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Three North African Dust Source Areas and their Geochemical Fingerprint

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22 Abstract

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

44

45 Keywords:

46 North Africa, dust source, Bodélé Depression, radiogenic isotopes, ϵNd , $87\text{Sr}/86\text{Sr}$

47

48 **1. Introduction**

49 Atmospheric dust is a key component of Earth's climate system, influencing the global
50 radiative budget both directly by controlling absorption and scattering of solar radiation,
51 and indirectly by stimulating cloud condensation and cover (Thompson et al., 2019). Today,
52 North Africa exports more dust to the atmosphere than any region on Earth with an
53 estimated annual production of 170 to 1600 Tg yr⁻¹ (Engelstaedter et al., 2006), over half the
54 total global flux (Ginoux et al., 2012). In some regions, these loadings constitute a serious
55 risk to human health; high concentrations of atmospheric particulate matter with an
56 aerodynamic diameter of less than 2.5 μm (PM_{2.5}) are estimated to have led to between
57 194,000 and 709,000 infant deaths in North Africa in 2015 alone (Heft-Neal et al., 2018).
58 Once lofted into the atmosphere, North African dust can be carried thousands of
59 kilometres, primarily to the west, in plumes that are visible from space and are suggested to
60 transport Fe and other fertilizing micronutrients to sites of primary production in the
61 surface waters of the North Atlantic Ocean (Jickells, 2005) and the rainforests of the
62 Amazonian Basin (Koren et al., 2006; Yu et al., 2015). The accumulation of this dust in
63 marine or lacustrine sediments also provides a way to investigate past changes in climate
64 variability on geological timescales (Cole et al., 2009; Grousset et al., 1998; Tiedemann et
65 al., 1994).

66

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

70 Schepanski et al., 2012) and establishing that there are two main transport pathways
71 westwards, a “northern route” towards the Caribbean which dominates during boreal
72 summer, and a “southern route” towards the Amazon during winter (Engelstaedter et al.,
73 2006; Meng et al., 2017). Yet despite these recent advances, major uncertainties remain in
74 our knowledge of North African dust-producing source regions and our ability to fingerprint
75 their contribution to these export routes (Bakker et al., 2019; Formenti et al., 2011;
76 Scheuvens et al., 2013). This limited knowledge base hinders the development of a
77 comprehensive source-to-sink understanding and adds uncertainty to climatic
78 interpretations that rely on the accumulation of dust in geological archives.

79

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

94

95 Once defined geographically by remote sensing analysis, preferential source areas of dust
96 generation (PSAs) are characterised geochemically to permit dust provenance studies (e.g.,
97 Figure 1C & 1D). The primary tools that have been used in this endeavour are the radiogenic
98 isotope analysis (mainly ϵ_{Nd} and $^{87}Sr/^{86}Sr$) of bedrock and associated lithogenic sediments.
99 Yet, there are two main problems associated with existing interpretations of this kind. First,
100 previous landmark studies noted the disparity in dust sources identified by the different
101 remote sensing techniques outlined above and combined the results of the these
102 approaches to identify six PSAs in North Africa (Fig. 1C, Scheuven et al., 2013). However,
103 this approach incorporates residual transport bias in dust emission estimates (Schepanski et
104 al., 2012, 2007). Second, a large fraction of the Sr and Nd isotope data set currently
105 available does not come from active deflating dust source regions (Abouchami et al., 2013;
106 Blanchet, 2019; Gross et al., 2016; Scheuven et al., 2013; Zhao et al., 2018). Dust in North
107 Africa is mostly derived from distinct palaeolake- and alluvial- deposits, which are located in
108 the foothills of Saharan mountain ranges (Bakker et al., 2019; Schepanski et al., 2009).
109 Surrounding bedrock and sand sediments contain very limited fine-grained material, which
110 inhibits dust production from these surfaces (Bullard et al., 2011). While samples from
111 bedrock and sand deposits in the vicinity of dust sources are useful for investigating local
112 sediment dynamics, they are not well-suited to fingerprinting the signature of dust
113 transported over thousands of kilometres. Moreover, existing radiogenic isotope data have
114 been generated on differing grain size fractions, ranging from bulk sediment measurements
115 to $< 2 \mu m$. While the Nd isotope composition of aeolian dust is considered insensitive to
116 grain size, Sr isotope data are widely suggested to be susceptible to a substantial grain-size
117 dependent fractionation (with an increase in $^{87}Sr/^{86}Sr$ of ~ 0.01 in the $< 2 \mu m$ fraction

