1	Variability in grain sediment particle size, mineralogy, and Fe mode of	
2	occurrence of Fe in surface sediments of preferential across dust-source inland	
3	drainage basins: The case of the Lower Drâa Valley, -S Morocco	
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53 54 Abstract 55 The effects of mineral dust emittededsert dust from arid and semiarid surfaces upon climate and 56 ecosystems is highly affected bydepends strongly ondepends fundamentally on theitsir particle 57 sizeparticle -size distribution (PSD) and size-resolved mineralogical compositionical compositiony. 58 However, there is very limited quantitative knowledge on the particle size and composition of the parent 59 sediments along with their variability within dust source regions, particularly in dust emission hotspots. 60 This article presents a relevant dataset of dust emitting sediments from, not only a Saharan, dust hotspot 61 but also its surroundings, which is important to better understand their respective impacts on climate and 62 air quality, and also their interactions. However, soil mineralogy atlases used for mineral-speciated dust 63 modelling are highly uncertain as they are derived extrapolating mineralogical analyses of soil samples 64 that are particularly scarce in dust-source regions. This extrapolation neglects the processes affecting the 65 formation of different dust-emitting surface sediments, such as dunes, crusts, and paved sediments. The 66 Lower Drâa Valley, an inland drainage basin and dust hotspot regionand preferential dust-source located 67 in southern Moroccothe Moroccan Sahara, was chosen for a comprehensive analysis of sediment grain 68 sizeparticle -size and mineralogy. Different sediment types samples (n=42) were collected, including 69 paleo-sediments, paved surfaces, crusts, and dunes, and analysed for particle -size distribution (through 70 PSD analysis of minimally and fully dispersed samples) and mineralogy, and X-ray diffraction mineralogical 71 analysis of bulk samples. We also performed Furthermore, Fe sequential wet extraction was carried out 72 to characterize the modes of occurrence of Fe, including Fe in Fe (Fe mineralogy, including the contents 73 of (oxyhydr)oxides, mainly from -{goethite and hematite}, which are key to dust radiative effects}, the and 74 ppoorly crystalline pool of Fe (readily exchangeable ionic Fe and Fe in nano-Fe-oxides), relevant to dust 75 impacts upon ocean biogeochemistry), and structural Fe. Based on the results Results yield we propose a 76 conceptual model where both particle sizeparticle -size and mineralogy are segregated by transport and 77 deposition of sediments during runoff of water across the basin, and by the precipitation of salts, which 78 causes a sedimentary fractionation. The proportions of ceoarser particles (substantially richerenriched in 79 quartz) is higherare more present in elevated areas in the high-lands, and while that of finer particles {rich 80 in clay, carbonates, and Fe-oxides, is higher in the low-landsare present in depressed areas, where dust 81 emission is maximized dust emission hotspots. There, wWhen water ponds and evaporates, secondary 82 carbonates and salts precipitate, and the clays are enriched in readily exchangeable ionic Fe, due to 83 sorption of dissolved Fe by illite. Our-These results differ from currently available mineralogical atlases 84 atlases and highlight the need for observationally-constrained global high-resolution mineralogical data 85 for mineral-speciated dust modeling. The obtained dataset represents an important resource for future 86 evaluation of surface mineralogy retrievals from spaceborne spectroscopy.

 Keywords: A<u>eolian, desert, sediments, rid regionsarid land, dust-sources, desert dust, dustemittingmineralogy, iron, <u>Morocce sediments formation model</u>, dust modelling.
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- 101 102 103 104 105 106 107 108 109 1. Introduction 110 Desert dust is atmospheric particulate matter (PM), mostly mineral in composition, emitted into 111 the atmosphere by wind erosion of arid and semi-arid surfaces. The global dust source regions 112 include North Africa, the Middle East, Central Asia, Western Australia, South America, Southern 113 Africa and Southern US-Northern Mexico. These regions include most of the so-called dust 114 115
- emission "hotspots", defined as localized, persistent areas of intense dust production within an overall landscape which generally does not emit dust (Gillette, 1999; Baddock et al., 2016). From 116 these regions, North Africa accounts for around 50 % of the global dust emissions, followed by 117 Central Asia, the Middle East and East Asia (Kok et al., 2021). Dust storms arise when strong winds generate a large amount of dust particles that drastically reduce visibility nearby and are 118 119 transported over distances of hundreds of kilometres (Prospero et al. 2002). During transport, 120 dust perturbs the energy and water cycles by direct radiative forcing and influences cloud 121 formation, precipitation and the associated indirect radiative forcing (Weaver et al., 2002). Dust 122 transports nutrients across the planet affecting ocean productivity (Boyd et al., 2007), plant 123 nutrient gain or loss (Sullivan et al., 2007, Doronzo et al., 2016, Alshemmari et al., 2013, Al-124 Dousari et al., 2020}, and glacier mass budgets (Goudie & Middleton, 2006). Dust can also 125 directly affect human health by inhalation or by favouring the propagation of diseases (Goudie 126 & Middleton, 2006, De Longeville et al., 2010; Karanasiou et al., 2012; Pérez García-Pando et al., 2014, Al-Dousari et al., 2018). It can reduce renewable solar energy output due to attenuation 127 128 of solar radiation and soiling of solar panels (Al-Dousari et al., 2019; Monteiro et al., 2022), 129 create poor visibility on roads increasing the risk of traffic accidents (Middleton, 2017) and cause
- disturbances in airport operations and air traffic (Monteiro et al., 2022)
- 131 (), and photovoltaic energy efficiency (),

132 Dust is emitted mostly from arid inland drainage basins (Dubief, 1977; Prospero et al., 2002; Goudie & Middleton, 2006; Bullard et al., 2011; Querol et al., 2019). These basins encompass 133 134 different sedimentary environments, many of which are potentially efficient sources of dust 135 emission hotspots, including unconsolidated aeolian deposits, endorheic depressions, and 136 fluvial and alluvial dominated systems (Bullard et al., 2011). Consolidated or compacted fine 137 sediments in the form of crusts and paved sediments, for instance on ephemeral lake beds, can 138 also be important dust emitting surfaces when loose sand size sediments provided by adjacent 139 sand dunes are available (Stout, 2003). These sand particles are efficiently mobilised by wind 140 and strike-blast the consolidated surface breaking the sediment aggregates and releasing dust 141 (Shao et al., 2011).

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Models developed to simulate the atmospheric dust cycle and its impact on climate represent 142 143 dust emission, transport, interactions with radiation and clouds, and removal by wet and dry 144 deposition (Tegen and Fung, 1994; Ginoux et al., 2001; Zender et al., 2004; Perez et al., 2011, 145 Klose et al., 2021). Modelling efforts have mostly focused on the representation of dust sources 146 and emission (Kok et al., 2021) and Tthe characterization of dust sources and hotspots is one of 147 the crucial aspects for representing dust mobilisation in models. Traditionally Initially, models 148 used aridity vegetation cover as a criterion to identify potential dust sources (e.g. Tegen and 149 Fung, 1994). Satellite retrievals subsequently showed that the most prolific sources occupy a 150 smaller fraction of arid regions (Prospero et al., 2002; Ginoux et al., 2012). These so-called 151 hotspots or "preferential sources" are found within enclosed basins, where easily eroded soil 152 particles accumulate after fluvial erosion and transport of from the surrounding high-lands. The 153 implementation of preferential source functions in global models based on topography (Ginoux 154 et al., 2001), hydrology (Tegen et al. 2002; Zender et al. 2003), geomorphology (Bullard et al., 155 2011), or satellite proxies (Prospero et al., 2002; Ginoux et al., 2012), has significantly improved 156 the skill of models by approximately locating large-scale natural sourcesdust hotspots. However, 157 models are not able yet to capture the small-scale spatial and temporal variability in emissions 158 apparent from observations. Some studies Models assume relatively homogeneous soil 159 properties everywheredue to the lack of data,- while- there can beis an important significant 160 heterogeneity. Furthermore Ssome studies have provided small-scale understanding on the role of geomorphology and sedimentology upon dust emissions (Bullard et al., 2011; Baddock et al., 161 162 2016). For instance, Bullard et al. (2011) developed a conceptual model of how different 163 geomorphologic surfaces affect the intensity and temporal variability in dust emissions. 164 While it is key to understand dust source-hotspot locations and emission intensity, climate 165 impacts by dust also depend upon its mineralogy (Alshemmari et al., 2013). Dust is a mixture of 166 different minerals including quartz, clay minerals (mica/illite, kaolinite, palygorskite, 167 chlorite/clinochlore and smectite/montmorillonite), feldspars (albite/anorthite and orthoclase), 168 carbonate minerals (mainly calcite and dolomite), salts (mainly halite and gypsum), Fe-oxides 169 and hydroxides (mostly goethite and hematite) and other oxides or hydroxides of Ti, Mn and Al 170 (Coudé-Gaussen et al., 1987; Schültz & Sebert, 1987; Molinaroli et al. 1993; Gomes, 1990; Sabre, 1997; Caquineau, 1997; Avila et al., 1997; Caquineau et al, 1998; Claquin et al., 1999; Al-Ghadban 171 172 et al., 1999; Cattle et al., 2002; Formenti et al., 2008; Nickovic et al., 2012; Alshemmari et al., 173 2013, Scheuvens et al., 2013; Journet et al., 2014; Scanza et al., 2015; Subramaniam et al., 2015; 174 Doronzo et al., 2016; Al-Dousari et al., 2018, 2019, 2020; Ito & Wagai, 2017; Querol et al., 2019).

