1	Rapid changes of dust geochemistry in the Saharan Air Layer
2	linked to sources and meteorology
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20	Abstract
21	Based at Izaña Observatory (~2400 m a.s.l. in Tenerife), we performed 1-hour
22	resolution measurements of elemental composition of dust in the Saharan Air Layer (SAL)
23	and studied the variability of the ratios of these elements to aluminium (elemental ratios).
24	In a period (~1 week) of continuous dust presence (50-200 $\mu$ g/m <sup>3</sup> ), we observed rapid
25	variations of dust composition; some elemental ratios changed by a factor 2 in a few (5 to
26	8) hours. The lowest variability (Normalized Variability Range, %) was found for Si/Al
27	(9%) and Fe/Al (9%), followed by the ratios of K, Ti, Mg, Mn, Ca and Sr to Al (20 to 80 %),
28	and the highest for S/Al, Na/Al and Cl/Al (110 to 160 %) and a number of trace metals (Cr,
29	Cu, Ni, Zn, Zr) and Br (>200%). This variability was induced by the alternating impacts of
30	three of the large North African dust sources: NE Algeria (rich in evaporite minerals
31	bearing Ca, S, Sr, K and Mg and in illite mineral), Western Sahara to Bechar region
32	(containing Na, S and Cl rich Yermosol soils) and SW Sahara – Western Sahel (rich in illite
33	and hematite). We traced the variability in large-scale meteorology using the so-called
34	North African Dipole Intensity (NAFDI: strength of the subtropical Saharan high -Morocco-
35	to the monsoon tropical low -Nigeria-). The mobilization of dust from the different sources
36	was associated with westward propagating Harmattan pulses linked to the change of
37	phase of NAFDI (- to +), the associated westward shifts of the Saharan Heat Low and

39 and NAFDI: moderate NAFDI values (0 to +2.5) were associated with Ca, K, Na, Mg and S

convective monsoon inflow. We found a correlation between dust composition in the SAL

rich dust linked to dust sources in NE Algeria, whereas higher NAFDI values (+2.5 to +4)
were linked to Fe rich dust (Ca, Na and S depleted) linked to dust sources in SW Sahara –
Western Sahel. The results of this study also show that some trace elements (Br, Cr, Ni, Zn and Zr) are influenced by industrial emissions into North Africa.

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45 Keywords: Saharan Air Layer, Saharan dust sources, dust geochemistry, North Africa,
46 Harmattan, NAFDI.

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1. Introduction

49 Desert dust emitted from soil by the action of wind is the second most abundant 50 atmospheric aerosol after sea salt (Andreae and Rosenfeld, 2008). Dense dust plumes 51 occur over thousands of kilometres beyond their desert source regions (Prospero et al., 52 2002). This has implications on radiative fluxes (Miller et al., 2014), cloud formation and 53 properties (Boose et al., 2016), nutrients deposition on ecosystems (Ravelo-Pérez et al., 54 2016) and human health (Zhang et al., 2016). Dust is a mixing of tens of minerals (Kandler 55 et al., 2009; Reid et al., 2003), each presenting physical and chemical properties which 56 modulate dust impacts, e.g. feldspars are active ice nuclei (Atkinson et al., 2013), hematite 57 absorb UV radiation (Alfaro et al., 2004), carbonates neutralize acid pollutants (Ito and 58 Feng, 2010), whereas iron bearing clays and oxides provide iron to ecosystems (Rizzolo et 59 al., 2017). Some models are already including such mineral diversity (e.g. Nickovic et al., 60 2012; Perlwitz et al., 2015). Experimental methods still have limitations for quantifying 61 the long-term variability of dust mineralogy, thus complementary elemental composition 62 data is being used (Rodríguez et al., 2012), even for model validation (Pérez García-Pando 63 et al., 2016).

64 North Africa is the largest and most active dust source region, accounting for 50 to 65 70 % of global emissions (Huneeus et al., 2011). In summertime, dust is uplifted to high 66 altitude (Cuesta et al., 2009). Then exported in the Saharan Air Layer (SAL; Prospero and 67 Carlson, 1972), i.e. the dusty airstream that flows over the North Atlantic at altitudes 1 - 5 68 km a.s.l. off North Africa and at < 2 km.a.s.l. over the Caribbean (Tsamalis et al., 2013). 69 Dust sources are mostly located in topographic lows, associated with drainage of ancient 70 watercourses and endorheic basins (Prospero et al., 2002) where fluvial sediments 71 accumulated during the so-called African Humid Periods (De Menocal and Tierney, 2012; 72 Middleton et al., 2018; Skonieczny et al., 2015). The variability of soil dust composition 73 across the North African sources is a topic of major interest; however in-situ aerosol 74 measurements are scarce (Scheuvens et al., 2013) and proxies based on dust mineralogy 75 in specific site extrapolated using the FAO Soil Map of the World are being used (Claquin

et al., 1999; Nickovic et al., 2012; Journet et al., 2014; Pérez García-Pando et al., 2016). The
variety of dust sources impacting in distant regions has also been identified with isotopic
characterization (Bozlaker et al., 2018).

79 The variability of dust composition in the SAL (Kandler et al., 2007; Rodríguez et 80 al., 2011) may also depend on meteorology, more specifically (i) on the occurrence of the 81 specific meteorological scenarios activating dust emissions (Fiedler et al., 2013; Flamant 82 et al., 2007), prompting regional dust mobilization (Schepanski et al., 2017) and dust 83 export to the Atlantic (Jones et al., 2003; Tsamalis et al., 2013), and (ii) on wind speed, 84 which influences the deposition rates of the minerals linked to the coarser dust fractions. 85 Meteorology linked to dust export in summertime, and its connection to climate related 86 processes (monsoon, Saharan Heat Low, etc...), is a topic of major interest (Engelstaedter 87 and Washington, 2007; Evan et al., 2016; Cuevas et al., 2017; Schepanski et al., 2017).

This study addresses three questions: how quick does dust composition change in the SAL?, what is the connection to dust sources?, and what is the role of meteorology?. We address these questions by performing temporal high resolved measurements of elemental composition of dust in the high altitude Saharan Air Layer.

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#### 2. Methods

#### 2.1 <u>Aerosol samples</u>

95 In this study we analysed a data set of 1-hour time resolution measurements of 96 elemental composition of aerosol samples collected at Izaña Observatory in Tenerife, 97 located at 2400 m.a.s.l. The samples were collected from 23-Aug 19h to 30-Aug 17h 2010. 98 The hourly resolution sampling was performed with two streakers (D'Alessandro et al., 99 2003): one to collect samples of total particulate matter (PM<sub>T</sub>) and another one to collect 100 samples of coarse (2.5-10  $\mu$ m) and fine (< 2.5  $\mu$ m) aerosol fractions, i.e. PM<sub>2.5-10</sub> and PM<sub>2.5</sub>, 101 respectively. A total of 166 hourly samples were collected with each streaker.  $PM_T$  and 102 PM<sub>2.5</sub> are collected on polycarbonate membranes while PM<sub>2.5-10</sub> is collected by impaction 103 on thin Kapton® foils. Here in after we will refer to PM<sub>T</sub>, PM<sub>2.5-10</sub> or PM<sub>2.5</sub> as PM<sub>x</sub>. We used 104 this dataset to study the variability of dust composition, the potential dust sources in 105 North Africa and the relationship between variability of dust composition and 106 meteorology.