118 compared to the >50 μm fraction, (Feng et al., 2009). Here we address these issues in
119 identifying the geochemical signature of exported dust by presenting new PSAs,
120 geographically defined based on the dust source activation frequency (DSAF) map of
121 Schepanski et al. (2012) and isotopically characterised using our new Sr and Nd data
122 combined with existing datasets from known dust sources. Our overarching aim is to
123 improve the reliability by which dust deposits can be geochemically fingerprinted to their
124 distant source regions.

125

126 **2. Materials and Methods**

127 2.1 Geographical definition of PSAs using remotely sensed data

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

137

138 2.2 Dust samples

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

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

144

145 2.3 Radiogenic Isotope analysis

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

154

155 Radiogenic isotope analyses were carried out in a clean chemistry laboratory at the
156 University of Southampton Waterfront Campus, NOCS. Approximately 100 mg of
157 decarbonated sample was digested overnight using a HF-HNO₃-HClO₄ acid mixture at 130 °C.
158 The digestion blanks (i.e. acid residue after full digestion procedure with no sample) were
159 below detection level (level of detection was 0.051 ppm & 0.266 ppm for Nd and Sr
160 respectively, calculated by $\bar{x}_{\text{blank}} + 3\sigma$). Nd and Sr were isolated using chromatographic
161 column separation (adapted from Bayon et al. (2002)). Nd was isolated using a cation
162 column (Bio-Rad AG-50W-X8 resin) followed by a reverse phase column (Ln-spec resin) (Pin
163 and Zalduegui, 1997). Sr was isolated using Sr-spec resin. The total column blanks (i.e. when

164 blank acid is run through the column procedure) were negligible (50 pg and 30 pg)
 165 compared to the total amounts analysed (1 µg and 200 ng) for Sr and Nd respectively.

166

167 Nd-isotope ratios were measured using a Thermo Scientific Neptune multi-collector
 168 inductively coupled plasma mass spectrometer (MC-ICP-MS). $^{143}\text{Nd}/^{144}\text{Nd}$ compositions
 169 were corrected following the method of Vance and Thirlwall (2002) through adjustment to a
 170 $^{146}\text{Nd}/^{144}\text{Nd}$ ratio of 0.7219 and a secondary normalisation to $^{142}\text{Nd}/^{144}\text{Nd} = 1.141876$.

171 Results for the JNdi-1 reference standard (Tanaka et al., 2000) measured as an unknown
 172 were 0.512115 with an external reproducibility of the ± 0.000006 (2SD) across 6 analysis
 173 sessions over two years. For convenience $^{143}\text{Nd}/^{144}\text{Nd}$ is reported here in epsilon notation
 174 (ϵ_{Nd}), where $^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}}$ represents the Chondrite Uniform Reservoir value of 0.512638
 175 (Jacobsen and Wasserburg, 1980):

176

$$177 \quad \epsilon_{\text{Nd}} = \left[\frac{^{143}\text{Nd}/^{144}\text{Nd}_{\text{sample}}}{^{143}\text{Nd}/^{144}\text{Nd}_{\text{CHUR}}} - 1 \right] \times 10^4$$