The relative abundances, size, shape, and mixing state of these minerals influence the effect of

dust upon climate. For instance, the absorption of solar radiation by dust depends upon the iron
 oxide content (Tegen et al., 1997; Sokolik and Toon, 1999; Reynolds et al., 2014, Di Biagio et al.,

178 2019), ice nucleation in mixed-phase clouds is highly sensitive to the amount of K-feldspar and

179 quartz (Boose et al., 2016b; Harrison et al., 2019), and the bioavailability of iron in dust depends

180 upon its iron mineralogy and speciation (Shi et al., 2012). <u>Recent studies have shown that cloud</u>

181 pH is controlled in great part by calcite from dust (Grider et al., 2023). Furthermore, Ca is

182 <u>controlling</u>controls heterogeneous reactions of acids on the surface of dust, which ultimately

183 <u>affect O3 production (Bauer et al., 2004; Paulot et al., 2016).</u> According to the geological, 184 geomorphological and climate (weathering) patterns of the desert regions, the type, and

proportions of minerals might greatly vary (Caquineau, 1997; Caquineau et al, 1998, Claquin et

al., 1999; among others). For exampleinstance, Sahelian dust is composed mainly of quartz,

187 kaolinite and hematite, the mineralogy-Middle Eastern dust is dominated by quartz and

<u>carbonates (Al-Dousari, 2018, 2019, 2020), while and</u> in North-eastern China and the Sahara <u>dust</u>
 mica/illite, kaolinite, quartz and carbonates prevail (Shen et al., 2009; Claquin et al., 1999).

190 Despite the potential importance of dust mineralogical variations, climate models typically 191 assume dust composition as globally uniform, which is partly due to our the limited knowledge 192 of the composition of the parent sources at global scale. The few models that explicitly represent 193 dust mineralogical composition (e.g., Scanza et al., 2015; Perlwitz et al., 2015, Li et al., 2021; 194 Gonçalves Ageitos et al., 2023) use global atlases of soil type and the relation of this variable to 195 soil mineralogy. This relation is inferred using massive extrapolation from a limited amount of 196 mineralogical analyses, particularly in dust source regionshotspots, ancillary information on soil 197 texture and colour, and a number of additional assumptions (Claquin et al., 1999; Journet et al., 198 2014). The mineralogical compositionConventionally, the particle -size of dust is -is 199 characterizsed by measuringin two traditional two particlegrain -size ranges (Wentworth (1922) 200 and Urquhart (1959)), i.e. clay (<2 μ m) and silt (2-63 μ m) linked to FAO (Food and Agricultural 201 Organization of the United States) soil texture datasets based on measurements following wet 202 sieving, a technique that disperses (breaks up) the mineral aggregates found in the undisturbed parent soil into smaller particles (Chatenet et al., 1996). Furthermore, the samples that underpin 203 204 these atlases consider the first 10-15 cm of soil sediment, which is much deeper than the thin 205 layer that is relevant to wind erosion and dust emission, and mineralogy is normally analysed 206 after removing organic matter with hydrogen peroxide (H₂O₂), which can partially dissolve 207 carbonate minerals.

208 The assumed relationship between mineralogy and soil type in these atlases neglects the role of 209 geomorphology and sedimentology affecting the formation of different dust-emitting surface 210 sediments, such as dunes, __crusts, and paved sediments. In tThis study, we provides a 211 comprehensive analysis of the variability in grain sizeparticle -size, mineralogical composition 212 and Fe mineralogy and speciationmode of occurrence of sediments collected across the Lower Drâa Valley, an inland drainage basin and prolific dust-source located in the north-western 213 214 border of the Saharan desert in southern Morocco (Figure 1). The data collection was performed 215 carried out during a wind erosion and dust field campaign in September 2019 in the context of the FRontiers in dust minerAloGical coMposition and its Effects upoN climaTe (FRAGMENT) 216 217 project. Based on the analysis of the results we propose a conceptual model is proposed that 218 links formation processes of potential dust-emitting sediments to their particle size 219 distribution (PSD) and mineralogy across the basin.

221 2. Methodology

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222 2.1 The FRAGMENT field campaign and the study area

223 The sediment samples analysed in this study were collected during a field campaign that took-

224 place in September 2019 in the Lower Drâa Valley, west of M'Hamid, between the Erg Chigaga

and L'Bour (Figure 1a), a dry inland drainage basin where dust emission is frequent as evidenced

by satellite data (Ginoux et al. 2012) (Figure 1b). The region lies where the Sahara Desert begins,

227 to the south of the Atlas Mountain, near the Algerian border, in the Drâa River Basin. Preliminary

results from the Earth Surface Mineral Dust Source Investigation, EMIT, (Green et al., 2020) show

229 the presence of a complex regional mineralogy with fine-grained goethite, hematite (with

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substantial nano-sized hematite), gypsum sulphate salts in the lowlands (depressions) and
 Illite/muscovite, with local outcrops of carbonates in the study area (Figure 1c). The EMIT
 mineral maps show that the study area is representative of the larger area.

233 The campaign was conducted in the framework of the FRAGMENT project, in which distinct 234 desert dust source regionhotspot regions are being characterised to better understand the size-235 resolved dust emission and composition for different meteorological and soil conditions. The 236 aim of FRAGMENT is to better understand dust emission, its mineralogical composition and the 237 effects of dust upon climate, by combining field measurements, laboratory analyses, remote and 238 in situ spectroscopy, theory and modelling. FRAGMENT field campaigns consist of intensive 239 sediment sampling and meteorological and airborne dust measurements in one specific 240 location, along with sediment sampling across the broader basin. The intensive meteorological 241 and airborne dust measurements were performed in the dry lake L'Bour and are analysed in e. 242 g., González-Florez et al., 2022; Panta et al., 2022; Yus et al., in prep. Here the we-focus is on the 243 sediment sampling across the basin.

244 The study area records very low annual precipitation (ranging from <50 to 800 mm80 mm) and 245 extremely variable droughts interrupted by extreme floodings (Berger et al., 2021). The Drâa 246 River was anthropogenically dried in this area mostly due to the construction of El Mansour 247 Eddahbi dam in 1972 (near Ouarzazate). The Jbel Hassan Brahim range reaches the highest 248 altitude in the area (840 m.a.s.l.), while the Drâa River is the lowest point (570 m.a.s.l.). The 249 study region corresponds to a low relief alluvial system, unarmored and unincised according to 250 Bullard et al. (2011). Rains are scarce, but sometimes they concentrate in the mountains (high-251 lands) and even more sporadically they can directly affect the area during convective storms, creating flash floods with a high sediment load canalised by torrents or wadis, such as wadi 252 253 Latache (high-lands) (Figure 1a), which flood flat areas. In specific areas across the basin, highly 254 sediment-loaded waters can be shortly ponded on the way to Drâa River in small depressions, 255 such as dry Lake Iriki, Erg Smar or L'Bour (low-lands) (Figure 1a), among other areas, along the floodplain. Dunes are concentrated in small flat areas, near depressions, where, after wind 256 erosion, sediment can be dragged and be entrapped by the very scarce and low vegetation. 257

258 2.2 Sediment sampling

259 The sampled sediments include paleo-sediments (hereafter named sediments), paved-260 sediments, crusts, and dunes, according to the classification by Watt & Valentin (1992) and 261 Valentin & Bresson (1992). Paved sediments result from cyclic drying and aeolian erosion of the 262 surface of paleo-sediments and range from 0.5 to 2 cm of vertical depth. Crusts ranged from 0.1 263 to 2 cm of vertical depth and we differentiated two types are differentiated: i) thin depositional 264 crusts formed as result of the deposition of sediments from running water during floods, and ii) 265 thicker sedimentation crusts resulting from the sedimentation and drying of highly sediment-266 loaded waters in ponded areas of different sizes. The difference between paved sediments and 267 crusts is mostly the period of formation. The former dates fromcan date up to thousands of 268 years ago, while the latter was formed recently. However, crust might have finer sediments 269 because these are formed by ponding. Sediments are below the crusts (not exposed to the 270 atmosphere) and dunes are aeolian deposits. We used aA 50 cm² inox steel shovel was used to sample surfaces (first top cm), sediments (below surface, from 1 to 5 cm in the vertical depth) 271 272 and dunes (from surface to 5 cm). We registered c<u>C</u>oordinates, type of sample, surroundings

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273 description, and we also recorded any other important information were registered and made

274 concept drawings. We obtained total of 42 sediment samples were obtained, including crusts

275 (12), dunes (12), paved sediments (11) and sediments (7) (Figure 2) from different locations in

the Drâa River Basin (Figure S1). These were considered representative because the study
 focuses on sediments (not deposited dust) and one basin.

278 2.3 Sample treatment

279To analyse mineral-size fractionation (<10 and 10-63 μm), we applied a fully dispersed size</th>280fractionation using MilliQ-grade water and shaking the samples was applied previous-prior to281separation for 12-24 h. First, samples were subjected to 250, 63 and 10 μm sieves to obtain the282<63 and <10 μm fractions. Due to availability, the smallest opening size of the sieve was 10 μm.</td>

283 Sonic sieving was applied for 60 s at maximum sustainable power for 3 min in every sieve. Finally,

284 subsequent drying at 80 °C was applied to recover the solid fraction from the suspension.

285 2.4 Analysis

286 2.4.1 Particle size Particle -size distribution and texture

287 The particle sizeparticle -size analysis was carried out for fully (natural aggregates totally 288 dispersed) and minimally (natural aggregates minimally dispersed) dispersed PSD to obtain the 289 fully dispersed particle sizeparticle -size distribution (FDPSD) and the minimally dispersed 290 particle sizeparticle -size distribution (MDPSD) to evaluate (i) how aggregates and particles occur 291 in natural conditions (MDPSD) and (ii) the distribution of single particles that form the 292 aggregates (FDPSD). The MDPSDs were obtained with laser diffraction using a Malvern 293 Mastersizer 2000 Scirocco accessory (hereinafter, Scirocco) for minimally dispersed conditions. 294 In this case, samples of 0.3-0.5 g of the fraction <2 mm were introduced into the Scirocco 295 vibration plate with a 2 mm aperture and 5 s measuring time. FDPSDs were determined using the Malvern Mastersizer 2000 Hydro G accessory (hereinafter, Hydro) with a water suspension 296 297 and ultrasound assistance for totally dispersed conditions. In that case, the samples were pretreated following the method by Sperazza et al. (2004). The suspension was introduced into the 298 299 Hydro's sample container, pumping at 1750 rpm and stirring at 500 rpm. Results were obtained

in both cases using the Fraunhofer method (Etzler et al., 1997).