A second dataset, based on sampling from 1-Aug to 31-Aug 2013 (sampling from 22h to 08h), was used to assess if the relationship between variability of dust composition and meteorology observed in 2010 was also observed in other summers. This additional data set includes the elemental composition of PM<sub>10</sub> aerosols collected on Teflon 47 mm filters at an airflow of 2.3 m<sup>3</sup>/h. Bulk PM<sub>10</sub> concentrations were determined by gravimetry, 112 by conditioning the filters at 20 °C following the EN-14907 procedure (except that RH was

113 kept to 30% instead of 50%).

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The data set used in this study is available at Rodríguez et al. (2019).

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- 116 2.2 <u>PIXE analysis</u>

117 PM<sub>x</sub> samples collected at Izaña have been analysed by Particle Induced X-ray 118 Emission (PIXE) and Particle Induced Gamma-ray Emission (PIGE) at the Tandetron 119 accelerator of the LABEC-INFN laboratory (Florence, Italy), where a specific, high 120 efficiency PIXE-PIGE set-up has been developed for the analysis of aerosol samples 121 (Lucarelli et al., 2014, 2018, Calzolai et al., 2015).

Each sample was irradiated with a 3.0 MeV proton beam (10-150 nA intensity) for 60-90 s. For Teflon samples, a filter scanning was carried out to analyse most of the deposit area, while hourly samples were analysed by a properly collimated beam, which scanned the deposit in steps corresponding to 1 h of aerosol sampling, thus providing the elemental concentrations with hourly time resolution (Calzolai et al., 2015).

127 PIXE spectra were fitted using the GUPIX code (Maxwell et al., 1995) and elemental 128 concentrations (Na, Mg, Al, Si, P, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Br, Rb, Sr, Zr, 129 Mo, Ba, Pb) were obtained by a calibration curve from a set of thin standards (Micromatter 130 Inc.). The lighter elements (Na, Mg, Al and Si) concentrations were corrected for self-131 absorption effects using experimental corrective factors obtained by PIGE measurements 132 of Na and Al (Calzolai et al., 2010; Formenti et al., 2010). Detection limits are within 1-10 133 ng/m<sup>3</sup> for elements from Na to V and 1 ng/m<sup>3</sup> for elements from Cr to Pb. A verification of 134 the overall accuracy was made by analysing the NIST SRM 2783 standard (Air Particulate 135 on Filter Media).

136 It is worth noting that PIXE is an appropriate technique for the study of particulate 137 matter, as it is multielemental, rapid, very sensitive, not destructive, and it does not 138 require any sample pre-treatment. In particular it is very effective for the study of mineral 139 dust as it is particularly sensitive for the detection of medium Z elements, including 140 important soil related elements, such as Al, Si, K, Ca, Ti, Sr, Mn and Fe.

Bulk dust concentration was determined by considering that Al accounts for 8% of dust in the Earth Crust (Mason, 1966). This is consistent with our observations: see the slope of Al versus bulk dust in PM<sub>10</sub> determined by gravimetry (Fig. 1A). These PM<sub>10</sub> samples are basically constituted by dust, as their ochre colour indicates (Fig. 1B).

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#### 2.3 Potential Source Areas of dust

148 The Potential Source Areas of dust were identified by analysing the "Median Ratios 149 At Receptor" plots, determined for the ratio of each element (X) to Al (X/Al), based on the 150 elemental composition of dust at Izaña receptor site and backtrajectories. These plots 151 represent the median ratio of each X/Al measured at the receptor site (Izaña in this study) 152 when the air mass has previously passed above each  $1^{\circ} \times 1^{\circ}$  degree pixel over North Africa 153 (e.g. Fig. 4, discussed below). Back-trajectories were calculated with HYSPLIT software 154 (Stein et al., 2015) using GDAS data. A total of 166 back-trajectories were calculated, one 155 for each hour observation of aerosol composition based on the streaker sampling: 23-Aug 156 19h to 30-Aug 17h 2010 (Fig. 1C). The identification of the Potential Source Areas (based 157 on Median Ratios At Receptor plots analysis; Fig.4) was performed only during the period 158 when Izaña was within the dusty SAL (dust > 40  $\mu$ g/m<sup>3</sup>; 24-Aug 11h to 30-Aug 17h 2010); 159 the back-trajectories density map (number of back-trajectory points that passed by each 160  $1^{\circ} \times 1^{\circ}$  degree pixel of the study domain) of this period is shown in Fig. 1D. In this group 161 (Fig 1D), the median value of the X/Al ratios was calculated for each  $1^{\circ} \times 1^{\circ}$  degree pixel 162 (Fig.4), only if at least 5 back-trajectories pass by that pixel. This calculation method of the 163 Median Ratios At Receptor is similar to that used by Rodríguez et al. (2011) for studying 164 the potential source areas of the pollutants observed in the dusty SAL and by García et al. 165 (2017) for studying the transatlantic transport of dust and aerosol pollution from North 166 America in the westerlies. This method does not use the vertical wind component. We 167 performed several tests, and calculated the MRAR plots considering only the back-168 trajectories points located at low altitude (below certain threshold altitudes). Results did 169 not differ significantly from those obtained without limiting the altitude of the back-170 trajectories (not shown for the sake of brevity).

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#### 2.4 Meteorology and satellite dust observations

173 Meteorology was studied using the NCEP/NCAR re-analysis data (Kalnay et al., 174 1996), whereas satellite Dust Optical Depth observations (dark target + deep blue MODIS 175 combination) were provided by the Giovanni system of NASA (Levy et al., 2010; Hsu et al., 176 2013). For studying some events, the hourly images and Dust Optical Depth (Brindley and 177 Russell, 2009; Legrand et al., 2001) measured by the Spinning Enhanced Visible and Infra-178 Red Imager (SEVIRI) radiometer boarded on Meteosat Second Generation and compiled 179 the WMO-SDSWAS (https://sds-was.aemet.es/forecast-products/dustby portal 180 observations/) were also used.

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#### 183 **3. Results and discussion**

184 Figure 2A illustrates how the dusty SAL impacted over the North Atlantic in the 185 study period. Figure 2C shows the so-called Potential Source Areas of North African dust 186 that may influence dust composition, according to Scheuvens et al. (2013), whereas Figure 187 2D-2I shows the meteorological fields (described below) that illustrate the complex 188 summer meteorological scenario in North Africa. The positive correlation observed between dust at Izaña and Harmattan wind speed (central Algeria), during 30 years (Fig. 189 190 2B), illustrates the key role of meteorology in the processes involved in dust export. We 191 studied the links between the variability of dust composition in the SAL (Fig.2A), dust 192 sources (Fig. 2C) and meteorology (Fig. 2D-2I).