178

179 After column separation, the Sr fraction was dried down and loaded onto an outgassed
 180 tantalum filament with 1µl of a tantalum activator solution. The samples were analysed on a
 181 ThermoScientific Triton Plus Thermal Ionisation Mass Spectrometer (TIMS) using a multi-
 182 dynamic procedure with an ^{88}Sr intensity of 2V. Fractionation was corrected using an
 183 exponential correction normalised to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. NIST987 (Yobregat et al., 2017) was
 184 run as a standard on each turret alongside our samples and was measured at $0.710241 \pm$
 185 0.000013 (2SD) on 4 analyses. The long-term average for NIST987 on this instrument is

186 0.710243 ± 0.000020 (2SD) from 464 analyses. Rock standard JB-1a was run through the
 187 same digestion and chemical separation procedures to give $^{87}\text{Sr}/^{86}\text{Sr}$ and ϵ_{Nd} values of
 188 0.704112 ± 0.000014 (2SE) and 2.97 ± 0.17 (2SD) respectively (within accepted values,
 189 GeoReM: Jochum et al., 2005)).

190

191 2.4 Generating geochemically representative PSA compositions

192 Our approach to geochemically defining PSA compositions differs in two important ways
 193 from the previous studies. First, our PSA compositions are defined using only data from dust
 194 producing substrates. We combined our new data with published data from dust-producing
 195 substrates such as lacustrine and riverine sediments and soils, but excluded data from
 196 bedrock and aerosol samples. Second, we applied a dust source activity weighting to the
 197 isotope data. In the past, when geographically defined using remote sensing methods that
 198 conflate dust deflation and transport, PSA signatures were defined using the ranges of
 199 isotopic data generated on all samples that fall within those PSA regions, defining a “box” or
 200 field in Sr-Nd space. While this simple method is a sensible first order approach, it has the
 201 important limitation that equal weighting is given to all isotopic measurements, regardless
 202 of the contribution of the substrate to atmospheric dust loading. We applied a weighting to
 203 the isotope data according to annual DSAF (in each case the DSAF value from the
 204 corresponding 1 ° x 1 ° square was used as a multiplier) (Schepanski et al., 2012). To reduce
 205 bias caused by uneven sampling, where there are multiple samples available within one 1 ° x
 206 1 ° square, the isotope data were averaged and the resulting value was weighted by DSAF.
 207 The weighted mean isotopic signature for each PSA is calculated by:

$$208 \quad \bar{x}_{PSA} = \frac{\sum(x_i \times w_i)}{\sum w_i}$$

210 *where x_i = sample isotope signature, w_i = DSAF*

209

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

$$212 \quad st. dev_{PSA} = \sqrt{\frac{\sum(w_i (x_i - \bar{x}_{PSA})^2)}{\frac{(N-1)\sum w_i}{N}}}$$

213 *where x_i = sample isotope signature, w_i = DSAF, N = no. of samples*

214 We also calculate the percentage coverage for each PSA (i.e. the proportion of $1^\circ \times 1^\circ$
215 squares of DSAF > 0 % that have an isotopic value assigned).

216

217 **3. Results and Discussion**

218 3.1 PSA geomorphology

219 The distribution of DSAF in North Africa changes seasonally. For most of the year,
220 particularly in boreal summer when dust export is greatest (Engelstaedter and Washington,
221 2007) (Figure 2B), there are three main geographically distinct hotspots of dust source
222 activation (i) West Sahara/Sahel (Southern Algeria, North East Mali, West Niger, North
223 Mauritania), (ii) Central Sahara/Sahel (Chad) and (iii) East Sahara/Sahel (North Eastern
224 Sudan) (Schepanski et al., 2007), hereafter the Western, Central and Eastern source regions
225 (Figure 2C). These hotspots are separated by topographic highs; the Hoggar and Air
226 mountains between the Western and Central source regions, and the Ennedi Mountains
227 between the Central and Eastern sources (Figure 2C). Furthermore, the three dust source
228 regions are sufficiently distinct geographically to mean that they lie within separate palaeo
229 river catchment basins (Drake et al., 2011).