To investigate the possible occurrence of vertical segregation of the PSD (top layers are the ones that are emitting dust), 7 crust and 5 paved sediment samples were selected for verticallyresolved PSD analyses. To this end, 3 sub-samples were extracted from each sample (top, middle, and bottom sections) by scratching the surface with a cutter from top to bottom and

305 were analysed separately. The thickness of these crusts varied between 4 to 8 mm.

The pipette method was also used to analyse the texture of a soil layer or boundary according to FAO-UNESCO (1990) of a total of six samples. This allows us-to separate a suspension of the sample in MilliQ-grade water into different size fractions (>63, 2-63 and <2 μ m), dry and analyse each size-fraction individually.

310 2.4.2 Mineralogical composition

311 Quantitative mineralogical analyses of bulk sediment samples and segregated size fractions

were carried out by means of powder X-Ray Diffraction (XRD), using a Bruker D8 A25 Advance,
 with Cu K_{a1} radiation of 1.5405 Å wavelength, a Bragg Brentano geometry and a LynxEyeXE

detector. Analysis was performed at 40 mA and 40 kV with a range of angles from 4 to 60° and 314 315 angle steps of 0.019° and 10 Hz for 1 h/sample. The mineral identification was made with the 316 EVA software package by Bruker. For quantitative analyses we used the method of the internal 317 reference material by Chung (1974) was applied, with quartz as the internal reference. The ratios 318 of intensities of the different minerals versus quartz were obtained by preparing and analysing binary mixtures of the specific minerals and quartz. The accuracy of the XRD quantitative 319 320 approach was tested by analysing 16 mixtures of reference materials with known concentrations 321 of minerals. Figure S21 summarises major results, which yield relative standard deviations 322 versus the known contents of quartz (13 % of error), albite/anorthite (10 %), calcite (31 %), dolomite (14 %), mica/illite (29 %), kaolinite (11 %), gypsum (27 %), anhydrite (19 %), goethite 323 324 (42 %), hematite (50 %).

325 For an in-depth evaluation of clay mineralogy, XRD analyses of oriented aggregates following 326 the procedure by Thorez (1976) were carried out for the same six samples of the texture. We 327 treat t_{T} he samples were treated for air drying (AO), glycolation (AG) and heating (AC). 328 Mica/illite, chlorite/kaolinite, palygorskite and smectite were found in all the samples, as 329 evidenced from the bulk XRD analysis. Calcite and dolomite were dissolved by acidifying soil suspension with a strong acid as HCl and the excess used to quantify stoichiometrically the 330 331 content of carbonates using the method proposed by Horváth et al. (2005) also for the same six samples of the clay-oriented aggregates and texture. 332

To investigate the possible occurrence of mineralogical vertical segregation, the 7 crust and 5 paved sediment unaltered samples used for <u>particle sizeparticle -size</u> analysis (see section 2.4.1) were also used for vertically-resolved mineralogy analyses.

336 2.4.3 Mode of occurrence of Fe

337 Fe is a key ingredient to climatic and biological processes affected by dust. For instance, the 338 amount, mixing state and size of Fe-oxy/hydroxides determine the degree of absorption of solar radiation by dust (Engelbrecht et al., 2016) and the potential solubility of the dust deposited into 339 340 the ocean (Shi et al., 2012). However, the XRD semiquantitative analysis for Fe-oxy/hydroxides 341 are affected by large uncertainties due to the low concentrations (increasing relative errors) and 342 is not sensitive to nano-Fe-oxides (Shi et al., 2012). We complemented the XRD analyses were 343 complemented by quantifying the levels and mode of occurrence of Fe in the bulk samples using 344 the methodologyas described in Shi et al. (2009). The method is based-latter being based -345 through which based on a sequential extraction protocol to obtain the proportions of , we 346 determine the amount of readily exchangeable (adsorbed) Fe ions and Fe in nano-Fe-oxides 347 (FeA) and the amount of <u>Fe in crystalline Fe-oxides</u>, mainly <u>from</u> hematite and goethite (FeD) in 348 the samples, were determined. We used 30 mg of Arizona Test Dust (ATD; ISO 12103-1, A1 349 Ultrafine Test Dust; Powder Technology Inc.) wasere used to test the accuracy of the method 350 and extractions were done with 15 ml of extractant solution. For total Fe content (FeT), we used a two-step wet acid digestion method developed by Querol et al. (1993, 1997) and a coal fly ash 351 352 (1633b) standard sample was were used to test accuracy. The 1633b gave 7.5 % with a standard 353 deviation of 0.14 % for total Fe (reference content of 7.8 % of Fe), while ATD gave 0.076 % with a standard deviation of 0.002 % of FeA and 0.49 % with a standard deviation of 0.07 % for FeD + 354 355 FeA (reference content of 0.067 % of FeA and 0.41 % of FeD). Furthermore, by subtraction, we 356 obtained the contents of structural Fe (FeS = FeT - (FeA + FeD)) were obtained, corresponding

to the Fe fraction as elemental Fe into the structure of minerals other than Fe-oxides, such as 357 illite or other Fe-bearing minerals. Furthermore, the FeD contents were converted 358 359 stoichiometrically to hematite (Fe_2O_3) and goethite (FeO(OH)) by using the hematite/goethite 360 proportions from XRD.

2.4.5 Electron microscopy of crust and paved sediment sections 361

362 The PSD, mineralogy and morphology of crust and paved sediments can vary along the vertical 363 profile, especially in crusts where progressive sedimentation and subsequent evaporation leads 364 to inter-layering of sediments with different properties. For that purpose, crust and paved 365 sediment sections were impregnated with epoxy resin, cut, and polished with diamond paste 366 for microscopy analysis. The polished samples were coated with graphite before analysis with a

JEOM JSM-7001F SEM-EDX Scanning Electron Microscope (SEM). 367

368 3. Results and discussion

369 3.1 Regional variability

370 3.1.1 Particle size Particle -size distribution

371 We analyse tThe PSDs of the samples collected across the basin were analysed to detect possible

372 trends or size segregation patterns from high- to low-lands for the different types of sediment.

373 The mean median diameter values of each group of sediments provided in this section represent 374

the mean and standard deviation of the median diameters. Because the PSDs are generally bi-

375 modal, other PSD metrics can be found in Table 1, including the maximum, minimum and mean 376 of the median diameters for different types of sediments, location, PSD type (MDPSD and FDPS),

377 and size fraction (full range, <63 μ m and >63 to < 2000 μ m).

378 MDPSDs, excluding dune samples, show a major mode centred around 100 μ m in diameter and

379 a secondary one between 2 to 20 μ m (Figure 3a; Table 1). FDPSD's also show two modes at 5

and 100 μm (Figure 3b; Table 1). The MDPSDs and FDPSDs of dune samples are very similar with 380

381 a main mode centred around 150 μ m and a secondary small one at 5 μ m (30 times lower) (Figure

382 3c and d). Crust samples show the largest fine (0-5 μ m) fraction in MDPSD, followed by paved 383 sediments and sediments (Figure 3e). FDPSDs show a similar trend but with a larger proportion

of fine particles compared to MDPSD (Figure 3f). 384

385 The mean median diameter of the MDPSDs (Figure 4a), excluding dune samples, is 88±63 μm; 386 and that of the FDPSDs, is 27±51 µm (Figure 4a). Therefore, aggregates are about 3 times coarser 387 than individual mineral particles. As expected, dunes were coarser than other types of 388 sediments, with a mean median diameter of 219±70 µm of the FDPSDs, which is very similar to 389 that of the MDPSDs (Figure 4b). The mean median diameters of MDPSDs are 70±48, 74±45 and 113±79 µm for sediments, paved sediments and crusts, respectively (Figure 4c); whereas the 390 391 mean diameters of FDPSDs are 19±11, 21±26 and 37±77 µm for sediments, paved sediments 392 and crusts respectively, about 3 to 4 times finer (Figure 4d).

The spatial variation of the mean diameter of the FDPSDs (Figure 5) shows coarser crusts (>40 393

394 μ m) close to the high-land areas, and finer crusts (<40 μ m) near the Drâa River, likely due to

395 flooding (causing transport and deposition of fine sediments, especially in the low-lands) caused

during scarce and intensive rains. For paved sediments, sediments and dunes, spatial PSD trends 396

were not evident, with mean median diameters ranging from 10 to 120, 10 to 40 and 120 to 300
 μm, respectively, randomly located across the basin (Table 1).

According to the size classification by Valentin & Bresson (1992) and using the FDPSD data (Figure S₃₂), dune samples can be classified as sand, loamy sand, and sandy loam; sediments as silt loam and loam; paved sediments as sandy loam, loam and silt loam; and crusts as sandy loam, loam, silty clay loam and silt loam. As shown in Figure S₃₂ and due the higher transport potential of clays during rain episodes, and their accumulation during ponding, crusts tend to be further enriched in clay fractions, especially in low-lands, compared to paved sediments and sediment samples (see section 3.4).