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#### 3.1 Temporal variability of dust composition

195 Fig. 3A shows 1-hour resolution measurements of dust concentration at Izaña 196 Observatory (Tenerife), from 23-Aug 19h to 30-Aug 17h 2010. Izaña is located above the 197 marine boundary layer, and directly exposed to the dusty SAL (see details in Rodríguez et 198 al., 2015). Although three size fractions were independently measured (PM<sub>T</sub>, PM<sub>2.5-10</sub> and 199  $PM_{2.5}$ ) for simplicity, we perform our descriptions just in terms of total dust (dust<sub>T</sub>). 200 Initially (23-Aug 19h to 24-Aug 02h) dust<sub>T</sub> concentrations were low (~15  $\mu$ g/m<sup>3</sup>), but 201 increased from 18 to  $\sim$ 70 µg/m<sup>3</sup> on 24-Aug (02h to 22h; Fig.2A). During most of the 202 sampling period (~6 days: 24-Aug 11h to 30-Aug 17h) Izaña Observatory was 203 permanently impacted by the dusty SAL (Fig. 2A), with dust<sub>T</sub> concentrations ranging from 204 40 to 200  $\mu$ g/m<sup>3</sup> (Fig. 3A). Fig. 3B-3I shows the ratios of Ca, S, Mg, Cl, Na, K, Si and Fe to Al 205 when Izaña was impacted by the SAL.

206 In order to compare the mean aerosol composition with the mean composition of 207 soil dust, we determined the Enrichment Factor (EF), using Al as tracer and the average 208 upper continental crust composition of Mason (1966). Results support the idea that the 209 analysed elements of the aerosol population in the SAL were dominated by soil desert 210 dust. On average we found low EFs (0.5 to 2) for most of the study elements (Ni, Na, Si, Cu, 211 K, Mn, Zr, Fe, Mg, Sr, Ca, Ti, P, Zn and Cr). As average, dust was slightly depleted in Na, Si, K 212 and Mn, and slightly enriched in Sr, Ca, Ti and P. The highest EFs were found for Br (30 to 213 60 in the different size ranges), S (~ 20 to 65) and Cl (40-45). Trace metals (Ni, Cr, Zn and 214 Zr) showed higher EF in the fine (2 to 4) than in the coarse and total (< 2) fractions. These 215 results of the EF are consistent with the ochre colour of the samples (Fig.1B) and the slope 216 of Al versus PM<sub>10</sub> (8%, Fig. 1A; similar to the mean content of Al in the Earth Crust).

Dust composition in the SAL experienced a significant variability, traced by the ratios of some elements to Al, e.g. those of Ca, S, Mg, Cl, Na, K and Fe (Fig. 3B-3I). The ratio

- Ca/Al changed by a factor ~2 in a few (5 to 8) hours. Top of Fig.3B highlights some events
  (E1 to E6) discussed below, e.g. pulses of Ca rich dust occurred around 25-Aug ~00h
  (event E1), the 27-Aug around 07h (E4) and the 29-Aug at 23h (E6; Fig. 3B). Pulses of dust
  rich in other elements were also recorded (Fig. 3C-3I).
- We quantified the variability of dust composition by determining the NOrmalized
  VAriability Range (NOVAR) of each ratio to aluminium, defined as:

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where the average (arithmetic mean), the percentile 98<sup>th</sup> (P98<sup>th</sup>) and the percentile 2<sup>nd</sup> (P2<sup>nd</sup>) refer to the ratio X/Al in the dust samples collected in the SAL. The values of X/Al (Table 1) are within the range of those observed in other studies (Rodríguez et al., 2011 and references within). The lowest variability was found for Si/Al (NOVAR: 9%) and Fe/Al (9%), followed by X/Al of key dust components, such as Ti, K, Mg, Mn, Ca and Sr (20 to 80 %) and S, Na and Cl (111 to 156 %). Trace elements, as Cr, Ni, Cu, Zn, Br and Zr, showed high X/Al NOVAR values (107 to 277%).

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#### 3.2 Dust source regions

NOVAR =  $100 \cdot (P98^{th} - P2^{nd})$  / average

237 We assessed how dust composition in the SAL (Fig. 3) (i.e. the X/Al) changes 238 depending on the source region of dust. We determined the Median Ratios At Receptor 239 (MRAR) plots, which represents the typical (median) X/Al of a given element measured at 240 Izaña receptor site when the air mass has passed (according to back-trajectories; Fig. 1C) 241 by each 1°x1° pixel of the study domain (Fig.4). The MRAR (Fig.4) allows identifying (i) 242 source areas of aerosols and (ii) transport pathways from the source to the receptor site, 243 especially when there are very few and small sources. The airstream that may influence 244 the transport pathways in North Africa are illustrated with the arrows shown in Fig 1C. 245 The potential influence of the industrial emission on trace elements was also assessed 246 (location of industrial sources shown in Fig. 1D, according to Rodríguez et al., 2011). We 247 determined the MRAR only using the data collected when Izaña was impacted by the dusty 248 SAL (dust > 40  $\mu$ g/m<sup>3</sup>; 24-Aug 11h to 30-Aug 17h 2010). The plots show that the X/Al of:

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- Si, Fe and Mn are high when dusty air arrives from southern Sahara (Fig.4A, 4C and 4E),
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• Ca, Sr, S, K and Mg are high when dust arrives from northeast Algeria (Fig.4B, 4D, 4F, 4K and 4H),

Na are Cl are high when dust airflow arrives from northwest Algeria (Fig.4G and
4I),

• trace elements (Br, Cr, Ni, Zn and Zr) shows transport pathways from regions affected by industrial emissions in Tunisia, Algeria and Morocco (Fig. 4M-4Q),

257 Our results were compared with the location of the so-called Potential Source 258 Areas (PSAs) proposed by Scheuvens et al. (2013) as results of a literature review. These 259 PSAs are plotted in Fig. 2C1 and 2C2 to highlight their location in topographic lows, some 260 of them associated with the watersheds (Skonieczny et al., 2015). Briefly: (1) PSA1 261 expands over Northeast Algeria and Tunisia; (2) PSA2 is placed south of Atlas mount, 262 along the northern side of the Tamanrasset paleo-river (ending in Arguin bay, 263 Mauritania); (3) PSA3 is located east of Hoggar massif, throughout the southern part of the 264 Tamanrasset paleo-river and the northern part of the Niger river basin; (4) PSA4 is the 265 northern slope of Tibesti massif, and (5) PSA5 is the Bodele depression.

266 Fig. 5 shows the scatter plot of some elements (Ca, Sr, S, Na, Cl, Si and Fe) to Al, 267 indicating the slope of the different trending groups. In order to put our results in the 268 context of the considered key large-scale dust sources (Scheuvens et al., 2013), the median 269 X/Al value measured at Izaña when the air masses have passed by the PSAs 1 to 3 is 270 shown in Table 2, even if the spatial variability of dust composition is most probably not 271 limited to these PSAs (Nickovic et al., 2012; Journet et al., 2014). The X/Al ratios in some 272 key Si-Al bearing minerals (Table 3) will be used to discuss the regional variability we 273 observe in dust composition (Fig.4). The location of the industrial areas in North Africa 274 (with potential emissions of trace elements) is shown in Fig. 1D.

