procedures to give $^{87}Sr/^{86}Sr$ and ε_{Nd} values of 0.704112 ± 0.000014 (2SE) and 2.97 ± 0.17 (2SD) respectively (within accepted values, GeoReM: Jochum et al., 2005)).

2.4 Generating geochemically representative PSA compositions

Our approach to geochemically defining PSA compositions differs in two important ways from the previous studies. First, our PSA compositions are defined using only data from dust producing substrates. We combined our new data with published data from dust-producing substrates such as lacustrine and riverine sediments and soils, but excluded data from bedrock and aerosol samples. Second, we applied a dust source activity weighting to the isotope data. In the past, when geographically defined using remote sensing methods that conflate dust deflation and transport, PSA signatures were defined using the ranges of isotopic data generated on all samples that fall within those PSA regions, defining a “box” or field in Sr-Nd space. While this simple method is a sensible first order approach, it has the important limitation that equal weighting is given to all isotopic measurements, regardless of the contribution of the substrate to atmospheric dust loading. We applied a weighting to the isotope data according to annual DSAF (in each case the DSAF value from the corresponding 1 ° x 1 ° square was used as a multiplier) (Schepanski et al., 2012). To reduce bias caused by uneven sampling, where there are multiple samples available within one 1 ° x 1 ° square, the isotope data were averaged and the resulting value was weighted by DSAF. The weighted mean isotopic signature for each PSA is calculated by:

$$\bar{x}_{PSA} = \frac{\sum(x_i \times w_i)}{\sum w_i}$$

$$where\ x_i = sample\ isotope\ signature, w_i = DSAF$$

The weighted mean standard deviation for each PSA is calculated by:

$$st.dev_{PSA} = \sqrt{\frac{\sum(w_i\ (x_i - \bar{x}_{PSA})^2)}{\frac{(N-1)\sum w_i}{N}}}$$

$$where\ x_i = sample\ isotope\ signature, w_i = DSAF,\ N = no. of\ samples$$

We also calculate the percentage coverage for each PSA (i.e. the proportion of 1 ° x 1 ° squares of DSAF > 0 % that have an isotopic value assigned).

3. Results and Discussion

3.1 PSA geomorphology

The distribution of DSAF in North Africa changes seasonally. For most of the year, particularly in boreal summer when dust export is greatest (Engelstaedter and Washington, 2007) (Figure 2B), there are three main geographically distinct hotspots of dust source activation (i) West Sahara/Sahel (Southern Algeria, North East Mali, West Niger, North Mauritania), (ii) Central Sahara/Sahel (Chad) and (iii) East Sahara/Sahel (North Eastern Sudan) (Schepanski et al., 2007), hereafter the Western, Central and Eastern source regions (Figure 2C). These hotspots are separated by topographic highs; the Hoggar and Aïr mountains between the Western and Central source regions, and the Ennedi Mountains between the Central and Eastern sources (Figure 2C). Furthermore, the three dust source regions are sufficiently distinct geographically to mean that they lie within separate palaeo river catchment basins (Drake et al., 2011).

The most active dust sources in North Africa are desiccated river and lake beds (Prospero et al., 2002), contributing approximately 36% and 64% of total North African winter dust respectively (Bakker et al., 2019). The Western source region (Figure 2) is dominated by dust production from alluvial deposits and palaeolakes in the deserts that surround the Aïr and Adrar Iforas Mountains spanning Algeria, Mali and Niger. Similarly, in the Eastern source region (Figure 2), dust derives largely from alluvial deposits in the Nubian desert (Bakker et al., 2019). The Central source region is the most active of the PSAs, and its main contributor is the Bodélé Depression. The Bodélé Depression is often described as the “dustiest place on Earth” and is estimated to contribute 50% - 64% of all North African dust (Bakker et al., 2019; Bristow et al., 2009; Engelstaedter et al., 2006; Evan et al., 2015) because of the combination of strong near surface winds funnelled between the nearby Tibesti and Ennedi mountains, and the large reservoir of easily deflatable, low density diatomite-rich sediment (Bristow et al., 2009; Washington et al., 2006). DSAF data may even underestimate the importance of this region as a dust source due to its extremely high dust loadings over a small geographical area (Evan et al., 2015).