406 3.1.2 Mineralogical composition

407 We describe hHere the mineralogy of samples collected across the basin is described to detect 408 possible trends or mineral segregation patterns from high- to low-lands for the different types 409 of sediment. The mineralogical composition (mass % composition of the bulk sample) of dunes, 410 crusts, paved sediments and sediment samples is summarised in Table 2. Dunes show a 411 homogeneous mineralogy across the study area, with mineral abundances of 74±9.7 % quartz, 5.8±2.9 % calcite, 6.7±3.6 % microcline, 6.9±3.1 % albite/anorthite, 4.1±2.3 % clay minerals, 412 413 1.0±1.4 % dolomite, 0.38±0.26 % goethite and 0.12±0.11 % hematite and trace amounts of halite and gypsum (<0.1 %) (Figure 6). In comparison to dunes, crusts are depleted in quartz (48±11 414 415 %) and feldspars (5.0±2.1 % albite/anorthite and 4.4±3.1 % microcline), and enriched in clay 416 minerals (17 \pm 8.0 %), calcite (19 \pm 8.0 %), dolomite (3.0 \pm 1.3 %) and Fe-oxides (0.24 \pm 0.28 % 417 hematite and 0.42±0.56 % goethite) (Figure 6). The content of gypsum (0.23±0.56 %) and halite 418 (2.9±5.1 %) is higher than in dune samples, but variability is large because it depends on the 419 exact point of crust sampling. Paved sediments have a similar mineralogy than crusts, for quartz (51±8.7%), calcite (17±4.9%), clay minerals (16±7.3%), albite/anorthite (6.3±1.8%), microcline 420 421 (4.5±2.5 %), dolomite (3.5±0.79 %), hematite (0.34±0.25 %), and goethite (0.38±0.38 %), but 422 with lower content of gypsum (<0.1%) (Figure 6). Sediments are also similar to paved sediments and crusts with a mean quartz content (55±11 %), calcite (17±4.6 %), clay minerals (14±6.8 %), 423 424 albite/anorthite (5.8±1.5 %), microcline (3.7±1.6 %), dolomite (3.4±1.3 %), hematite (0.28±0.37 425 %) and goethite (0.37±0.32 %). Trace amounts of gypsum (<0.1 %) and halite (0.32±0.55 %) were 426 also found in sediments (Figure 6).

427 In comparison with the bulk sediment, the fully dispersed silt fraction (10-63 µm) shows a lower 428 amount of quartz (35±6.4 %) and feldspars (7.4±2.5 %), a higher content of carbonates (25±5.2 429 %), clays (22±10 %) and hematite (1.07±0.38 %) and a similar content of goethite (0.61±0.32 %). 430 In the fully dispersed <10 μ m sieved fraction, the amount of quartz (23±5.2 %) and feldspars (4.7±1.1%) is two times lower than in the bulk sediments. The fraction of carbonates remains 431 432 similar (21±9.0 %) and the content of clays increases substantially (38±9.8 %) compared to the 433 bulk and silt-size mineralogy. The Fe-oxide content increases by about a factor two for both hematite (2.2±2.0 %) and goethite (1.8±1.2 %). Table 3 compares our-the mineralogical results 434 435 in the clay and silt size ranges, both with the fully dispersed separation and the pipette methods, 436 against the corresponding values provided by the available global mineralogical atlases of 437 Claquin et al. (1999) and Journet et al. (2014), which assume our-the sample locations to be 438 either fluvisols or yermosols in terms of soil type. In the silt-size fraction, we find-similar contents 439 of quartz, total clay, mica/illite, chlorite+kaolinite, calcite and Fe-oxides, were found, but 3 times 440 less feldspars and 5 times more dolomite. Compared to the clay-size fraction in the atlases, our 441 <u>this study</u>the <10 μ m fraction in this study, shows larger content of quartz and feldspars (by 442 factor of 2 to 4), a 30 % lower total clay content and similar contents of calcite and Fe-oxides, 443 which can only be partly explained by the difference in the size fraction considered (<10 µm vs 444 <2 µm) as shown by the results obtained with the pipette method. Because kaolinite and chlorite 445 have coincident spacing at 7 Å in the XRD spectra, in current atlases these minerals may be 446 confounded, whereas in our this study we quantified chlorite was quantified separately by 447 identifying other minor peaks in the spectra. This is relevant as both minerals are very different 448 in terms of chemical composition. In thisour study, we also detected minor concentrations of 449 dolomite and traces of smectite and palygorskite, were also detected. The large differences in 450 the silt-size feldspar content may be largely due to the lack of data and coarse assumptions used 451 in current atlases.

452 Table 4S1 in the supplemental material compares the silt+clay and sand proportions and the 453 mineral contents of the crusts from this study in Morocco with those from deposited dust in 454 different arid regions of the world. The FD-PSD data from this study evidences that 72% of the 455 particles in the crusts fall in the clay+silt fraction (<63 µm), while 28% in the sand size-range. 456 This is close to the average value (74 and 26%, respectively) calculated from the existing studies 457 on deposited dust. Concerning the mineralogy, the crusts of this study are enriched in clays and 458 depleted in carbonate minerals and feldspars compared with the average of the mineralogy of 459 deposited dust shown in(-Table 4S1).

460

461 In our this analysis of trends in mineralogy from the high-lands to the low-lands, we considered. 462 all sample types except dunes, were considered. The low-lands, such as L'Bour and Erg Smar, 463 are enriched in clay minerals (17±9.6 and 14±3.4 %, respectively) compared to the high-lands 464 (9.1±0.97 %) (Figure 6). Mica/illite is the most common clay mineral reaching mean contents of 465 9.1±4.8, 8.1±2.0 in Erg Smar and L'Bour, respectively, and 5.0±0.70 % in the high-lands. Kaolinite reaches 7.2±5.4, 4.9±2.1 and 3.5±0.30 % and clinochlore 1.7±1.8, 1.3±0.67 and 0.49±0.38 %, 466 467 respectively. Smectite and palygorskite were detected only in trace amounts (<0.1 %) in most 468 samples, with only palygorskite at Erg Smar and high-lands reaching a mean content of 469 0.34±0.58 and 0.15±0.06 %, respectively. The same trend is found for calcite (24±13, 16±3.1 and 470 11±2.7 %, Erg Smar, L'Bour and high-lands), dolomite (5.0±5.1, 3.6±0.51 and 1.7±0.50 %, at Erg Smar, L'Bour and high-lands) and Fe-oxides (0.78±1.4, 0.37±0.43 and 0.08±0.04 % for hematite 471 472 at Erg Smar, L'Bour and high-lands and 0.42±0.51, 0.39±0.35 and 0.32±0.21 % for goethite at Erg 473 Smar, L'Bour and high-lands) being steeper for hematite than goethite (Figure 6). Quartz follows 474 an opposite trend, increasing towards the high-lands (42±18, 53±5.0 and 61±5.4 %, at Erg Smar, 475 L'Bour and high-lands, respectively) (Figure 6). Albite/anorthite and microcline do not show a 476 clear trend, with 5.5±2.3, 5.9±1.8 and 5.4±1.2 % at Erg Smar, L'Bour and high-lands, and 3.4±2.4, 477 5.0±3.4 and 4.6±1.7 %, respectively (Figure 6). Salt concentrations peak randomly and depend 478 on very local scale conditions, being higher at concave areas where ponding is favoured (see 479 section 3.4). The mean content of halite is 1.0±2.2, <0.1 and 4.0±7.7 % at Erg Smar, L'Bour and high-lands and that of gypsum is 0.18±0.35, <0.1 and 0.15±0.92 %, respectively (Figure 6). 480

A soft crust occurred on the surface of several dunes (Figure 2). The PSD and mineralogical
 analysis of the crust and the underlying sands did not reveal significant differences. Pye & Tsoar
 (2015) reported that surface hardening of dunes is due to the scavenging and deposition of clays

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- from suspended dust in light rains and by cementation of sand grainparticles (meniscus) by precipitation of carbonates and silica in the retained interstitial pore water. In both cases the potential variability caused by the slight increase of this clay and carbonate/silica cementation
- 487 is obscured by variations in the bulk mineralogy.

488 3.2 Vertical segregation in crust and paved sediments

The examination of thin vertical cross-sections provides insight into how particle sizeparticle size and mineral composition vary within the top few μm or mm of the surface. These differences
 are relevant to the mineralogy and PSD of newly emitted dust.

492 The MDPSDs of crust sections (top, middle and bottom) are very similar, with two modes of 493 occurrence at 5-7 and 200 μm (Figure S43a). Yet, while the FDPSDs show similar two modes at 494 1-5 and 100 μm for the top and middle sections and a second mode at 300 μm for the bottom 495 section (Figure S43b). The MDPSD mean median diameter of the 7 crust profiles reach 25±25, 496 54±80 and 25±26 μm for the top, middle and bottom sections, respectively, while FDPSD means 497 are 9.4±9.4 and 11±9.5 μ m in the top and middle sections and 94±145 μ m in the bottom one 498 (Figure S43c and d). Therefore, during the initial stages of ponding, coarser particles are 499 deposited first while finer particles remain suspended (see section 3.4) in the later stages before 500 evaporation of the water. Even some oxides, carbonates and salts may precipitate in the top layers of the crust as water evaporates and the ionic strength increases. 501

- 502 No vertical PSD segregation is observed in paved sediments, but some top sections analysed 503 show enrichment in coarser fractions in FDPSD (the median diameter increases from bottom 504 and middle sections (14 ± 6.8 and 12 ± 5.8 µm) to the top section (23 ± 28 µm)), likely due to
- 505 preferential erosion of finer fractions through sandblasting (see section 3.4).