230

231 The most active dust sources in North Africa are desiccated river and lake beds (Prospero et
232 al., 2002), contributing approximately 36% and 64% of total North African winter dust
233 respectively (Bakker et al., 2019). The Western source region (Figure 2) is dominated by dust
234 production from alluvial deposits and palaeolakes in the deserts that surround the Aïr and
235 Adrar Iforas Mountains spanning Algeria, Mali and Niger. Similarly, in the Eastern source
236 region (Figure 2), dust derives largely from alluvial deposits in the Nubian desert (Bakker et
237 al., 2019). The Central source region is the most active of the PSAs, and its main contributor
238 is the Bodélé Depression. The Bodélé Depression is often described as the “dustiest place on
239 Earth” and is estimated to contribute 50% - 64% of all North African dust (Bakker et al.,
240 2019; Bristow et al., 2009; Engelstaedter et al., 2006; Evan et al., 2015) because of the
241 combination of strong near surface winds funnelled between the nearby Tibesti and Ennedi
242 mountains, and the large reservoir of easily deflatable, low density diatomite-rich sediment
243 (Bristow et al., 2009; Washington et al., 2006). DSAF data may even underestimate the
244 importance of this region as a dust source due to its extremely high dust loadings over a
245 small geographical area (Evan et al., 2015).

246

247 The Bodélé Depression is located within the palaeolake Megachad basin which, during its
248 mid-Holocene high-stand, reached 360,000 km² in size (Figure 3) and 170 m in depth (Drake
249 and Bristow, 2006). Modern day Lake Chad is located within the southern part of the
250 palaeolake basin and covers only ~ 5% of its mid-Holocene area (Bristow et al., 2018) (Figure
251 3). The Bodélé Depression lies within the northern part of the basin and remains dry today,
252 separated from modern-day Lake Chad by a 285-m-high sill. During humid intervals in the
253 past when lake levels exceeded 285 m, water flowed from Lake Chad in the south to the
254 Bodélé Depression in the north via a palaeoriver system known as the Bahr El Gazel (Figure

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

265

266 3.2 PSA geochemical fingerprints

267 3.2.1 Three geochemically distinct PSAs

268 Regional bedrock geology can have a major impact on the isotopic signature of aeolian
269 sediments. The three main dust source regions that we define in Figure 2 are located within
270 differing geological settings (Begg et al., 2009; Van Hinsbergen et al., 2011). The Western
271 source region is strongly influenced by the West African Craton where bedrocks are of
272 Paleoproterozoic age. The Central source region is characterized by younger basement rocks
273 of Neoproterozoic age, and the Saharan Metacraton. The Eastern source region is much
274 younger geologically and characterised by outcropping basic volcanic rocks. However,
275 surface sediments from dust generation hotspots may have already been transported
276 thousands of kilometres from their bedrock sources by the action of winds or rivers, which
277 can result in a smoothing of spatial heterogeneities in bedrock geology (Reynolds et al.,
278 2006).

279

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

294

295 The data that we present indicate a clear isotopic distinction between the three PSAs (Table
296 2 & Figure 5). The Western source region has the most radiogenic weighted mean $^{87}Sr/^{86}Sr$
297 signature (0.72788 ± 0.00520 (1sd)) and the most unradiogenic ϵ_{Nd} signature (-14.79 ± 2.16
298 (1sd)). The Eastern source region has a weighted mean $^{87}Sr/^{86}Sr$ of 0.70580 ± 0.00142 (1sd)
299 and ϵ_{Nd} of -1.34 ± 2.46 (1sd). The Central source region has a weighted mean $^{87}Sr/^{86}Sr$ of
300 0.71863 ± 0.00530 (1sd) and a weighted mean ϵ_{Nd} of -9.96 ± 3.85 (1sd), but is highly
301 heterogeneous and is therefore discussed in detail in a separate section below (Section
302 3.2.2). These broad trends from west to east in $^{87}Sr/^{86}Sr$ (to less radiogenic values) and ϵ_{Nd}