The Bodélé Depression is located within the palaeolake Megachad basin which, during its mid-Holocene high-stand, reached 360,000 km^2 in size (Figure 3) and 170 m in depth (Drake and Bristow, 2006). Modern day Lake Chad is located within the southern part of the palaeolake basin and covers only ~ 5% of its mid-Holocene area (Bristow et al., 2018) (Figure 3). The Bodélé Depression lies within the northern part of the basin and remains dry today, separated from modern-day Lake Chad by a 285-m-high sill. During humid intervals in the past when lake levels exceeded 285 m, water flowed from Lake Chad in the south to the Bodélé Depression in the north via a palaeoriver system known as the Bahr El Gazel (Figure

3). Today, exposed diatomites in the Bahr El Gazel also act as an important dust source, contributing ~10% of North African winter dust (Bakker et al., 2019). To the east of the Bodélé Depression, a palaeoriver system originating in the Ennedi, Wadi Fira, Quaddai and Sila provinces feeds into palaeolake Megachad. From north to south, these rivers are locally known as Ouadi Archei, Ouadi Chili, Ouadi Oum Hadjer, Ouadi Haouach, Ouadi Yedinga, Ouadi Haddad and Ouadi Enne, and hereafter collectively referred to as the Eastern palaeorivers. A palaeoriver system originating in the Tibesti mountains feeds in from the north west, of which the main rivers are known as Enneri Ké and Enneri Modragué. Today the alluvial deposits that remain from all of these palaeoriver systems are important dust sources.

3.2 PSA geochemical fingerprints

3.2.1 Three geochemically distinct PSAs

Regional bedrock geology can have a major impact on the isotopic signature of aeolian sediments. The three main dust source regions that we define in Figure 2 are located within differing geological settings (Begg et al., 2009; Van Hinsbergen et al., 2011). The Western source region is strongly influenced by the West African Craton where bedrocks are of Paleoproterozoic age. The Central source region is characterized by younger basement rocks of Neoproterozoic age, and the Saharan Metacraton. The Eastern source region is much younger geologically and characterised by outcropping basic volcanic rocks. However, surface sediments from dust generation hotspots may have already been transported thousands of kilometres from their bedrock sources by the action of winds or rivers, which can result in a smoothing of spatial heterogeneities in bedrock geology (Reynolds et al., 2006).

Here, we combine new and existing data from known dust-producing substrates to quantify the isotopic signature of material emitted from each of our three newly outlined PSAs. The data set that we present includes samples from locations with a wide range of dust source activation frequencies (Figure 4A, section 2.4). The extent to which the locations with the highest DSAF have been sampled is variable by PSA (Figure 4B-D) and some geographical regions of data sparsity are unavoidable (Figure 2A). Least well sampled is the Western PSA, especially in its central and south-eastern regions where political unrest has limited ground-access in recent years. Nevertheless, there is reasonable isotopic agreement between our weighted mean ε_{Nd} signature for the Western PSA and the ε_{Nd} of airborne trap samples of dust collected on the Senegalese coast, which was backtracked to Mauritania, Western Sahara and the hotspot between Mali, Niger and Algeria (Skonieczny et al., 2013), all within our Western PSA. This similarity suggests that our data provide a good first approximation of the Western PSA (Supp. Figure 1) but the mismatch in $^{87}Sr/^{86}Sr$ (approximately 0.01, Supp. Figure 1) suggests that more coverage is needed.