The mean levels of quartz and feldspars are enriched in the bottom sections of the crusts (46±17 506 507 and 8.7±4.6 %, respectively) compared to the middle (38±11 and 8.3±2.5 %) and top sections 508 (41±12 and 6.9±2.2 %) due to the higher quartz content of the coarse fraction that is deposited 509 first (see section 3.4). The content of clay minerals, salts and Fe-oxides is similar in the top 510 (20±7.2, <0.1 and 3.3±1.9 %, respectively), middle (21±5.0, <0.1 and 2.8±1.6 %), and bottom 511 sections (19±9.1, <0.1 and 1.9±1.0 %). Carbonate minerals are relatively homogeneous, but slightly enriched in the middle and top sections (29±9.7 and 28±7.9 %, respectively) compared 512 to the bottom section (24±8.4 %). This can arise from both detrital carbonate particles and 513 514 precipitation from high ionic strength waters that are ponded and dried in the low-lands.

515 The mineral composition of the paved sediment profiles differs slightly from that of crust profiles. This is because the latter are affected by particle segregation during transport and 516 517 subsequent sedimentation. The top section of the paved sediment profiles has more quartz than 518 in the middle and bottom sections (44±8.1, 38±5.7 and 40±9.8 %, respectively), whereas 519 feldspars decrease from the bottom and middle to the top sections (9.1±4.2, 9.3±2.2 and 6.9±2.7 %). Carbonates and clay are relatively homogeneous (26±4.9, 26±2.0 and 25±4.2 % for 520 521 carbonates, and 22±8.4, 23±9.2 and 25±4.9 % for clays, respectively). The slight depletion of 522 minerals in the top section may be due to sandblasting, which tends to erode the fine fraction of the surface over time (see section 3.4). Fe-oxides are more present in the top section than in 523 the middle and bottom sections (2.1±0.47, 2.0±0.38 and 1.7±0.27 %, respectively) and the 524 525 presence of salts is very low (<0.1 % for all sections).

526 3.3 Mode of occurrence of Fe

We implemented a<u>A</u> sequential Fe extraction procedure <u>was implemented</u> to evaluate the levels
 and mode of occurrence of Fe in dust samples from the basin. Due to limitations of XRD analysis
 for low Fe-oxide contents, this procedure provided a much more precise quantitative evaluation.

530 The mean FeT content of bulk crusts, paved sediments and sediments was found to be 3.6±0.71, 531 3.4±0.47, and 3.2±0.44 %, respectively, while bulk dunes had a much lower FeT content 532 (2.0±0.33 %). Fe-speciation studies reveal that FeS percentage from FeT (FeS/FeT) is the prevailing Fe mode of occurrence (67±2.4, 69±3.0, 68±2.7 and 73±5.9 % in crusts, paved 533 534 sediments, sediments and dunes, respectively), followed by FeD percentage from FeT (FeD/FeT) (31±2.3, 29±3.0, 30±3.0, 26±5.8 %), and FeA percentage from FeT (FeA/FeT) (1.9±0.55, 1.7±0.56, 535 536 1.4±0.55 and 1.0±0.54 %). These results show that FeT is very similar between crusts, paved 537 sediments and sediments while FeT in dunes is depleted by almost 50 %. Compared to Shi et al. 538 (2011) samples from northwestern Africa, our the samples of this study areis depleted in total 539 iron (4.7 % FeT from Shi et al. (2011)), quite similar in FeS (67 % from Shi et al. (2011)), similar in FeD (33 % from Shi et al. (2011)) and much higher in FeA (0.43 % from Shi et al. (2011)). 540

The mean FeT content in the basin is similar in Erg Smar (3.6±0.27 %) and L'Bour (3.2±0.66 %) 541 542 compared to high-lands (3.0±0.24%). The ratio FeA/FeT was slightly higher at Erg Smar (1.9±0.53 543 %) but similar at L'Bour and high-lands (1.3±0.44 and 1.5±0.47 %, respectively). This is probably 544 due to the preferential accumulation of exchangeable and nano-Fe-Oxides (FeA) in the low-545 lands, where flooding results in red-water ponds and red surfaces after drying. Subsequently, 546 highly concentrated ionic Fe is trapped in the last stages of ponding, and nano-Fe-oxides may 547 precipitate during drying. Once the ponded is dried, the crusts of the low-lands tend to have a reddish patina (see section 3.4). However, a slightly higher mean FeD/FeT of 33±2.4 % is 548 549 obtained in the high-lands compared to 31±2.7 and 29±2.4 % at L'Bour and Erg Smar, respectively. The FeS/FeT mean content is slightly lower at the high-lands (65±2.5 %) compared 550 551 to Erg Smar and L'Bour (69±2.6 and 68±2.6 %, respectively).

FeD levels were apportioned between hematite and goethite using XRD proportions. These results show that in crusts, 0.79±0.66 % of hematite and 0.55±0.67 % of goethite are present, in paved sediments 0.83±0.51 and 0.64±0.54 %, in sediments 0.73±0.58 and 0.69±0.59 %, and in dunes 0.20±0.17 % and 0.68±0.24 %.

556 The proportions of FeD + FeA are higher in crusts, probably due to preferential transport of non-557 FeS to the low-lands and the trapping of Fe ions (FeA) by clay adsorption during ponding, and 558 the formation of nanosized Ferrihydrite (Fe4-5(OH,O)12. This readily exchangeable Fe has very low 559 impact on radiative forcing but a high impact in Fe fertilisation of oceans during dust events 560 (Gobler et al., 2001), as ionic Fe adsorbed by clays and nano-Fe-oxides are easily released in water solutions. The correlation of FeS, FeD and FeA with FeT is linear, with coefficients of 561 562 determination (R²) reaching 0.96, 0.89 and 0.67 for FeS, FeD and FeA respectively (Figure S54). Thus, when increasing total Fe content all modes of occurrence of Fe increase, but the increase 563 564 is preferentially driven by FeS, while it seems that the basin FeA segregation causes a lower 565 correlation with FeT.

566 **3.4 Conceptual model for** grain sizeparticle -size and mineralogy fractionation in crusts and 567 paved sediments

According to Bullard et al. (2011) and as previously discussed in this study, heavy rainfall results in the selective deposition of coarser particles from runoff and floodwaters in higher elevations. Conversely, smaller particles enriched in clays, colloidal Fe-oxides (which give the water a reddish hue), and dissolved salts tend to be transported to lower elevations. Figure 7 summarises a conceptual model that outlines the formation of crusts and paved sediment in the study area, with a focus on particle sizeparticle -size and mineralogical fractionation.

In the low-lands, floodwaters carrying fine sediments flood extensive flat areas, such as Erg Smar 574 575 or Iriki lake. Prospero et al. (2002), Bullard et al (2011) and Ginoux et al (2012), among others, 576 have shown that dust emissions originate from relatively small and localised areas where 577 sediments are supplied by floodwaters, and that the occurrence of dust emissions from these 578 areas may be partly due to the occurrence or absence of floodings. During ponding in low-lands, 579 coarser particles deposit first and form a high sand-rich bottom layer of the crust (as described in section 3.2) (Figure 7a & 8a). Subsequently, the clay fraction deposits on top of the bottom 580 581 layer until total dryness (Figure 7a & 8a) forming a second clay-rich fraction layer in the crust. 582 However, the particle size particle -size in crust surfaces is heterogeneous (Figure S65 & S76), 583 which can result in erodible dust-emitting sediment (heterogeneity enhances sandblasting). The 584 finer and more easily exchangeable FeA fraction remains in suspension until the last drying 585 stages on the most superficial layer of the crust, during drying out of the remaining ponds (as 586 described in section 3.3) (Figure 7a & 8a). During this ponding, dissolved Fe ions interact with 587 clays in such a way that they can be adsorbed on clay surfaces according to the ionic composition 588 of the waters (as described in section 3.3) (Echeverría et al., 1998). This typical ion adsorption 589 by clays is higher for montmorillonite than for other clays but the content of montmorillonite is 590 low compared to illite. In this study a high correlation is obtained for FeA and illite contents crusts 591 S<u>8</u>7). Furthermore. contain proportion (Figure а higher of 592 hematite(oxide)/goethite(hydroxide) in the FeD, due to the weathering with water during 593 transport and ponding and precipitation of nano-Fe-oxides during drying.

594 After the pond drying, the continuous heating of the clay rich surface layer causes the hardening 595 of the crust and mud-cracking, giving a 'ceramic-like' compactness to the thick crusts in the low-596 lands, usually with a reddish colour induced by the Fe-oxides (Figure So-5a). Complete drying causes mud cracks due to loss of volume, breaking the crust into polygonal pieces, whose 597 598 thickness and area depend on the amount of clay deposited. Furthermore, these concave mud-599 crust pieces resulting from the cracking usually have a grey-colour patch in the middle due to 600 the superficial precipitation of salts, which together with carbonates accumulate by capillarity 601 (see section 3.1.2) (Figure S65b). This capillary ascension and precipitation of salts (the latter 602 being an expansive process) causes sponging and breaking of the surface layers. Thus, a third 603 (top) layer is formed in the crusts of the low-lands, which is very easily eroded by wind because 604 of the spongy structure and enriched in clay and readily exchangeable Fe. In some cases, in Erg 605 Smar, we observed an additional breaking and sponging of the third (upper layer) due to 606 expansive clays, was observed. Both the ceramic-like compactness and the cementing of salts 607 give the fine-clay rich crusts in the low-lands a compact pattern with coarser MDPSD compared 608 with the high-lands where ponding is limited and very thin crusts occur. This could explain why

the crusts from the low-lands have finer FDPSDs and coarser MDPSDs compared to the high-lands (see section 3.1.1). Also, wind erosion of the few top millimetres of these crusts may result

611 in dust with higher contents of clay, Fe-oxide and salts compared to a 15 cm sediment profile.