303 (to more radiogenic values) are consistent with large-scale North African bedrock geology
304 which gets progressively younger from west to east (Begg et al., 2009; Van Hinsbergen et al.,
305 2011). This result shows that the mixing effects of dust transportation and deposition across
306 North African have a subordinate influence on regional isotope composition compared to
307 underlying geology at a continental scale. Compared to previous PSA isotopic definitions
308 (Abouchami et al., 2013; Pourmand et al., 2014; Scheuvens et al., 2013), weighting our data
309 by DSAF leads to less overlap between the isotopic signatures of our new PSAs, especially
310 between the Western and Central PSAs, permitting effective identification of source areas
311 from downwind sample analyses.

312

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

325

326 In the Eastern source region, our new data from alluvial sediments in the Nubian Desert
327 align with existing data from Sudan. Compiled new and existing data (Figure 5) range from -
328 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
329 PSAs, this Eastern region has the most radiogenic Nd and non-radiogenic Sr isotope
330 signatures, resulting from deposits left by palaeorivers draining young rocks. Data from the
331 Eastern source region are extremely consistent, especially in Sr. This region produces dust
332 year-round, but production is greatest during boreal summer (Engelstaedter and
333 Washington, 2007; Schepanski et al., 2007). Dust is primarily transported from the Eastern
334 source region via north-westerly winds to the Arabian Peninsula and the northern Arabian
335 Sea. Sudan is historically under-appreciated as a dust source, however, Bakker et al. (2019)
336 show that this is an important region of dust production, generating approximately 7.5% of
337 North African wintertime dust. Our isotopic characterisation of high dust-producing
338 substrates such as desiccated river beds will allow an improved definition of the
339 contribution of this North African dust source to the Arabian Sea. To better understand the
340 significance of this eastward dust flux from northeast African over geological timescales,
341 further work is required to isotopically fingerprint other potential dust sources to the
342 Arabian Sea, particularly over the Arabian Peninsula and eastern Horn of Africa.

343

344 3.2.2 Isotopic characterisation of the Bodélé Depression and surrounding area

345 The Central source region has the highest dust production of our three PSAs and includes
346 Earth's greatest single dust source, the Bodélé Depression. It is particularly important to
347 fingerprint the contribution of the Bodélé Depression to North African dust plumes to better
348 understand its role in fertilising the Amazon rainforest and modifying the global radiative
349 budget. Yet that task has been a challenging one because, while some isotope data are

350 available from the wider region, the Central PSA is highly heterogeneous, with a broad
351 range of isotopic signatures (ϵ_{Nd} values between -16.3 and -2.42 and $^{87}Sr/^{86}Sr$ between
352 0.70968 and 0.73181, Figure 5) (Abouchami et al., 2013; Gross et al., 2016; Grousset and
353 Biscaye, 2005; Kumar et al., 2014), and very few data have been published from within the
354 Bodélé Depression itself. Here, we present new radiogenic data from the Bodélé Depression
355 and the surrounding river systems to better understand the origin and signature of the dust
356 exported from this region.

357

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

367

368 Samples taken north of the Bodélé Depression, in the Angamma Delta, near to the Tibesti
369 Mountains have highly radiogenic Nd and non-radiogenic Sr compositions (ϵ_{Nd} : -3.4 to -2.4,
370 $^{87}Sr/^{86}Sr$: 0.70968 to 0.71050). This is the first time that dust sources with such a radiogenic
371 Nd signature have been reported in the central Sahel/Sahara. The Tibesti Massif is volcanic
372 and younger than the surrounding basement. Isotopic measurements on samples from
373 Wadi Yebigue, a granitic pluton from within the Tibesti Massif, range from -6.01 to -1.83 in