The data that we present indicate a clear isotopic distinction between the three PSAs (Table 2 & Figure 5). The Western source region has the most radiogenic weighted mean $^{87}Sr/^{86}Sr$ signature (0.72788 ± 0.00520 (1sd)) and the most unradiogenic ε_{Nd} signature (-14.79 ± 2.16 (1sd)). The Eastern source region has a weighted mean $^{87}Sr/^{86}Sr$ of 0.70580 ± 0.00142 (1sd) and ε_{Nd} of -1.34 ± 2.46 (1sd). The Central source region has a weighted mean $^{87}Sr/^{86}Sr$ of 0.71863 ± 0.00530 (1sd) and a weighted mean ε_{Nd} of -9.96 ± 3.85 (1sd), but is highly heterogeneous and is therefore discussed in detail in a separate section below (Section 3.2.2). These broad trends from west to east in $^{87}Sr/^{86}Sr$ (to less radiogenic values) and ε_{Nd}

(to more radiogenic values) are consistent with large-scale North African bedrock geology which gets progressively younger from west to east (Begg et al., 2009; Van Hinsbergen et al., 2011). This result shows that the mixing effects of dust transportation and deposition across North African have a subordinate influence on regional isotope composition compared to underlying geology at a continental scale. Compared to previous PSA isotopic definitions (Abouchami et al., 2013; Pourmand et al., 2014; Scheuvens et al., 2013), weighting our data by DSAF leads to less overlap between the isotopic signatures of our new PSAs, especially between the Western and Central PSAs, permitting effective identification of source areas from downwind sample analyses.

The Western source region is most active during boreal summer (Schepanski et al., 2007) and dominates dust transport to the nearshore eastern subtropical Atlantic (Engelstaedter and Washington, 2007; Meng et al., 2017). Data sparsity for the Western PSA are currently limiting but based on the large-scale bedrock geology (Begg et al., 2009) we anticipate that, as more data become available, it may be possible to distinguish Western Sahara and Mauritania as separate PSAs. Nevertheless, even based on the data available, the Western source region appears geochemically distinct from the other two North African PSAs (Figure 5). A key priority for future sampling is the high dust emission frequency region where the borders of Algeria, Mali and Niger meet. Evan et al., (2015) report that this region is responsible for producing up to 13% of North African dust emissions, making it the second largest dust source on the continent, yet to our knowledge, no radiogenic isotope data currently exist for this region (Figure 2A).

In the Eastern source region, our new data from alluvial sediments in the Nubian Desert align with existing data from Sudan. Compiled new and existing data (Figure 5) range from -7.5 to +3.2 in ε_{Nd}, and from 0.70405 to 0.71691 in $^{87}Sr/^{86}Sr$. Of our three newly defined PSAs, this Eastern region has the most radiogenic Nd and non-radiogenic Sr isotope signatures, resulting from deposits left by palaeorivers draining young rocks. Data from the Eastern source region are extremely consistent, especially in Sr. This region produces dust year-round, but production is greatest during boreal summer (Engelstaedter and Washington, 2007; Schepanski et al., 2007). Dust is primarily transported from the Eastern source region via north-westerly winds to the Arabian Peninsula and the northern Arabian Sea. Sudan is historically under-appreciated as a dust source, however, Bakker et al. (2019) show that this is an important region of dust production, generating approximately 7.5% of North African wintertime dust. Our isotopic characterisation of high dust-producing substrates such as desiccated river beds will allow an improved definition of the contribution of this North African dust source to the Arabian Sea. To better understand the significance of this eastward dust flux from northeast African over geological timescales, further work is required to isotopically fingerprint other potential dust sources to the Arabian Sea, particularly over the Arabian Peninsula and eastern Horn of Africa.

3.2.2 Isotopic characterisation of the Bodélé Depression and surrounding area

The Central source region has the highest dust production of our three PSAs and includes Earth's greatest single dust source, the Bodélé Depression. It is particularly important to fingerprint the contribution of the Bodélé Depression to North African dust plumes to better understand its role in fertilising the Amazon rainforest and modifying the global radiative budget. Yet that task has been a challenging one because, while some isotope data are available from the wider region, the Central PSA is highly heterogeneous, with a broad range of isotopic signatures (ε_{Nd} values between -16.3 and -2.42 and $^{87}Sr/^{86}Sr$ between 0.70968 and 0.73181, Figure 5) (Abouchami et al., 2013; Gross et al., 2016; Grousset and Biscaye, 2005; Kumar et al., 2014), and very few data have been published from within the Bodélé Depression itself. Here, we present new radiogenic data from the Bodélé Depression and the surrounding river systems to better understand the origin and signature of the dust exported from this region.