In the high-lands, washout erosion occurs during rainfall, leading to the formation very thin 612 crusts in reduced areas. This results in sources of dust made of very thin crusts and fields of 613 614 stony surfaces with lower emission rates compared to the low-lands (Bullard et al., 2011). As 615 illustrated in Figure SZ6 the surfaces of paved sediments and their thin crusts might resemble 616 crusts profiles, but with the top section depleted on clay minerals due to preferential erosion over time, and with a very thin layer (a few micrometres) of clay minerals from the previous 617 intact formed crust after flooding or running water. The top paved sediments are more compact, 618 619 finer and have homogeneous distribution of the particle than crusts, which makes them less 620 erodible and less likely to emit dust compared to crusts (which have heterogeneous particle 621 sizeparticle-size, see section 3.2).

622 4. Conclusions

623 This study analysed the particle size particle -size and mineralogy of dust-emitting sediments in 624 the region of Drâa basin in Northern Africa, at the northwesternnorth-western fringe of the 625 Sahara. The study aimed to compare these patterns for different types of dust-emitting sediments and their variations across the basin. The results are consistent with the conceptual 626 627 models of dust emission sources in desert areas of Prospero et al. (2002) and Bullard et al. 628 (2011), which predict higher dust emissions in the low-lands than in the high-lands. The study 629 shows a clear size and mineralogical fractionation between paleo-sediments and low-land dust-630 emitting sediments, indicating that collecting samples of parent paleo-sediments for particle 631 sizeparticle -size and mineralogy may not fully represent the highly emitting-dust emission

632 sourceshotspots.

633 Both PSDs and mineralogy are segregated by transport and deposition of sediments during 634 runoff of water across the basin, and by the precipitation of salts, which causes a sedimentary 635 fractionation. Coarser particles such as quartz, feldspars, and carbonates (detrital) deposit first 636 due to friction and gravity and are enriched in high-lands. In contrast, waters reaching the low-637 lands are enriched in fine particles (clays), carbonate, salt and Fe ions from partial dissolution of 638 minerals of the source lands. When these waters are ponded in low-lands, coarser minerals 639 deposit first, followed by a second layer enriched in clays minerals. Evaporation of the last 640 ponded water layer causes the deposition of the finest particles and clays enriched in readily exchangeable ions of Fe. Once dried, the heating of the surfaces by insolation causes 641 642 evaporation of interstitial solutions moving towards the surface by capillarity, leading to the 643 precipitation of salts and secondary carbonates in the upper layer. This expansive process 644 sponges the surface of the crust, in some cases accelerated by the occurrence of expansive clays, 645 which might favour dust emission from a top clay-Fe-salts rich micro-layer. Therefore, dust emission is not only higher but also has a different mineral composition in the low-lands than in 646 647 high-lands that is also controlled by the type of sediment.

648 Our <u>The results of this studyresults</u> show that modeling mineral-speciated dust emission 649 requires understanding of the the mineralogical and <u>particle</u> size fractionation of accumulated sediments across inland enclosed basins. Large areas may act as sediment suppliers, while
 reduced areas may act as dust emitters with differences in sediment composition. Models that
 represent mineral-speciated dust emission and transport should be developed to properly

653 account for these factors.

654 Our-The results have also shown that global atlases fail to describe the clay-size fraction of dust-655 emitting sediments in the region, overestimateing __the clay mineral content and 656 underestimating_underestimate that of quartz, feldspars, and Fe-oxides. Quartz and feldspars 657 are overestimated and clay minerals underestimated in the silt-size fractions. Kaolinite-chlorite 658 are not differentiated, while our this study observes finds major differences. The classical 659 procedure loses salts during fractionation, and Fe-oxides are detected mainly by colour without 660 precision. Our-This_study detects dolomite, palygorskite, and smectite, and provides more precision for Fe-oxides, with the mode of occurrence of Fe in different types of samples and 661 662 locations. However, the study was unable to obtain a sample below 10 μ m without losing salts 663 in the process.

664 Dust models need global observationally constrained high-resolution mineral maps, which will 665 soon become available based on high-quality spaceborne spectroscopy measurements 666 performed from the International Space Station (Figure 1c, Green et al., 2020). A key challenge 667 of mineral mapping based on spectroscopy for dust emission modeling is to constrain not only 668 the presence (Figure 1c) but also the abundance of the different surface minerals. The data 669 gathered and analysed_analyzed_in this study will be used to evaluate these spaceborne 670 retrievals in forthcoming studies.

671 The large dam built in the Drâa River has caused the drying of this part of the basin, a reduction 672 of vegetation and probably increased dust emissions. The region exemplifies how anthropogenic 673 activities can promote wind erosion and represents a unique location for research on the topic. 674 Future studies may indeed explore many other aspects related to sedimentology, mineralogy, 675 wind erosion, dust emission and anthropogenic impacts, including the study of the introduction 676 of native plants and green belts to reduce wind erosion as has already been done in other 677 regions (Al-Dousari et al. 2020). 678 679 680 681 682 683 684 685 686 687

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718 Credit authorship contribution statement

CPG-P proposed and designed the field campaign with contributions of AA, KK, MK and XQ. The Campaign was implemented by CPG-P, AA, CGF, AGR, KK, MK, AP, XQ, CR and JYD. The samples were collected by CPG-P, AA, AGR, MK and XQ and analysed by CB, PC, AGR, CR and ZS. Spectroscopy was analysed by RNC. AGR performed the visualization and writing of the original draft manuscript and CPG-P and XQ supervised the work. CPG-P and XQ re-edited the manuscript and all authors contributed in data discussion, reviewing and manuscript finalization.

726 Declaration of competing interest

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727 Some authors are members of the editorial board of journal ACP. The peer-review process was

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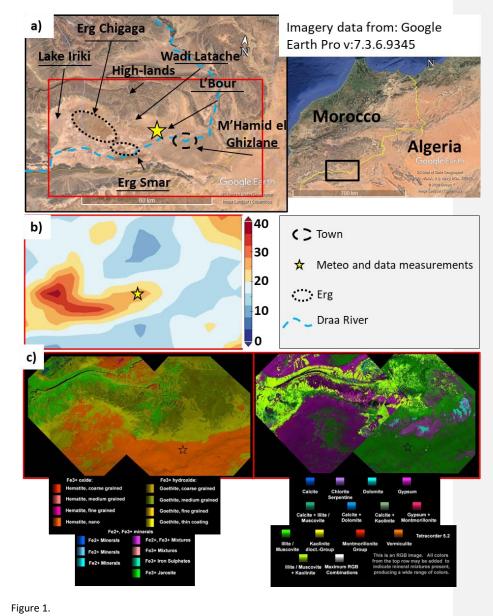
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1025 Figure captions

- 1026 Figure 1. a) Location of the study area (exact location of data measurement "star": 29°49'30"N, 1027 5°52'25"W), near M'Hamid el Ghizlane, into the Drâa basin in S Morocco. Base layer from world imagery of Google Earth Pro v:7.3.6.9345. b) Frequency of ocurrence (%) of dust 1028 1029 optical depth above 0.2 in September, October and November between 2003 and 2016 1030 derived from MODIS Deep Blue. c) EMIT scenes 1031 emit20220903t082303_o24606_s001_l2a_rfl_b0106_v01 and emit20230206t101334_003707_s000_l2a_rfl_b0106_v01 at 60 meters per pixel show the 1032 1033 diversity of Fe2+ and Fe3+ bearing minerals (left) and the EMIT 8 phyllosilicates, carbonates, 1034 and sulphates (right). The mineral maps were produced by tetracorder 5.27c1 (Clark, 2023). 1035 There is some mapped mineralogy difference at the scene boundaries, possibly due to the 1036 changing viewing geometry, and variation in atmospheric removal between the two scenes. 1037 Cirrus clouds in the scene on the right may also be impacting derived mineralogy.
- Figure 2. Images of samples collected during a field campaign near M'Hamid el Ghizlane, into
 the Draa Basin, S Morocco.
- Figure 3. Median minimally and fully dispersed PSDs of crusts, sediments, paved sediments and dunes. (a) MDPSDs and (b) FDPSDs combined from crust, sediment and paved sediment samples; (c) and (d) are MDPSDs and FDPSDs for dune samples; (e) and (f) are MDPSDs and 1043
 FDPSDs differentiated by type of sample.
- 1044Figure 4. Boxplot of median particle sizeparticle-sizediameters in μm including both fully and1045minimally dispersed analysis (a) for all samples combined excluding dunes and (b) for dune1046samples only. Also particle sizeparticle-size1047sediment for (c) minimally dispersed and (d) fully dispersed results. Means median1048diameters for each sediment type are shown with crosses.
- 1049 Figure 5. Spatial variation map with crust fully dispersed mean median particle diameter.
- Figure 6. Mean mineral group content of dune, crust, paved sediment and sediment samples,
 and also at Erg Smar, L'Bour and High-lands. Solid lines mark the mean content of all the
 samples (excluding dune samples). The dashed line divides between type and location of
 the samples.
- **Figure 7.** Schematic model of sedimentation and deposition processes in our <u>the</u> study site from high-lands to low-lands for a) crusts and for b) paved sediments.
- Figure 8. Dust emission conceptual model integrating particle sizeparticle-size distributions and mineralogy of dust source-hotspots sediments. a) Refers to the conceptual thickness and particle sizeparticle-size distributions along the basin, b) to the particle sizeparticle-size distribution and segregation of mineralogy and c) to the dust emission quantity expected depending on the place in the basin.

