

1 **Variability in sediment particle size, mineralogy, and Fe mode of occurrence**  
2 **across dust-source inland drainage basins: The case of the Lower Drâa Valley,**  
3 **Morocco**

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53 **Abstract**

54 The effects of desert dust upon climate and ecosystems depends strongly on its particle size and size-  
55 resolved mineralogical composition. However, there is very limited quantitative knowledge on the particle  
56 size and composition of the parent sediments along with their variability within dust source regions,  
57 particularly in dust emission hotspots. The Lower Drâa Valley, an inland drainage basin and dust hotspot  
58 region located in the Moroccan Sahara, was chosen for a comprehensive analysis of sediment particle size  
59 and mineralogy. Different sediment types samples (n=42) were collected, including paleo-sediments,  
60 paved surfaces, crusts, and dunes, and analysed for particle size distribution (minimally and fully dispersed  
61 samples) and mineralogy. Furthermore, Fe sequential wet extraction was carried out to characterize the  
62 modes of occurrence of Fe, including Fe in Fe (oxyhydr)oxides, mainly from goethite and hematite, which  
63 are key to dust radiative effects, the poorly crystalline pool of Fe (readily exchangeable ionic Fe and Fe in  
64 nano-Fe-oxides), relevant to dust impacts upon ocean biogeochemistry, and structural Fe. Results yield a  
65 conceptual model where both particle size and mineralogy are segregated by transport and deposition of  
66 sediments during runoff of water across the basin, and by the precipitation of salts, which causes a  
67 sedimentary fractionation. The proportions of coarser particles enriched in quartz is higher in the high-  
68 lands, while that of finer particles rich in clay, carbonates, and Fe-oxides, is higher in the low-land dust  
69 emission hotspots. There, when water ponds and evaporates, secondary carbonates and salts precipitate,  
70 and the clays are enriched in readily exchangeable ionic Fe, due to sorption of dissolved Fe by illite. The  
71 results differ from currently available mineralogical atlases and highlight the need for observationally-  
72 constrained global high-resolution mineralogical data for mineral-specified dust modeling. The obtained  
73 dataset represents an important resource for future evaluation of surface mineralogy retrievals from  
74 spaceborne spectroscopy.

75 **Keywords:** Aeolian, desert, sediments, dust, mineralogy, iron, dust modelling.

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## 96 1. Introduction

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

118 Dust is emitted mostly from arid inland drainage basins (Prospero et al., 2002; Goudie &  
119 Middleton, 2006; Bullard et al., 2011; Querol et al., 2019). These basins encompass different  
120 sedimentary environments, many of which are potentially dust emission hotspots, including  
121 unconsolidated aeolian deposits, endorheic depressions, and fluvial and alluvial dominated  
122 systems (Bullard et al., 2011). Consolidated or compacted fine sediments in the form of crusts  
123 and paved sediments, for instance on ephemeral lake beds, can also be important dust emitting  
124 surfaces when loose sand size sediments provided by adjacent sand dunes are available (Stout,  
125 2003). These sand particles are efficiently mobilised by wind and blast the consolidated surface  
126 breaking the sediment aggregates and releasing dust (Shao et al., 2011).

127 Models developed to simulate the atmospheric dust cycle and its impact on climate represent  
128 dust emission, transport, interactions with radiation and clouds, and removal by wet and dry  
129 deposition (Ginoux et al., 2001; Zender et al., 2004; Perez et al., 2011, Klose et al., 2021). The  
130 characterization of dust sources and hotspots is one of the crucial aspects for representing dust  
131 mobilisation in models. Initially, models used vegetation cover as a criterion to identify potential  
132 dust sources (e.g. Tegen and Fung, 1994). Satellite retrievals subsequently showed that the most  
133 prolific sources occupy a smaller fraction of arid regions (Prospero et al., 2002; Ginoux et al.,  
134 2012). These so-called hotspots or “preferential sources” are found within enclosed basins,  
135 where easily eroded soil particles accumulate after fluvial erosion and transport from the  
136 surrounding high-lands. The implementation of preferential source functions in global models  
137 based on topography (Ginoux et al., 2001), hydrology (Tegen et al. 2002; Zender et al. 2003),  
138 geomorphology (Bullard et al., 2011), or satellite proxies (Prospero et al., 2002; Ginoux et al.,  
139 2012), has significantly improved the skill of models by approximately locating large-scale