Our data from the region immediately surrounding the Bodélé Depression show extreme isotopic heterogeneities, particularly in ε_{Nd}, that have not previously been documented (Figures 5 & 6). Palaeohydrological reconstructions (Drake et al., 2011) indicate that, during African humid periods of the last glacial cycle, the Bodélé Depression was fed by rivers flowing from three different areas: the Tibesti Mountains, the region surrounding the Ennedi Mountains (Eastern palaeorivers) and modern Lake Chad via the Bahr el Gazel (Figures 3 and 6A). These three hinterlands have different geology and therefore provide lithogenic material with very different isotopic signatures to the Bodélé Depression, as well as acting directly as dust sources themselves (Bakker et al., 2019).

Samples taken north of the Bodélé Depression, in the Angamma Delta, near to the Tibesti Mountains have highly radiogenic Nd and non-radiogenic Sr compositions (ε_{Nd}: -3.4 to -2.4, $^{87}Sr/^{86}Sr$: 0.70968 to 0.71050). This is the first time that dust sources with such a radiogenic Nd signature have been reported in the central Sahel/Sahara. The Tibesti Massif is volcanic and younger than the surrounding basement. Isotopic measurements on samples from Wadi Yebigue, a granitic pluton from within the Tibesti Massif, range from -6.01 to -1.83 in

ε_{Nd} (back-calculated from a crystallisation age of 550 Ma), and 0.7254 to 0.87549 in $^{87}Sr/^{86}Sr$ (Suayah et al., 2006). This provides a possible endmember from which alluvial sediments sampled in the Angamma Delta, draining the Tibesti Mountains, have derived. While the ε_{Nd} data fit well, the $^{87}Sr/^{86}Sr$ data of the Wadi Yebigue are far more radiogenic than those measured in the Angamma Delta. It is unlikely that the Wadi Yebigue pluton is representative of the entire catchment surrounding the Tibesti Massif, so more data are required to fully constrain the isotopic endmember leading to highly radiogenic ε_{Nd} values at the northern extremity of the Bodélé Depression.

Samples taken within the Eastern palaeo-river system, to the south of the Ennedi Mountains, have very different signatures to those of the Angamma Delta, with ε_{Nd} values between -14.2 and -11.6, and $^{87}Sr/^{86}Sr$ between 0.71530 and 0.72063. There are no isotope data available from the highland source of these rivers, but they are likely well-approximated by the data that we present from the Eastern palaeoriver sediments. Furthermore, the consistency of the Eastern palaeoriver data in Nd-Sr space, across a large geographic area covering several branches of the palaeo river system, implies that this system stemmed from a single source, in this case a Precambrian basement with Lower Palaeozoic sandstone (Wolff, 1964).

Samples taken from south of the Bodélé Depression, in the Bahr El Gazel palaeoriver system, exhibit highly radiogenic $^{87}Sr/^{86}Sr$ (0.72054 to 0.73181) and comparatively unradiogenic ε_{Nd} (-13.1 to -9.9). Reconstructions (Drake and Breeze, 2016) suggest that the Bahr El Gazel flowed out of Lake Chad when water levels surpassed the 285m sill, carrying sediment north into the Bodélé Depression. While samples from modern day Lake Chad are very limited, the similarity between the signatures of Lake Chad (ε_{Nd} of -12.7; Grousset and Biscaye, (2005)) and the Bahr El Gazel support this reconstruction.