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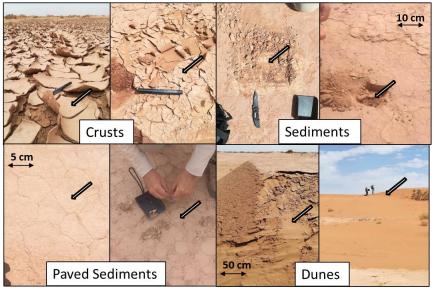
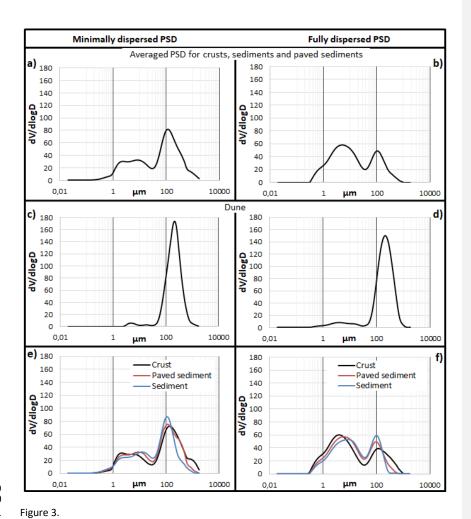
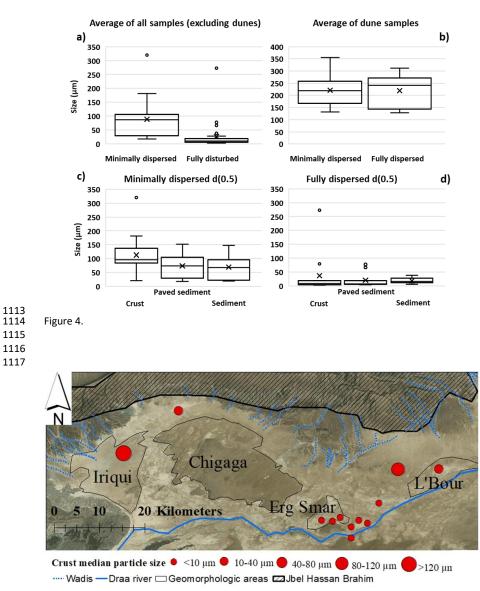
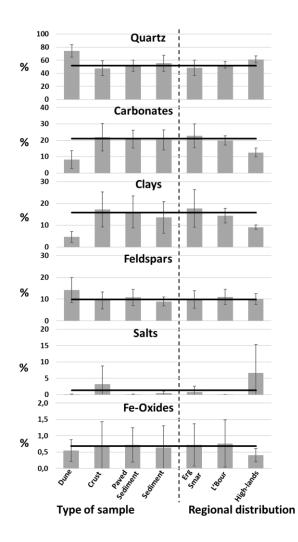


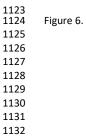
Figure 2.

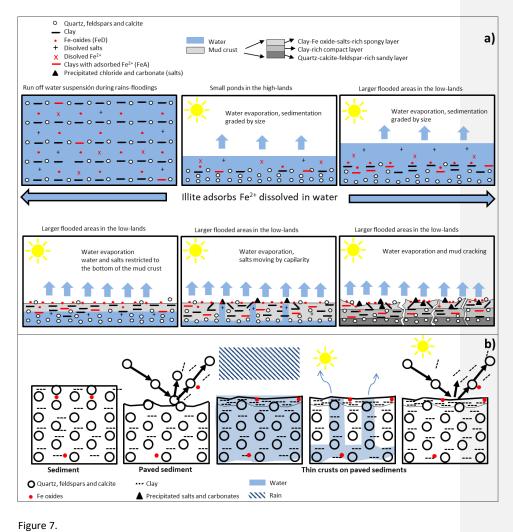




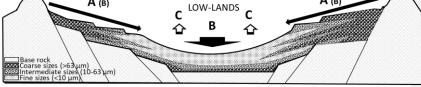
- 1119 Figure 5.





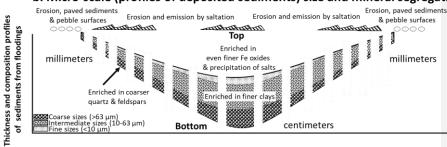






A: Washout, erosion and sporadic flooding with deposition of coarser sediments enriched in quartz and feldspars B: Flooding and deposition of finer sediments enriched in clays and Fe oxides

C: Evaporation and deposition of fine clays and readily exchangeable Fe oxide, salt crystallization in upper layers



b. Micro-scale (profiles of deposited sediments) size and mineral segregation

c. Higher dust emissions (high Fe oxide and clay) in low-lands with thicker & finer deposited sediments



a+b+c= Emitted dust might be markedly enriched in clays and Fe oxides compared to the parent sediments/soils

1150 1151	
1152	Figure 8.
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<u></u>	<u>icie-size</u> disti									
e ce		Nº of		MDPSD ≤ 63 μm						
burface Type	Location	samples	Full range	>63 to 2000 µm						
5			Mean of medians ± sd [Min,Max]							
	Mean	12	221 ± 64 [132,355]	32 ± 9.3 [20,46]	252 ± 65 [142,364]					
Dunes	High-Land	3	212 ± 27 [195,243]	45 ± 1.3 [44,46]	259 ± 22 [243,284]					
Dui	Erg Smar	4	286 ± 49 [244,355]	32 ± 8.1 [25,41]	295 ± 52 [238,364]					
	L'Bour	5	174 ± 45 [132,244	27 ± 7.4 [20,36]	214 ± 76 [142,332]					
	Mean	12	113 ± 79 [20, 320]	15 ± 3.7 [7.7,19]	308 ± 146 [146,635]					
ists	High-Land	3	94 ± 5 [89,99]	18 ± 1.1 [17,19]	219 ± 28 [187,238]					
Crusts	Erg Smar	8	131 ± 89 [21,320]	13 ± 3.4 [7.7,17]	362 ± 151 [193,635]					
	L'Bour	1	20 ± NA [NA,NA] 15 ± NA [NA,NA]		146 ± NA [NA,NA]					
Ś	Mean	11	74 ± 48 [19,152]	237 ± 71 [146,387]						
Paved Sediments	High-Land 0		NA	NA	NA					
Paved edimen	Erg Smar	8	68 ± 46 [19, 148]	17 ± 7.0 [11,33]	240 ± 43 [167,320]					
Š	L'Bour	3	90 ± 61 [29,148]	18 ± 7.1 [13,26]	230 ± 137 [146,387]					
<u>s</u>	Mean	7	70 ± 45 [20,147]	18 ± 5.1 [15,29]	175 ± 58 [129,302]					
Sediments	High-Land	1	97 ± NA [NA,NA]	18 ± NA [NA,NA]	149 ± NA [NA,NA]					
edin	Erg Smar	2	115 ± 45 [83,147]	22 ± 11 [15,29]	229 ± 104 [155,302]					
Š	L'Bour	4	40 ± 23 [20,68]	17 ± 0.79 [16,17]	155 ± 21 [129,178]					
				FDPSD						
	Mean	12	219 ± 70 [128,312]	24 ± 13 [9.0,46]	247 ± 72 [145,355]					
Dunes	High-Land	3	250 ± 73 [169,312]	41 ± 6.8 [33,46]	290 ± 77 [205,355]					
Dui	Erg Smar	4	263 ± 32 [239,308]	20 ± 6.2 [13,25]	279 ± 33 [238,319]					
	L'Bour	5	166 ± 61 [128,272]	16 ± 7.5 [9.0,26]	195 ± 68 [145,310]					
	Mean	12	37 ± 77 [2.7,272]	9.8 ± 3.6 [3.6,16]	196 ± 76 [119,389]					
Crusts	High-Land	3	124 ± 132 [20,272]	13 ± 1.1 [12,14]	251 ± 121 [162,389]					
Crt	Erg Smar	8	7 ± 3 [2.7,10]	7.9 ± 2.5 [3.6,11]	183 ± 44 [130,236]					
	L'Bour	1	17 ± NA [NA,NA]	16 ± NA [NA,NA]	119 ± NA [NA,NA]					
ts	Mean	11	21 ± 26 [2.3,78]	13 ± 4.8 [8.2,21]	157 ± 36 [120,221]					
Paved Sediments	High-Land	0	NA	NA	NA					
Pa	Erg Smar	8	18 ± 24 [5.9,78]	12 ± 4.6 [8.2,21]	169 ± 34 [129,221]					
S	L'Bour	3	29 ± 33 [5.3,67]	14 ± 6.0 [8.3,20]	122 ± 2.2 [120,124]					
ts	Mean	7	19 ± 11 [5.8,39]	14 ± 3.9 [7.7,19]	128 ± 9.6 [117,144]					
nen	High-Land	1	12 ± NA [NA,NA]	9.9 ± NA [NA,NA]	133 ± NA [NA,NA]					
Sediments	Erg Smar	2	22 ± 23 [5.8,39]	13 ± 8.1 [7.7,19]	126 ± 13 [117,135]					
5	L'Bour	4	19 ± 6.3 [13,28]	15 ± 1.3 [13,17]	128 ± 11 [122,144]					

Table 1. Full range, $<63\mu$ m and >63 to 2000 μ m mean diameter, standard deviation, min., max. and for Minimally dispersed particle sizeparticle-size distribution and fully dispersed particle sizeparticle-size distribution.

Table 2. Mineral results from samples and type of sample. In type of samples, C: Crust, PS: Paved sediment, S: Sediment, D: Dune. In Loc (Location), ES: Erg Smar, LB: L'Bour, HL: High-lands. Sme: Smectite, Mca: Mica/Illite, Kln: Kaolinite, Chl: Chlorite, Plg: Palygorskite, Qtz: Quartz, Cal: Calcite, Dol: Dolomite, Hl: Halite, Gp: Gypsum, Mc: Microcline, Ab: Albite and anorthite, Hem: Hematite, Gt: Goethite. <0.1 indicates below limit of detection.