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

382

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

392

393 Samples taken from south of the Bodélé Depression, in the Bahr El Gazel palaeoriver
394 system, exhibit highly radiogenic $^{87}Sr/^{86}Sr$ (0.72054 to 0.73181) and comparatively
395 unradiogenic ϵ_{Nd} (-13.1 to -9.9). Reconstructions (Drake and Breeze, 2016) suggest that the
396 Bahr El Gazel flowed out of Lake Chad when water levels surpassed the 285m sill, carrying
397 sediment north into the Bodélé Depression. While samples from modern day Lake Chad are

398 very limited, the similarity between the signatures of Lake Chad (ϵ_{Nd} of -12.7; Grousset and
399 Biscaye, (2005)) and the Bahr El Gazel support this reconstruction.

400

401 Samples taken from within the Bodélé Depression itself have ϵ_{Nd} values between -11.9 and -
402 7.4, and $^{87}\text{Sr}/^{86}\text{Sr}$ between 0.71523 and 0.71858. This is more radiogenic in ϵ_{Nd} and more
403 narrowly defined in $^{87}\text{Sr}/^{86}\text{Sr}$ than reported previously (Abouchami et al., 2013; Scheuvens
404 et al., 2013). We find that the isotopic signature of the Bodélé Depression can be explained
405 by mixing of the three isotopically distinct riverine endmembers (the Bahr El Gazel, Tibesti
406 and Eastern palaeorivers) (Figure 6B), in line with palaeohydrological reconstructions (Drake
407 et al., 2011). Several smaller palaeolakes (Figure 6B) located to the west of the Bodélé
408 Depression exhibit isotopic values similar to it, indicating that they were also fed by a
409 mixture of the Bahr El Gazel, and palaeorivers draining the Tibesti Mountains and Eastern
410 highlands. This contrasts with samples from modern lakes south of the Bodélé Depression
411 (Lake Fitri), which exhibit a different isotopic signature ($^{87}\text{Sr}/^{86}\text{Sr}$ of 0.72423, ϵ_{Nd} of -16.3 to -
412 13.6, this study), indicating a different source of sediment to this locality.

413

414 The disparity between our isotopic definition of the Bodélé Depression and those previously
415 reported (Abouchami et al., 2013; Scheuvens et al., 2013) primarily stems from differing
416 naming protocols for samples from the Bodélé Depression and its surrounding
417 geomorphological features. While several studies (Abouchami et al., 2013; Gross et al.,
418 2016; Grousset and Biscaye, 2005; Kumar et al., 2014) sampled close to the Bodélé
419 Depression, until our work (this study), data from samples taken from within the palaeolake
420 basin were extremely sparse (Figure 6A). Many samples taken from nearby regions, several
421 close to modern day Lake Chad, and others within the Bahr El Gazel, were originally

422 classified as “Bodélé” samples. Our data show that the term “Bodélé Depression” should be
423 reserved for the diatomite-rich palaeolake basin in the north of palaeolake Megachad.
424 Abouchami et al. (2013) distinguished between Si-rich and Ca-rich components of Chadian
425 dust sources, however the variability seen in Sr-Nd isotope space in their data is modest in
426 comparison to that documented for our sample set, most likely because their samples
427 spanned a smaller geographical range. Their Si-rich source is the diatomite-rich alluvial
428 deposits of the Bahr El Gazel whereas their Ca-rich source is a palaeolake bed neighbouring
429 the Bodélé Depression where the isotopic signature is very similar to that of our Eastern
430 palaeoriver endmember, indicating its primary source.

431

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

443

444 3.2.3 Effect of grain size on Sr and Nd isotope signature

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

452

453 We analysed five samples from the Central Source region across four different size fractions:
454 63 - 45 μm , 45 - 32 μm , < 32 μm and <10 μm . Overall, the magnitude of variation evidenced
455 in our samples due to changing grain size is small in comparison to isotopic differences
456 between the newly defined PSAs. Yet, variability is documented in both Nd and Sr isotope
457 composition with grain size (Figure 7), and the magnitude and sign of these signals varies by
458 location.