Samples taken from within the Bodélé Depression itself have ε_{Nd} values between -11.9 and -7.4, and $^{87}Sr/^{86}Sr$ between 0.71523 and 0.71858. This is more radiogenic in ε_{Nd} and more narrowly defined in $^{87}Sr/^{86}Sr$ than reported previously (Abouchami et al., 2013; Scheuvens et al., 2013). We find that the isotopic signature of the Bodélé Depression can be explained by mixing of the three isotopically distinct riverine endmembers (the Bahr El Gazel, Tibesti and Eastern palaeorivers) (Figure 6B), in line with palaeohydrological reconstructions (Drake et al., 2011). Several smaller palaeolakes (Figure 6B) located to the west of the Bodélé Depression exhibit isotopic values similar to it, indicating that they were also fed by a mixture of the Bahr El Gazel, and palaeorivers draining the Tibesti Mountains and Eastern highlands. This contrasts with samples from modern lakes south of the Bodélé Depression (Lake Fitri), which exhibit a different isotopic signature ($^{87}Sr/^{86}Sr$ of 0.72423, ε_{Nd} of -16.3 to -13.6, this study), indicating a different source of sediment to this locality.

The disparity between our isotopic definition of the Bodélé Depression and those previously reported (Abouchami et al., 2013; Scheuvens et al., 2013) primarily stems from differing naming protocols for samples from the Bodélé Depression and its surrounding geomorphological features. While several studies (Abouchami et al., 2013; Gross et al., 2016; Grousset and Biscaye, 2005; Kumar et al., 2014) sampled close to the Bodélé Depression, until our work (this study), data from samples taken from within the palaeolake basin were extremely sparse (Figure 6A). Many samples taken from nearby regions, several close to modern day Lake Chad, and others within the Bahr El Gazel, were originally classified as “Bodélé” samples. Our data show that the term “Bodélé Depression” should be reserved for the diatomite-rich palaeolake basin in the north of palaeolake Megachad. Abouchami et al. (2013) distinguished between Si-rich and Ca-rich components of Chadian dust sources, however the variability seen in Sr-Nd isotope space in their data is modest in comparison to that documented for our sample set, most likely because their samples spanned a smaller geographical range. Their Si-rich source is the diatomite-rich alluvial deposits of the Bahr El Gazel whereas their Ca-rich source is a palaeolake bed neighbouring the Bodélé Depression where the isotopic signature is very similar to that of our Eastern palaeoriver endmember, indicating its primary source.

The Central PSA is an active dust source all year round, but most important during boreal winter when the Western and Eastern PSAs become distinctly less active than in summer (Ben-Ami et al., 2012; Schepanski et al., 2007). Dust from this region is transported via the Harmattan Trade winds towards the Eastern Equatorial Atlantic and onwards to South America (Meng et al., 2017). North African dust is hypothesised to have a significant fertilising effect on the nutrient-poor soils of the Amazon rainforest (Yu et al., 2015). Our radiogenic isotopic data will help to assess the role of the Bodélé Depression in this process. Comparison of the isotopic signature of dust accumulating in marine sediment cores off the North West African margin to the central dust source will help to evaluate the relative contributions of dust sources and also shed light on the palaeo-history of the Bodélé Depression and palaeolake Megachad.

3.2.3 Effect of grain size on Sr and Nd isotope signature

Sr and Nd isotope systems provide conservative fingerprints of PSAs over long geological timescales and geographical distances. Nd isotopes are not considered to fractionate isotopically during weathering or transport, so the isotopic signature is passed directly from source rock to sink (Feng et al., 2009; Goldstein et al., 1984; Meyer et al., 2011). Conversely, Sr isotopes are suggested to fractionate during weathering and transport processes, with several studies demonstrating increased $^{87}Sr/^{86}Sr$ with increasing weathering intensity and with decreasing grain size (Feng et al., 2009; Grousset et al., 1992; Meyer et al., 2011).

We analysed five samples from the Central Source region across four different size fractions: 63 - 45 µm, 45 - 32 µm, < 32 µm and <10 µm. Overall, the magnitude of variation evidenced in our samples due to changing grain size is small in comparison to isotopic differences between the newly defined PSAs. Yet, variability is documented in both Nd and Sr isotope composition with grain size (Figure 7), and the magnitude and sign of these signals varies by location.