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3.2.1. <u>Group -1: Ca, Sr, S, K, Mg</u>

These elements show a high X/Al in PSA1. Main features:

278 Ca/Al has a high variability (NOVAR = 78%; Table 1). The scatter plot of Ca vs Al • 279 shows two main groups of dust particles: Ca depleted (ratio, slope - S1 = 0.39; Fig. 280 5A1) and Ca rich (S2 = 0.68; Fig. 5A1). Ca rich dust is observed in PSA1 (ratio to Al 281  $\sim$  0.62; Fig.4B), whereas PSA3 ( $\sim$ 0.40) is Ca depleted. This variability also occurs 282 in the coarse and fine fractions (Fig. 5A). The rather high ratio of Ca/Al ( $\sim 0.5$ ) 283 observed in northern PSA2 is probably influenced by a transport pathway from 284 PSA1 (Fig. 4B). A hot spot rich in Ca is also observed in inner Morocco (in-Mo, a 285 Potential Source Area to which we will refer as PSA-in-Mo-), where Ginoux et al. 286 (2012) attributed dust emissions to anthropogenic use of soil (probably the 287 agriculture fields western of Marrakesh, which match with the location of PSA-in-288 Mo pointed in Fig.4B. Ca mostly occurs in calcite (CaCO<sub>3</sub>) and gypsum/anhydrite 289  $(CaSO_4 \cdot 2H_2O / CaSO_4)$ , and secondarily as dolomite  $((CaMg)_2CO_3)$  and anorthite 290 (CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub>). The first two evaporite minerals are abundant in PSA1 and 2, and

- scarce in PSA3 (according to the soil mineralogical maps of Nickovic et al., 2012 and
  Journet et al., 2014), a distribution consistent with our observations (Fig. 4B) and
  with those of Kandler et al. (2007).
- Sr/Al has a NOVAR=87% (Table 1). Roughly two types of dust particles are observed: Sr depleted (S1 =  $4.0 \cdot 10^{-3}$ ; Fig. 5B1) in PSA3 (Fig.4D) and Sr rich (S2 =  $6.2 \cdot 10^{-3}$ ; Fig. 5B1) near PSA1 (Fig. 4D). The spatial distribution resembles that of Ca, with high levels in PSA1, northern PSA2 and In-Mo. This Sr-Ca co-variability is probably due to the fact that Sr has a high geochemical affinity with Ca and uses to substitutes it in calcite and gypsum (CaSO<sub>4</sub>·2H<sub>2</sub>O). A similar Sr-Ca co-variability was found by Moreno et al. (2006).
- 301 S/Al shows a very high variability (NOVAR = 111%; Table 1). Two trends are 302 observed: S poor (slope = 0.06; Fig.5C1) dust in PSA3 (Fig.4D) and S rich (slope = 303 0.15; Fig.5C1) dust in PSA1, northern PSA2 and In-Mo (Fig.4D). The aerosol 304 samples collected (under dust free conditions) from 23-Aug 19h to 24-Aug 10h 305 were not included in this analysis (i.e. neither Fig. 4 and Table 1); they showed a 306 much higher S/Al ratio (S3 = 0.44; Fig 5C1) attributed to the transport of fine 307 sulphate pollution (Fig. 5C3) from the Mediterranean (back trajectories not shown 308 for the sake of brevity). Sulphur is usually present as gypsum/anhydrite in soil 309 dust, with higher amounts in PSA1 and 2, and lower in PSA3 (Claquin et al., 1999).
- Mg/Al showed a smoother variability (NOVAR=25%), with typical ratios between
  0.23 and 0.29 (Table 1), slightly higher than that characteristic of the illite (Table
  3), a dominant clay in North African dust (Reid et al., 2003). Mg also occurs in
  dolomite (CaMg)<sub>2</sub>CO<sub>3</sub>), and other clays (e.g. chlorite, vermiculite) and feldspars
  (Table 3). The presence of Ca-Mg carbonate (dolomite) in northern Algeria (Fig.
  4B), accounts for the observed Mg rich dust in PSA1 (0.28) compared to PSA2 and
  PSA3 (0.24; Fig. 4H).
- K/Al also has a smooth variability (NOVAR=25%; Table 1). A northward gradient is observed, with values < 0.20 south of 25°N, and the highest ratios (0.22) in PSA1 (Fig. 4K). The high K/Al ratio in PSA1 (Fig.4K) is consistent with the atlases that report the presence of soils rich in illite in this specific area (Nickovic et al., 2012), a clay mineral characterised by a high K/Al ratio (=0.67; Table 3), that contributes to increase the K/Al ratio.</li>
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The dust rich in Ca, S, Sr, K and Mg in PSA1 are associated with a Cretaceous limestone-rich basement, also rich in illite (Nickovic et al., 2012), and with the occurrence of dusty dry lakebeds, so-called chotts, where evaporite minerals (rich in Ca, Na, Sr, K 327 carbonates and sulphates) occur (Hamdi-Aissa et al., 2004 and references within
328 Rodríguez et al., 2011). Dust emission in this region, where the chotts Felrhir and Melghir
329 (eastern Algeria) and the chotts el-Djerid and el-Gharsa (Tunisia) are located, is detected
330 by satellite (Prospero et al., 2002). Its impact in the SAL was described by Rodríguez et
331 al.(2011).

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#### 3.2.2. <u>Group -2: Na and Cl</u>

334 Na/Al and Cl/Al exhibit a high NOVAR (116% and 156%, respectively; Table 1) 335 and a northward gradient, with the highest values along the northern PSA2 (ratio  $\sim 0.3$  for 336 Na and  $\sim 0.14$  for Cl; Table 2), compared to PSA3 (ratio  $\sim 0.15$  for Na and  $\sim 0.05$  for Cl; 337 Fig. 4G and 4I). There are some hot spots rich in these elements in PSA1 (ratio  $\sim 0.27$  for 338 Na and ~ 0.10 for Cl; Fig. 4G and 4I). The slope of Na vs Cl (~2.2,  $r^2 = 0.92$ ) is much higher 339 than that in sea salt (which typically ranges from 0.5 to 0.6) and this discards marine 340 emissions as the main source of Na. This is consistent with the observations of Prospero 341 and Carlson (1972), who showed that sea salt is not present in the SAL because the 342 airstream lies above the marine boundary layer. The scatter plots give evidence for the 343 occurrence of two types of dust particles: depleted (S1 = 0.17 for Na and S1 = 0.05 for Cl) 344 and enriched (S2 = 0.31 for Na and S2 = 0.11 for Cl) in these elements (Fig.5D and Fig.5E). 345 Northern PSA2 is characterised by Halip Yermosol soil (Nickovic et al., 2012), a FAO 346 category for Na-rich saline lands, which in this region occurs around the abundant 347 sebkhas (Rodríguez et al., 2011), where precipitated Na-sulphate, Na-carbonate and NaCl 348 evaporites are dust components (Fehlberg and Stahr, 1985; Hamdi-Aissa et al., 2004). This 349 is probably the origin of the high Na/Al and Cl/Al ratios in this region (Fig. 4G and 4I). 350 High ratios of none-ammonium sulfate / Al ratios have been observed in the SAL when 351 dust originates in northern PSA2 (Rodríguez et al., 2011).