459

460 We find minimal grain-size variability in ϵ_{Nd} and $^{87}\text{Sr}/^{86}\text{Sr}$ in the sample from the Bahr El
461 Gazel (BEG2), and the sample from the Bodélé Depression (BOD1) shows only a modest
462 increase in radiogenic ϵ_{Nd} values with decreasing grain size, with no significant change in
463 $^{87}\text{Sr}/^{86}\text{Sr}$. Conversely, three samples from the Eastern palaeorivers (EN4, EN13, EN14) show
464 more pronounced grain size variation but with no consistent sign of change in either ϵ_{Nd} and
465 $^{87}\text{Sr}/^{86}\text{Sr}$. This result suggests that the sediments proximal to the Ennedi Highlands are
466 immature and poorly sorted when compared to sediments in the Bodélé Depression and the
467 Bahr El Gazel.

468

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

481

482 **4. Conclusions**

483 We identify three readily discernible North African dust PSAs, defined geographically using a
484 high resolution Dust Source Activation Frequency map (Schepanski et al., 2012) and
485 characterised geochemically using Sr and Nd isotopes (Figure 8). We weight geochemical
486 data by locality dust source activation to produce representative estimates of the isotopic
487 signature of emitted dust. The Western Source area is characterised by the most
488 unradiogenic Nd and most radiogenic Sr signature, reflecting the old cratonic bedrock
489 geology. More work is needed to validate the isotopic signatures of the major sites of dust
490 activation in this understudied region, particularly targeting the alluvial sediments in
491 Southern Algeria, Eastern Mali and North West Niger. The historically underestimated
492 Eastern source region is geochemically well defined, and shows radiogenic Nd and non-

493 radiogenic Sr values. The Central dust source region is highly geochemically heterogenous.
494 We find that the isotopic signature of the Bodélé Depression is the result of mixing from
495 three geochemically distinct palaeo-river systems: the Bahr El Gazel, and rivers draining
496 from the Tibesti Mountains and Eastern highlands. We identify a strong grain-size effect on
497 the Nd and Sr isotopic signature of the immature Eastern palaeoriver sediments in
498 particular, highlighting the need to determine the nature and importance of grain size
499 effects in a specific study region.

500

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

514

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732 **Figure Captions:**

733 **Figure 1: Previous analyses of North African preferential dust source areas (PSAs).** 1 ° x 1 ° maps of
734 North African dust sources from March 2006 to February 2010, based on A) OMI aerosol index (Map
735 adapted from Schepanski et al., 2012; frequency-based remote sensing method) B) MSG SEVIRI IR
736 dust index (Adapted from Schepanski, *et al.*, 2012; employs backtracking method therefore
737 removing dust transport bias). C) Geographical definition of North African Potential Source Areas
738 (PSAs) of (Adapted from Scheuvens et al., 2013) based on a variety of remote sensing techniques
739 that conflate dust transport and emission. D) Geochemical characterisation of North African PSAs, as
740 defined in Figure 1C, in Nd-Sr isotope space (PSAs 1 - 4 & 6 as defined by Scheuvens et al., 2013, PSA
741 5 from Abouchami et al., 2013).

742

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

751

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

754

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

760

761 **Figure 5: Isotopic composition of our three North African PSAs.** New data (squares, this study) and
 762 published data (circles) (Abouchami et al., 2013; Gross et al., 2016; Grousset et al., 1998; Kumar et
 763 al., 2014; Padoan et al., 2011; Zhao et al., 2018) from North African dust source regions. Size
 764 corresponds to annual DSAF (Schepanski et al., 2012). Crosses denote mean isotopic values for each
 765 source region weighted by annual DSAF +/- one weighted standard deviation. Where only ϵ_{Nd} or
 766 $^{87}Sr/^{86}Sr$ data is available, sample is not plotted, but the available data still contribute to the
 767 weighted PSA mean.

768

769

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

778

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

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

785

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

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