We find minimal grain-size variability in ε_{Nd} and $^{87}Sr/^{86}Sr$ in the sample from the Bahr El Gazel (BEG2), and the sample from the Bodélé Depression (BOD1) shows only a modest increase in radiogenic ε_{Nd} values with decreasing grain size, with no significant change in $^{87}Sr/^{86}Sr$. Conversely, three samples from the Eastern palaeorivers (EN4, EN13, EN14) show more pronounced grain size variation but with no consistent sign of change in either ε_{Nd} and $^{87}Sr/^{86}Sr$. This result suggests that the sediments proximal to the Ennedi Highlands are immature and poorly sorted when compared to sediments in the Bodélé Depression and the Bahr El Gazel.

Although there appears to be a wide acceptance in the literature (e.g. Feng et al., 2009; Grousset et al., 1992; Meyer et al., 2011; Újvári et al., 2018) that Nd isotopes do not fractionate with increased weathering and decreasing grain size, this perception stems from data reported on only one or two samples (Goldstein et al., 1984; Grousset et al., 1998). Our results show that significant grain size effects can occur in both strontium and neodymium isotopes in chemically immature sediments, therefore highlighting the need for more detailed research to better understand the extent to which sediment grain size can influence Nd isotopes in different geological settings, and how the isotopic signature of sediment evolves with transport history. It is important to isolate one consistent grain size fraction when analysing sediment for Sr and Nd isotope composition. Alternatively, work is needed to establish the scale of isotopic variability with varying grain size in the study region, especially when working with chemically immature sediments.

4. Conclusions

We identify three readily discernible North African dust PSAs, defined geographically using a high resolution Dust Source Activation Frequency map (Schepanski et al., 2012) and characterised geochemically using Sr and Nd isotopes (Figure 8). We weight geochemical data by locality dust source activation to produce representative estimates of the isotopic signature of emitted dust. The Western Source area is characterised by the most unradiogenic Nd and most radiogenic Sr signature, reflecting the old cratonic bedrock geology. More work is needed to validate the isotopic signatures of the major sites of dust activation in this understudied region, particularly targeting the alluvial sediments in Southern Algeria, Eastern Mali and North West Niger. The historically underestimated Eastern source region is geochemically well defined, and shows radiogenic Nd and non-radiogenic Sr values. The Central dust source region is highly geochemically heterogenous. We find that the isotopic signature of the Bodélé Depression is the result of mixing from three geochemically distinct palaeo-river systems: the Bahr El Gazel, and rivers draining from the Tibesti Mountains and Eastern highlands. We identify a strong grain-size effect on the Nd and Sr isotopic signature of the immature Eastern palaeoriver sediments in particular, highlighting the need to determine the nature and importance of grain size effects in a specific study region.

Our new characterisation of North African dust source regions provides distinct fingerprints to facilitate analysis of downwind dust trap samples, and help determine the role of specific dust sources in fertilising North Atlantic surface waters and the Amazon rainforest during different seasons (Kumar et al., 2018; Yu et al., 2015). Our data set also provides a framework to establish changes in the contribution of different continental dust source regions on geological timescales, through comparison to downcore records preserved in marine and lacustrine sediments, and to help to shed new light on the history of palaeolake Megachad. On finer spatial scales, the improved understanding of the isotopic signature of the region surrounding the Bodélé Depression corroborates palaeo river drainage reconstructions (Drake et al., 2011). Our study demonstrates the importance of understanding the geomorphological context of dust source regions in order to accurately define them geochemically, and highlights where future sampling should be focussed to further improve the geochemical characterisation of North African dust sources.