Dust emission in northern PSA2, Bechar basin, was also identified by Fiedler et al.(2013) and Knippertz et al.(2007). The Na and Cl rich dust in PSA2 is linked to the presence of Yermosol soils, a feature consistent with the hot spots of Ca, S and Sr observed in this region (Fig. 4B, 4D, 4F).

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#### 3.2.3. <u>Group -3: Si, Fe and Mn</u>

This group exhibits high X/Al in PSA3 (Fig. 4A, 4C and 4E and Table 2). A hot spot is also observed in PSA1. The ratio of:

Si/Al varied in a narrow interval (NOVAR = 9%), with values of ~ 2.04, 1.98 and
2.10 in PSAs 1, 2 and 3 respectively (Fig. 4A and Table 2). This narrow variability
(Fig. 5F1 and 5F2) is consistent with the results of Reid et al. (2003) and Kandler

363 et al., (2007), who found that silicon in the SAL was mainly bounded to 364 aluminosilicate. The higher Si/Al in PSA1 and northern-PSA3 is consistent with the 365 mineralogical map that report on soils rich in illite clays in these regions (Nickovic 366 et al., 2012; Journet et al., 2014), a mineral with a high Si/Al ratio (=2.8; Table 3) 367 that contribute to increase the Si/Al ratio in the mineralogical cocktail constituting 368 the bulk dust mass. A similar Si/Al variability, linked to the content of illite, was 369 observed in Cape Verde (Caquineau et al., 1998). Some feldspar, as orthoclase, 370 exhibits high Si/Al ratios (3.12, Table 3), but the soils of northern PSA3 are 371 depleted in these minerals, according to Nickovic et al. (2012). The Si/Al ratios 372 observed in the SAL over the North Atlantic (1.9 to 2.2, according to this study and 373 to Kaldler et al., 2007 in Tenerife; Reid et al., 2003, in Puerto Rico and Caquineau et al., 1998, in Cape Verde) are lower than those observed near the dust sources in 374 375 North Africa (3 to 6) according to Scheuvens et al. (2013). The quartz in the North 376 African soils mostly occurs in the coarse - silt fraction (Journet et al. 2014), so 377 deposition during long-range transport in the SAL accounts for the observed 378 decreased in the Si/Al ratio. Nonetheless, northern PSA3 soils are rich in quartz 379 (Nickovic et al., 2012), so even if this mineral is present in low amounts in small 380 size fractions (Kandler et al., 2007) it may also contribute to the high Si/Al ratio 381 observed in the dust arriving from this region (Fig. 4A).

- 382 Fe/Al also had a narrow variability (Fig. 5G1-5G3) and a low NOVAR (= 9%), with 383 values  $\sim 0.48$ , 0.46 and 0.49 in PSA 1, 2 and 3, respectively (Fig. 4C and Table 2). 384 This variability range is similar to that observed in previous studies (Moreno et al., 385 2006; Formenti et al., 2008; Scheuvens et al., 2013). Fe is regularly included in (i) 386 Si-Al-bearing minerals (Reid et al., 2003) and (ii) nanoparticles of iron 387 oxides/hydroxides (hematite:  $Fe_2O_3$  and goethite:  $\alpha$ -FeO(OH))) attached to the 388 surface of other dust particles (Reynolds et al., 2014; Moskowitz et al., 2016) that 389 contributes to increase the observed Fe/Al in comparison with the Fe/Al ratios in 390 the Si-Al-bearing minerals (0.16 for illite and 0.43 for vermiculite; Table 3). The 391 high Fe/Al ratio we observe in PSA3 (Fig. 4C) is consistent with the mineralogical 392 maps that report on hematite rich soils in western-PSA3 (Journet et al., 2014). The 393 higher Fe vs Al slope in the fine than in coarse fraction (Fig. 5G1-5G3) is consistent 394 with the study of Kander et al. (2007), who found a decrease in the amount of 395 hematite with increasing particle size.
- Mn/Al exhibited a wider variability (NOVAR = 46%), with values ~ 7.9, 7.3 and
   8.3·10<sup>-3</sup> in PSA 1, 2 and 3 (Fig. 4E and Table 2). Because of its geochemical affinity,
   Mn may partially replace to Fe in Fe-oxides.

400 PSA3 is among the most active dust sources in summer (Engelstaedter and 401 Washington, 2007; Flamant et al., 2007; Knippertz, 2008). The high Si/Al and Fe/Al we 402 observe in this dust from this area is consistent with the soils rich in illite and hematite 403 reported for this region (Nickovic et al., 2012; Journet et al., 2014).

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3.2.4. <u>Group -4: trace elements</u>

406The MRAR plots of Ti, Cu, Br, Cr, Ni, Zn and Zr are shown in Fig. 4J-4P. A detailed407analysis is out of the scope of this study, so we did just a brief exploration.

408 The ratios of Br, Cr, Ni, Zn and Zr to Al exhibit high values downwind of the main 409 industrial areas of Tunisia, Algeria and Morocco (shown in Fig.1D and Fig.4M), where 410 oil refineries, fertilizing industry, coal power plants and urban areas occur (see details 411 in Rodríguez et al. 2011). The Atlantic coast of Morocco shows high ratios of Br, Cr, Ni, 412 Zn, Ti and Zr to Al (Fig. 4M-4Q). A clear transport pathway of Br, Cr, Ni, Zn and Zr from 413 Hassi Messaoud industrial area, in Eastern Algeria, is observed (Fig. 4M-4Q). Although 414 marine emissions may contribute to Br, anthropogenic emissions (e.g. pesticide 415 application, chemical manufacturing, coal burning, and PVC usage and disposal; 416 Lammel et al., 2002) play a role. A fact that is consistent with the absence of Na-Cl 417 marine sea salt described above.

418 • Soil emissions may accounts for the high ratios of Ti and Cu to Al in PSA2 (Fig. 4J and
419 4L).

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#### 3.3 Large scale meteorology

In this final section we assess if the day-to-day variability in meteorology in North
Africa accounts for the observed temporal variability of dust composition in the SAL
(Fig.3) in relation to the location of the dust sources (Fig. 2C and 4).

425 The summer meteorological scenario in North Africa (Fig. 2D-2I) is characterised 426 by (i) the so-called North African High (NAFH, centred over Northern Algeria at standard 427 levels above 850Pha; Fig. 2G; Font-Tullot, 1950, UK Meteorological Office, 1962), (ii) the 428 InterTropical Convergence Zone (ITCZ) - also so-called Intertropical Discontinuity (ITD, 429 Scott, 1952) – located between 16 and 23 °N (Fig.2A), (iii) the monsoon inflow south of the 430 ITCZ, and (iv) the Saharan Heat Low (SHL; Lavaysse et al., 2009), which results in high 431 temperatures and high thickness of the 1000 – 500 hPa layer (Fig.2H-2I). North (dry side) 432 of the ITCZ, Harmattan (subsiding) wind prevails (Fig.1D-1E), whereas South (wet side) of 433 the ITCZ the (uplifting) monsoon inflow occurs (Fig.1F). Dust emission is associated with 434 the vertical transfer of moment linked to the morning breakdown of low level jets (Fiedler

et al., 2013; Schepanski et al., 2017) and the occurrence of afternoon deep convection cells
(Knippertz et al., 2007; Marshman et al., 2008; Schepanski et al., 2017). These emission
mechanisms tend to have a meso (and even smaller) scale (Fiedler et al., 2013; Flamant et
al., 2007), and they occur embedded in the larger scale patterns involved in the regional
dust mobilization (e.g. SHL, Harmattan, monsoon inflow, etc., Allen and Washington, 2013;
Engelstaedter et al., 2015) and dust export in the SAL (Jones et al., 2003; Tsamalis et al.,
2013).