Type Loc Qtz Mc Ab Cal Dol Sme Mca Kln Chl Plg HI Gp Hem Gt C ES 55 2,6 4,8 20 3,3 <0.1 11 <0.1 1,2 <0.1 <0.1 1,2 <0.1 <0.1 1,2 <0.1 <0.1 1,2 <0.1 <0.1 1,2 <0.1 <0.1 1,2 <0.1 <0.1 1,2 <0.1 <0.1 <0.1 1,2 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1
C ES 55 2,6 4,8 20 3,3 <0.1 11 <0.1 1,2 <0.1 <0.1 1,2 <0.1 1,1,2 <0.1 1,2 <0.1 1,2 <0.1 1,2 <0.1 1,2 <0.1 1,2 <0.1 1,2 <0.1 1,2 <0.1 1,2 <0.1 1,2 <0.1 1,2 <0.1 0.2 1,2 <0.1 0.2 1,2 <0.1 0.2 1,2 <0.1 0.2 0.1 1,2 <0.1 0.2 0.1 0.1 0.2 0.1 0.3 <0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 0.2 0.1 <th0.2< th=""> <th0.1< th=""> <th0.2< th=""></th0.2<></th0.1<></th0.2<>
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$
C ES 36 2,2 10 21 2,7 <0.1 15 10,0 1,4 <0.1 <0.1 0,20 1,2 <0. C ES 32 1,7 3,3 29 3,4 <0.1
C ES 32 1,7 3,3 29 3,4 <0.1 10 17 2,2 0,20 <0.1 <0.1 0,24 1,5 C ES 38 3,7 4,7 18 6,2 <0.1 14 9,0 1,3 0,14 3,5 0,14 0,95 <0.
C ES 38 3,7 4,7 18 6,2 <0.1 14 9,0 1,3 0,14 3,5 0,14 0,95 <0.
C ES 50 5,5 5,5 14 2,8 <0.1 12 7,9 1,3 <0.1 <0.1 <0.1 0,21 0,8
C LB 50 13 5,1 12 3,6 <0.1 8,1 5,7 0,46 <0.1 <0.1 <0.1 0,92 <0.
C HL 63 6,9 6,8 12 2,2 <0.1 4,5 3,9 0,19 <0.1 <0.1 <0.1 0,11 0,4
C ES 45 3,7 3,2 26 3,2 <0.1 11 5,4 1,8 0,21 <0.1 <0.1 <0.1 0,1
C ES 30 2,6 3,4 35 2,5 0,57 8,8 14 1,4 1,5 0,14 <0.1 0,14 0,1
C HL 60 3,7 5,5 11 0,98 <0.1 5,7 3,4 0,97 0,19 8,1 0,21 0,41 0,6
C HL 54 4,7 3,9 7,1 1,79 <0.1 5,4 3,3 0,60 <0.1 16 2,0 0,22 0,6
S ES 35 1,8 4,1 24 5,6 <0.1 17 8,3 2,2 <0.1 1,1 0,19 1,1 <0.
S ES 67 6,6 5,1 10 2,1 <0.1 3,0 3,6 0,64 <0.1 1,1 <0.1 0,42 0,2
S LB 51 4,6 7,9 15 4,2 <0.1 8,9 6,6 0,89 <0.1 <0.1 <0.1 <0.1 0,8
S LB 57 2,7 7,8 16 3,8 <0.1 9,6 1,9 0,49 <0.1 <0.1 <0.1 0,93 <0.
S LB 57 3,4 5,4 18 3,2 <0.1 6,5 3,5 2,1 <0.1 <0.1 <0.1 0,33 0,6
S HL 67 3,2 5,3 13 1,7 0,13 4,3 3,2 0,20 0,20 <0.1 <0.1 <0.1 0,9
<u>S LB 51 3,4 5,1 21 3,3 <0.1 8,5 4,5 2,2 0,15 <0.1 <0.1 0,66 0,5</u>
PS ES 44 3,0 5,7 15 3,1 <0.1 16 11 1,5 <0.1 <0.1 <0.1 0,35 0,6
PS ES 44 2,2 5,4 22 4,7 <0.1 13 6,8 0,54 <0.1 <0.1 <0.1 1,1 <0.
PS ES 55 2,3 5,4 24 3,6 <0.1 7,8 0,84 0,28 0,17 <0.1 <0.1 0,98 <0.
PS ES 40 5,3 4,7 20 4,3 <0.1 13 10 1,1 <0.1 0,29 0,77 0,2
PS ES 67 8,8 8,7 8,9 1,8 <0.1 3,1 0,38 0,30 <0.1 <0.1 <0.1 0,30 0,2
PS LB 48 5,5 4,0 16 4,3 <0.1 11 8,7 1,3 0,13 <0.1 <0.1 1,1 <0.
PS ES 61 3,5 6,3 12 3,6 <0.1 7,9 3,6 0,78 0,16 <0.1 <0.1 0,41 0,3
PS ES 46 9,1 9,0 14 3,3 0,29 6,6 8,8 1,0 0,42 <0.1 <0.1 0,42 0,6
PS ES 48 2,3 7,3 22 3,7 <0.1 8,1 6,3 1,3 <0.1 <0.1 0,16 0,17 1,1
PS LB 61 4,0 8,1 13 3,3 <0.1 4,3 4,2 1,2 <0.1 <0.1 <0.1 0,16 0,6
PS LB 51 3,6 4,4 22 3,1 <0.1 8,5 4,3 2,0 0,17 <0.1 <0.1 0,68 0,5
D ES 80 7,1 7,0 3,1 0,69 <0.1 1,4 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1
D ES 65 14 8,3 4,1 0,90 <0.1 4,8 1,9 <0.1 <0.1 <0.1 <0.1 <0.1 <0.1 0,3
D LB 73 7,0 11 6,6 0,31 <0.1 1,2 0,19 0,23 <0.1 <0.1 <0.1 0,11 0,3
D ES 89 2,5 3,0 2,0 <0.1 <0.1 0,69 1,4 <0.1 <0.1 <0.1 <0.1 0,45 0,6
D ES 65 12 5,4 11 1,3 0,13 2,7 2,1 0,38 <0.1 <0.1 <0.1 0,12 0,3
D LB 64 5,0 6,8 10 5,0 <0.1 3,1 4,3 0,50 <0.1 <0.1 <0.1 <0.1 <0.1 0,6
D LB 76 4,1 6,7 6,6 0,52 <0.1 2,6 2,2 0,21 <0.1 0,28 <0.1 0,25 0,2
D LB 77 3,9 6,7 7,5 0,53 0,26 1,5 1,6 0,49 <0.1 0,34 <0.1 0,13 0,5
D LB 57 11 14 7,5 1,8 <0.1 3,4 3,0 0,74 <0.1 <0.1 <0.1 0,16 0,3
D HL 85 4,8 4,0 3,7 0,33 <0.1 1,1 0,22 0,51 <0.1 <0.1 <0.1 <0.1 0,6
D HL 82 9,2 3,3 2,8 0,17 0,15 1,2 0,67 <0.1 <0.1 <0.1 <0.1 0,20 0,2
D HL 77 6,9 7,3 4,5 0,34 <0.1 1,7 1,3 0,39 <0.1 <0.1 <0.1 <0.1 0,4

			Carbo	nates		Clays				Salts		Fe-oxides		
	Qtz	Feld	Cal	Dol	Mca	Chl	Sme	Plg	Kln	Tot.clay	Gp	Hal	Hem	Gt
Bulk	58	9.5	15	2.4	6.4	1.0	0.1	0.2	3.8	11	0.2	8.1	0.5	0.5
Clay Ye Claquin	5	NA	6	NA	89	NA	NA	NA	NA	≈89	NA	NA	NA	NA
Clay Ye Journet	8	3	18	NA	67	NA	NA	1	3	≈71	NA	NA	NA	NA
Clay Fl Claquin	12	NA	11	NA	77	NA	NA	NA	NA	≈77	NA	NA	NA	NA
Clay Fl Journet	NA	NA	NA	NA	98	NA	NA	1	1	≈100	NA	NA	NA	NA
Clay classic Drâa	17	7.1	8.9	0.5	23	9.9	1.2	1.0	22	57	NA	NA	0.7	5.2
<10µm FD Drâa	23	4.7	19	2.4	19	4.7	0.4	0.2	14	38	NA	NA	2.2	1.8
Silt Ye Claquin	58	31	8	NA	NA	NA	NA	NA	NA	NA	2	NA	1	NA
Silt Ye Journet	43	21	20	NA	9	6	NA	NA	NA	15	NA	NA	1	NA
Silt Fl Claquin	30	38	29	NA	NA	NA	NA	NA	NA	NA	2	NA	NA	NA
Silt Fl Journet	39	19	12	NA	19	10	NA	NA	NA	29	NA	NA	1	NA
Silt classic Drâa	30	8	12	4.9	19	6.4	0.3	0.1	13	39	NA	NA	0.2	0.6
Silt FD Drâa	39	8.0	23	5.0	12	2.8	0.2	<0.1	7.5	23	NA	NA	1.2	0.7

Table 3. Mineralogy of specific soils according to Claquin et al. (1999) and Journet et al. (2014) and comparison with the one obtained in this study for six selected samples. Bulk, clay and silt fractions mineralogy (obtained from texture fractionation) and <10 μ m and silt (10-63 μ m) fractions mineralogy using fully dispersed separation. All content is in mass %.

Fl: Fluvisol sediment type; Ye: Yermosol; Qtz: Quartz; Feld: Feldspars; Cal: Calcite; Dol: Dolomite; Mca: Mica/illite; Chl: Chlorite; Sme: Smectite; Plg: Palygorskite; Kln: Kaolinite; Gp: Gypsum; Hal: Halite; Hem: Hematite; Gt: Goethite; FD: fully dispersed.