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Figure Captions:

Figure 1: Previous analyses of North African preferential dust source areas (PSAs). 1 ° x 1 ° maps of North African dust sources from March 2006 to February 2010, based on A) OMI aerosol index (Map adapted from Schepanski et al., 2012; frequency-based remote sensing method) B) MSG SEVIRI IR dust index (Adapted from Schepanski, *et al.*, 2012; employs backtracking method therefore removing dust transport bias). C) Geographical definition of North African Potential Source Areas (PSAs) of (Adapted from Scheuvens et al., 2013) based on a variety of remote sensing techniques that conflate dust transport and emission. D) Geochemical characterisation of North African PSAs, as defined in Figure 1C, in Nd-Sr isotope space (PSAs 1 - 4 & 6 as defined by Scheuvens et al., 2013, PSA 5 from Abouchami et al., 2013).

Figure 2: A new analysis revealing three North African preferential dust source areas (PSAs, this study). A) Annual dust source activation frequency (DSAF) (Schepanski et al., 2012) and location of existing (white circles) and new (red circles, this study) dust source samples with Sr and/or Nd isotope data (published data from Abouchami et al., 2013; Gross et al., 2016; Grousset et al., 1998; Kumar et al., 2014; Padoan et al., 2011; Zhao et al., 2018) . B) DSAF in boreal summer (JJA) (Schepanski et al., 2007). C) Three new PSAs (this study) based on the data in (A) and the topographic highs used to separate them. Coloured shading denotes annual DSAF > 5 % (bold) and DSAF < 5% (pale).

Figure 3: Location, areal extent and palaeohydrology of palaeolake Megachad (adapted from Armitage et al., 2015).

Figure 4: Distribution of dust source activation frequency (DSAF) and isotopic data coverage. Histograms showing the total area (i.e. number of 1 ° x 1 ° squares) covered by each DSAF bracket (bold colours, Schepanski et al., 2012) and the corresponding area characterised by isotopic data (pale bars, this study) for A) the whole of North Africa and for the B) Western, C) Central and D) Eastern PSAs. Percentage coverage shown above each bar.

Figure 5: Isotopic composition of our three North African PSAs. New data (squares, this study) and published data (circles) (Abouchami et al., 2013; Gross et al., 2016; Grousset et al., 1998; Kumar et al., 2014; Padoan et al., 2011; Zhao et al., 2018) from North African dust source regions. Size corresponds to annual DSAF (Schepanski et al., 2012). Crosses denote mean isotopic values for each source region weighted by annual DSAF +/- one weighted standard deviation. Where only ε_{Nd} or $^{87}Sr/^{86}Sr$ data is available, sample is not plotted, but the available data still contribute to the weighted PSA mean.

Figure 6: The Chadian region of the central preferential dust source area. A) Sampling locations for new data (squares, this study) and published data (circles, Abouchami et al., 2013; Gross et al., 2016; Grousset and Biscaye, 2005; Kumar et al., 2014) from Chad, in the central PSA. Satellite image taken from Google Earth, overlain with palaeo river reconstructions (Drake and Breeze, 2016). B) Sr and Nd isotope data from Chad, in the Central PSA. Shading highlights main contributors determining the isotopic signature of the Bodélé Depression (red): the Tibesti (orange), Eastern (green) and Bahr El Gazel (yellow) palaeorivers. Smaller palaeolakes (lilac) are also likely fed by a mixture of these palaeorivers.

Figure 7: Effect of grain size on Sr and Nd isotopes within the central dust source region. Symbol size denotes grain size (63 - 45 µm, 45 – 32 µm, <32 µm and <10 µm). One sample from each of the

Bodélé (BOD1, red) and Bahr El Gazel (BEG2, yellow), and three from the Eastern palaeorivers (EN4, EN13, EN14, green) were analysed. Sample locations shown in Figure 6. Black bars show 2 standard error (often smaller than the symbol). $^{87}Sr/^{86}Sr$ for sample BEG2 < 10 µm did not successfully run, but the ε_{Nd} is -12.26.

Figure 8: $^{87}Sr/^{86}Sr$ (top) and ε_{Nd} (bottom) isotope composition of our three North African PSAs (mean values, +/- 1sd, weighted by activation frequency of the source. Dust sources (1 ° x 1 °) with activation frequency > 5% (Schepanski et al., 2012) shown in bold colours, < 5% in pale colours.