442 For studying the variability of dust composition, we traced the variability of the 443 meteorology using the concept of North African Dipole Intensity (NAFDI), which measures 444 the intensity of the subtropical NAFH to the tropical low pressure of the monsoon. The 445 NAFDI was previously used by Rodríguez et al. (2015) for studying three decades of dust 446 export to the Atlantic, and by Cuevas et al. (2017) and Schepanski et al. (2017) for 447 studying intraseasonal dust export. We determined the daily values of the NAFDI, using 448 the NCEP/NCAR re-analysis data (Kalnay et al., 1996) as the difference of the anomalies of 449 the daily values (d) of the 700hPa geo-potential height ( $\Phi_{\text{(d)}}$ ) over Morocco (Mo, 30–32°N, 450  $5-8^{\circ}W$ ) and Nigeria (Ni,  $30-32^{\circ}N$ ,  $5-8^{\circ}E$ ), with respect to the climatological (average) 451 values from 1980 to 2010 ( $\Phi_{\text{(clim)}}$ ; Cuevas et al., 2017):

 $NAFDI = \frac{1}{10} \left( \left( \Phi_{(d)} - \Phi_{(clim)} \right)_{Mo} - \left( \Phi_{(d)} - \Phi_{(clim)} \right) \right)_{Ni} Eq.1$ 

The selected point in Morocco is sensitive to the location (East-West shifts) of the NAFH (see M in Fig 1G), whereas the point in Nigeria is within a main path of the northward monsoon inflow (see N in Fig. 1D) associated with uplifting air (see N in Fig. 1F). We analysed the relationship between dust composition and NAFDI in two periods: August 2010 (Fig.6B1-6H1) and August 2013 (Fig.6B2-6H2).

458 Daily dust concentrations at Izaña Observatory are positively correlated with the 459 daily values of NAFDI (lag -1; Figure 6B1 and 6B2), indicating that positive values of this 460 index are associated with enhanced dust export to the subtropical North Atlantic. Fig. 7 461 shows the average meteorological fields (Fig. 7A-7E) and Dust Optical Depth (Fig. 7F) 462 during the negative and positive phases of the NAFDI; results are consistent with those 463 obtained by Cuevas et al. (2017) and Schepanski et al. (2017). Cuevas et al. (2017) showed 464 that the changes of phase of NAFDI are connected with the mid latitude Rossby waves. The 465 change of phase of NAFDI (negative to positive) is associated with a westward shift of the 466 NAFH (centred over Tunisia in the NAFDI negative phase, Fig.7A1, and shifted to Morocco 467 in the NAFDI positive phase, Fig.7A2) and a westward propagation of the Harmattan Band 468 from Eastern Algeria - Libya (Mediterranean inflow, Kallos et al., 1998; Fig.7B1) to the

469 Atlantic coast of the Western Sahara (Fig.7B2). The change of NAFDI negative to positive is 470 associated with a westward propagation of the dusty air mass within North Africa 471 (Fig.7B1 to 7B2). The enhanced dust export that we observe to the Mediterranean during 472 negative NAFDI (Fig. 7F1) was previously found by Cuevas et al. (2017) and Schepanski et 473 al. (2017). Cuevas et al. (2017) also described the westward shift of the SHL that we 474 observe when the NAFDI changes from negative (Fig.7C1-7D1) to positive (Fig.7C2-7D2) 475 phase (also linked to enhanced deep convection in Mauritania, Fig.7E1-7E2), which is 476 associated with the westward propagation of cooler Mediterranean air in the Harmattan 477 Band (Fig.7B1-7B2). Analogously, Wang et al. (2017) also observed a reinforcement of the 478 NAFH during the westward shift of the SHL.

479 The meteorological changes in North Africa associated with the change of phase of 480 NAFDI have implications for the three large dust sources in western North Africa (PSA 1-3 481 highlighted in Fig.7B1-7B2) and on the composition of the dust exported in the SAL. Fig.6 482 shows bulk dust concentrations (Fig.6B1) and the ratios of Fe/Al, Ca/Al, K/Al, Na/Al, 483 Mg/Al and S/Al (Fig.6C1-6H1) in the SAL (measured at Izaña) versus daily NAFDI (lag -1) 484 values during our study period, August 2010. In order to verify if the observed 485 relationships were observed in other periods, we performed a similar analysis with a data 486 set collected in August 2013 Fig. (6B2-6H2). Increasing values of NAFDI (in the positive 487 range) are correlated with higher dust concentrations in the SAL (Fig.6B1-6B2), with an 488 increase in the Fe/Al ratio (dust richer in Fe, Fig.6C1-6C2), and with a decrease in the 489 ratios of Ca, K, Na, Mg and S to Al (Fig.6D-6H). In summary, moderate values of NAFDI (0 490 to +2.5) are associated with Ca, K, Na, Mg and S rich dust in the SAL linked to Northern 491 Saharan sources (northern PSA1 and PSA2; Fig.2C and Fig.4-4H), whereas higher NAFDI 492 values (2.5 to +4) are associated with Fe rich dust from Southern Saharan regions (PSA3; 493 Fig.2C and 4C)

These results are consistent with our previous meteorological description (Fig.7).
This interpretation is also supported by the detailed meteorological analysis that we did
for each of the events E1 to E6 shown on top of Fig.3B. Three brief examples:

<u>Event E1</u> (Fig. 8). Calcium rich dust impacted on Izaña the 24-Aug 2010 (Fig. 3B). Dust mobilization started the 19-Aug at PSA1 (Fig. 8A1 and Fig.11A; NE Algeria to Tunisia).
 The dusty air mass shifted westward across North Africa (Fig. 8A1-8F1, back-trajectory plotted in white colour; the arrow points to the daily location of the airmass) linked to a westward shift of the NAFH (Fig. 9A3-9F3), the Harmattan Band (Fig. 8A2-8F2), the SHL (Fig. 8A5-8F5) and the uplifting monsoon inflow south of the ITCZ (Fig. 8A4-8F4), in a sequence similar to that described above. The evolution of

the NAFDI values (change from - to +) is indicated on the top right side of plots of Fig.8A1-8F1.

<u>Event E2</u> (Fig.9). Na and Cl rich dust (Fig. 3E and 3F) impacted at Izaña on the 26 Aug
 2010. The surge of dust emissions occurred in northern edge of PSA2 (Bechar basin,
 border between Algeria and Morocco; Fig. 9A1-9A2), where the previous cited
 Yermosol occurs, in a scenario (Fig.11B) similar to that described by Fiedler et al.
 (2013) and Rodríguez et al. (2011). Again, a westward shift of the Harmattan Band and
 the NAFH (Fig.9B1-9B3 and 9C1-9C3) prompted dust export to the Atlantic.

- 512 Event E5 (Fig. 10) impacted at Izaña on the 27 and 28 Aug 2010 (Fig 3B). This is the • 513 only case in which an impact of Si rich (Si/Al: 2.12; Fig. 3H), Fe rich (Fe/Al: 0.51; Fig. 514 31), linked to hematite rich dust of western-PSA3, and Ca and S depleted dust (Fig. 3B-515 3C) was recorded. Dust uplift was linked to a deep convective cell that propagated, 516 within the monsoon inflow (Fig. 10C-10D), from Mali to Mauritania (Fig. 10A1-10D1) 517 in a scenario similar to those described by Marsham et al. (2008) and Bou Karam et al. 518 (2008). Enhanced dust export is clearly observed in the satellite Dust Optical Depth 519 (Fig.10A1 and 10D1). It is associated with a westward shift of the NAFH (Fig 10A3-520 10D3), of the monsoon inflow (purple contour of the negative omega domain; Fig. 521 10A4-10D4) and of the SHL region (Fig 11C-11D); all these movements were traced by 522 the increasing values of the NAFDI (top right of plots Fig. 10A1-10D1), from +1.76 to 523 +2.86 (lag -1).
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#### 4. Conclusions

526 The high temporal resolution measurements performed in this study shows that 527 dust composition in the Saharan Air Layer (SAL) experiences rapid variations, induced by 528 the (meteorologically modulated) alternated impacts of different regional sources of 529 North African dust. Dust composition (ratios of elements to Al) in the SAL experiences a 530 significant variability in a few (5 to 8) hours, in such a way that, up to eight impacts of 531 three of the large North African dust sources (NE Algeria, Western Sahara to Bechar region 532 and Southern Sahara - Sahel) may occur in less than 1 week. The mobilization of dust from 533 the different sources is associated with the westward propagating Harmattan pulses, the 534 associated westward shifts of the Saharan Heat Low and convection, including processes 535 embedded within the monsoon inflow. The North African Dipole Intensity (NAFDI: 536 strength of the subtropical North African High, at Morocco, to the monsoon tropical low at 537 Nigeria) traces the observed variability in dust composition. Moderate values of NAFDI (0 538 to +2.5) are associated with Ca, K, Na, Mg and S rich dust (linked to Northern Sahara 539 sources) in the Atlantic SAL, higher NAFDI values (2.5 to 4) are associated with Fe rich

dust in the SAL (linked to Southern Sahara), whereas negative values of NAFDI promoted
(Ca, K, Na, Mg and S rich) dust export to the Mediterranean. Trace metals (Br, Cr, Ni, Zn
and Zr) are influenced by industrial emissions in North Africa.

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#### 5. Acknowledgements

545 This study was performed within the project VARDUST-SAL (PGC2018-099166-B-546 100), funded by the Ministry of Science, Research and Innovation of Spain, the Research 547 State Agency of Spain and the European Regional Development Fund (ERDF). JLD is 548 awarded with a posdoc contract Agustín de Bethencourt, funded by the Program Fomento 549 de Transferencia del Conocimiento, funded by the Cabildo de Tenerife. A stage of JLD at 550 the INFN was funded by the Universidad de La Laguna. The authors gratefully 551 acknowledge (i) the NOAA Air Resources Laboratory for providing the HYSPLIT transport 552 and dispersion model, (ii) the NOAA NCEP Reanalysis data provided by the 553 NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their Web site at 554 https://www.esrl.noaa.gov/psd/ and (iii) the Giovanni online data system, developed and 555 maintained by the NASA GES DISC. We also thank to Dr. Albert Solé for his useful 556 comments on soils and sabhkas.

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- 794

Table 1. Ratio of elements to Al (based on 128 hourly samples) for major (Si to Cl,  $\mu g/\mu g$ ) and trace (Ti to Sr,  $ng/\mu g$ ) elements registered in the dusty SAL from 24-Aug 17h to 30-Aug 00h 2010 at Izaña Observatory. Includes percentiles 98<sup>th</sup>, percentiles 2<sup>nd</sup> and average (arithmetic mean) for total particles. NOVAR (%) for total, coarse and fine particles. NA: none available.

	total	total	total	total	coarse	fine	
	98 <sup>th</sup>	$2^{nd}$	average	NOVAR, %	NOVAR, %	NOVAR, %	
Si	2.13	1.95	2.03	9	11	NA	
Fe	0.51	0.46	0.48	9	11	18	
Κ	0.24	0.19	0.21	25	26	42	
Mg	0.29	0.23	0.25	25	32	36	
Ca	0.74	0.37	0.47	79	107	76	
S	0.19	0.06	0.12	111	172	119	
Na	0.36	0.10	0.22	116	152	117	
Cl	0.16	0.04	0.08	156	212	177	
Ti	64.3	53.0	57.2	20	35	44	
Mn	8.83	6.32	7.66	33	77	74	
Sr	6.73	3.33	4.60	74	104	83	

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Table 2. Ratios of several elements to Al for major (Si to Cl,  $\mu g/\mu g$ ) and trace elements (Ti to Sr, ng/ $\mu g$ ) measured in the dusty SAL at Izaña when the air mass has passed by several potential source areas (PSA), according to the MRAR analysis. Data extracted from the Median Ratios At Receptor matrix.

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	PSA1	northern-PSA2	PSA3	PSA-in-Mo
Si	2.04	1.98	2.10	2.01
Fe	0.48	0.47	0.50	0.48
Κ	0.22	0.22	0.20	0.22
Mg	0.27	0.25	0.24	0.27
Ca	0.60	0.49	0.43	0.60
S	0.18	0.18	0.09	0.16
Na	0.24	0.30	0.15	0.26
Cl	0.07	0.14	0.05	0.09
Ti	55.0	57.0	57.0	57.0
Mn	8.3	7.0	8.3	7.7
Sr	6.0	5.0	4.0	6.0

811 Table 3. Properties of key minerals components of North African dust. Includes a qualitative description of the mass size distribution between the clays (with most 812 of the mass  $< 2\mu$ m) and silt (with most of the mass between 2 to 60 $\mu$ m) size range 813 typically used in modeling (source: Journet et al., 2014) and the ratios to Al of Al-814 815 bearing minerals determined using their empirical formulas (www.webmineral.com). 816

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Mineral	clay size	silt size	Si/Al	Fe/Al	Mg/Al	K/Al	Ca/Al	Na/Al
Clay group:								
Illite	dominant	negligible	2.80	0.16	0.21	0.67		
Kaolinite	dominant	negligible	1.04					
Smectite group:	dominant	negligible						
Montmorillonite			2.08				0.07	0.09
Chlorite group:	present	present						
Clinochlore			1.56	1.29	1.69			
Chamosite			1.56	3.62	0.68			
Feldspar group:	minority	majority						
Orthoclase			3.12			1.45		
Evaporites group:								
Gypsum	minority	majority						
Calcite	present	present						
Dolomite	present	present						
Oxides group:								
Hematite	dominant	negligible						
Goethite	present	present						
Quartz	minority	majority						

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### 828 Figures

829 **Figure 1**. A) Scatter plot of Aluminium vs  $PM_{10}$  concentrations in aerosol samples 830 collected in the SAL at Izaña in August 2013. B) Picture of a PM<sub>10</sub> aerosol sample 831 collected in the SAL at Izaña is shown to illustrate the ochre colour. C) Back-832 trajectories to Izaña (red point) calculated for every hour of the study period, 23-Aug 19h to 30-Aug 17h 2010 (total: 166 back-trajectories). Key airflow (1.1 to 2) 833 834 are highlighted with arrow. D) Back-trajectories density map (number of back-835 trajectory points that passed by each  $1^{\circ} \times 1^{\circ}$  degree pixel of the study domain) for the back-trajectories group (24-Aug 11h to 30-Aug 17h 2010) used for identifying 836 the potential source areas. The industrial areas over Tunisia, Algeria and Morocco 837 838 are plotted (see details in Rodríguez et al., 2011).

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840 Figure 2. A) Dust Optical Depth (DOD) measured by MODIS (average 23-30 Aug 841 2010). B) dust record at Izaña and zonal component of the Harmattan wind in the 842 Subtropical Saharan Stripe (SSS 25-28°N, 7°W-2°E) from 1987 to 2017. 843 Topographic (C1) and watershed (C2) maps showing the Potential Source Areas 844 (PSA) of dust (Scheuvens et al., 2013). D-I) Mean meteorological fields (23-30 Aug 845 2010). Key components of the North African summer meteorology: Harmattan 846 Band along the SSS (HB, white arrow), ITCZ (dotted white line), SHL, African 847 Easterly Jet (AEJ) and the SAL is highlighted. Inner Morocco (in-Mo) potential source area cited in the text is highlighted. White line circles - M and N - highlights 848 849 the location of the Morocco and Nigeria for the calculation of NAFDI.

850

**Figure 3**. Time series of hourly concentrations of dust (A) and of ratios (B-H) of several elements (Ca, S, Mg, Cl, Na, K, Si and Fe) to aluminium in total, coarse (2.5-10  $\mu$ m) and fine (<2.5 $\mu$ m) dust fractions at Izaña Observatory from 23 Aug 19h to 30 Aug 17h 2010. Ratios are shown in the period when Izaña was impacted by the Saharan Air Layer.

856

Figure 4. Median Ratios At Receptor –MRAR- plots for several elements (Si, Ca, Fe,
S, Mn, Sr, Na, Mg, Cl, Ti, K, Cu, Br, Cr, Ni, Zn and Zr) measured in the dusty SAL at
Izaña (24-Aug 17h to 30-Aug 16h), based on 144 back-trajectories. The plots
include the location of the potential source areas of dust (1, 2, 3 and In-Mo in A and
G) and the location of the industrial (M).

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**Figure 5**. Scatter plot of several elements to Al measured by PIXE in 1h resolution samples collected at Izaña Observatory with two streakers: one for total particles (first column) and other for coarse (central column) and fine (right column) particles. Blue dots in the plot of sulphur (C) indicates samples collected under dust free conditions (23 -16h- to 24 -16h- Aug 2010), previous to the dusty – SAL impact. The slopes (S1 to S3) indicate the different trends, since they are representative of the X/Al.

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Figure 6. A) Location of three of the potential source areas (PSA) of dust in North
Africa, Izaña and the two reference sites (M: Morocco and Nigeria) to calculate
NAFDI. B-H) Concentrations of dust and elemental ratios to Aluminum (X/Al)
versus daily NAFDI for 24-31 Aug 2010 (column 1, left) and 1-31 Aug 2013

(column 2, right). A decreasing scale is used for NAFDI to highlight the westward
transport to the Atlantic (with respect to the PSA) under positive NAFDI values.

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Figure 7. Meteorological fields (A-E) and satellite Dust Optical Depth (F) during
negative (right column) and positive (left column) phases of NAFDI. HB:
Harmattan Band. SHL: Saharan Heat Low. Vertical wind is shown in terms of
omega parameter (negative and positive values indicate upward and downward
movements, respectively).

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884 Figure 8. Event E1 (24-Aug 16h- 25-Aug 01h 2010): Ca rich dust linked to transport from PSA1 to Izaña Observatory. The evolution (from 19-Aug to 24 Aug 885 886 2010) of the Dust Optical Depth and back-trajectory (A1-F1), horizontal wind at 850 hPa (A2-F2), height of the geopotential of 850 hPa (A3-F3), vertical wind at 887 888 850hPa (A4-F4) and thickness of the 1000-500 hPa layer (A5-F5) is shown. Column 1: white arrow indicates the track of the back-trajectory for each day. 889 890 Column 2: white arrow highlights the Harmattan Band (HB). Column 3: H highlight 891 the location of the core of the North African anticyclone. Column 4: vertical wind is 892 shown in terms of omega parameter (negative and positive values indicate upward 893 and downward movements, respectively). Daily value of NAFDI is shown on the 894 right top of the plots of column 1. HB is also shown with arrow in column 2, 4 and 895 5.

896

897 Figure 9. Event E2 (25-Aug 18h – 26 Aug 07h 2010): transport of Cl and Na rich 898 dust linked to transport from PSA2 to Izaña Observatory. The evolution (21-Aug to 899 23-Aug 2010) of the Dust Optical Depth and back-trajectory (A1-C1), horizontal 900 wind at 925hPa (A2-C2) and height of the geopotential of 850hPa (A3-C3). Column 901 1: white arrow indicates the track of the back-trajectory for each day, white point 902 position at 00h. Column 2: white arrow highlights the the Harmattan Band (HB). 903 Column 3: H highlights the location of the core of the North African anticyclone. 904 Daily value of NAFDI is shown on the top right of the plots of column 1.

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906 Figure 10. Event E5 (27-Aug 22h – 28 Aug 13h 2010): transport of Fe rich dust 907 from PSA3 to Izaña Observatory. The evolution (25-Aug to 28-Aug 2010) of the 908 Dust Optical Depth and back-trajectory (A1-D1), horizontal wind at 850hPa (A2-909 D2), height of the geopotential of 850hPa (A3-D3), vertical wind (A4-D4) and 910 temperature at surface (A5-D5). Column 1: white arrow indicates the track of the 911 back-trajectory for each day. Column 2: the location of the Harmattan Band (HB) is 912 highlighted. Column 3: H highlights the location of the core of the North African 913 anticyclone. Column 4: vertical wind component is shown in terms of the omega 914 parameter (negative and positive values indicate upward and downward 915 movements, respectively). Daily value of NAFDI is shown on the top right of the 916 plots of column 1.

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Figure 11. Satellite dust observations based on SEVIRI infrared (10.8 μm) product (A
and C) and Aerosol Optical Depth – SEVIRI based (B and D) during dust events occurring
the 19 (18h), 22 (10h), 25(12h) and 25(13h) of Aug 2010. Arrows show the direction of
transport of dust.

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- 923
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Figure 7

#### E1 - Ca rich dust – PSA1





E2 - CI and Na rich dust – northern PSA2

#### E5 - Fe rich dust – PSA3



## dust observations



19-Aug 18h



2.0

1.0

0.0



Figure 11

# Graphical abstract