140 natural sources. However, models are not able yet to capture the small-scale spatial and  
141 temporal variability in emissions apparent from observations. Models assume relatively  
142 homogeneous soil properties due to the lack of data, while there can be significant  
143 heterogeneity. Some studies have provided small-scale understanding on the role of  
144 geomorphology and sedimentology upon dust emissions (Bullard et al., 2011; Baddock et al.,  
145 2016). For instance, Bullard et al. (2011) developed a conceptual model of how different  
146 geomorphologic surfaces affect the intensity and temporal variability in dust emissions.

147 While it is key to understand dust hotspot locations and emission intensity, climate impacts by  
148 dust also depend upon its mineralogy (Alshemmari et al., 2013). Dust is a mixture of different  
149 minerals including quartz, clay minerals (mica/illite, kaolinite, palygorskite, chlorite/clinoclone  
150 and smectite/montmorillonite), feldspars (albite/anorthite and orthoclase), carbonate minerals  
151 (mainly calcite and dolomite), salts (mainly halite and gypsum), Fe-oxides and hydroxides  
152 (mostly goethite and hematite) and other oxides or hydroxides of Ti, Mn and Al (Caquineau et  
153 al, 1998; Claquin et al., 1999; Al-Ghadban et al., 1999; Cattle et al., 2002; Formenti et al., 2008;  
154 Nickovic et al., 2012; Alshemmari et al., 2013, Scheuvens et al., 2013; Journet et al., 2014; Scanza  
155 et al., 2015; Subramaniam et al., 2015; Doronzo et al., 2016; Al-Dousari et al., 2018, 2019, 2020;  
156 Ito & Wagai, 2017; Querol et al., 2019). The relative abundances, size, shape, and mixing state  
157 of these minerals influence the effect of dust upon climate. For instance, the absorption of solar  
158 radiation by dust depends upon the iron oxide content (Sokolik and Toon, 1999; Reynolds et al.,  
159 2014, Di Biagio et al., 2019), ice nucleation in mixed-phase clouds is highly sensitive to the  
160 amount of K-feldspar and quartz (Boose et al., 2016b; Harrison et al., 2019), and the  
161 bioavailability of iron in dust depends upon its iron mineralogy and speciation (Shi et al., 2012).  
162 Recent studies have shown that cloud pH is controlled in great part by calcite from dust (Grider  
163 et al., 2023). Furthermore, Ca controls heterogeneous reactions of acids on the surface of dust,  
164 which ultimately affect O<sub>3</sub> production (Bauer et al., 2004; Paulot et al., 2016). According to the  
165 geological, geomorphological and climate (weathering) patterns of the desert regions, the type,  
166 and proportions of minerals might greatly vary (Caquineau et al, 1998, Claquin et al., 1999;  
167 among others). For instance, Sahelian dust is composed mainly of quartz, kaolinite and hematite,  
168 the Middle Eastern dust is dominated by quartz and carbonates (Al-Dousari, 2018, 2019, 2020),  
169 and in North-eastern China and the Sahara dust mica/illite, kaolinite, quartz and carbonates  
170 prevail (Shen et al., 2009; Claquin et al., 1999).

171 Despite the potential importance of dust mineralogical variations, climate models typically  
172 assume dust composition as globally uniform, which is partly due to the limited knowledge of  
173 the composition of the parent sources at global scale. The few models that explicitly represent  
174 dust mineralogical composition (e.g., Scanza et al., 2015; Perlwitz et al., 2015, Li et al., 2021;  
175 Gonçalves Ageitos et al., 2023) use global atlases of soil type and the relation of this variable to  
176 soil mineralogy. This relation is inferred using massive extrapolation from a limited amount of  
177 mineralogical analyses, particularly in dust hotspots, ancillary information on soil texture and  
178 colour, and a number of additional assumptions (Claquin et al., 1999; Journet et al., 2014).  
179 Conventionally, the particle size of dust is characterized by measuring two particle size ranges,  
180 i.e. clay (<2 µm) and silt (2-63 µm) linked to FAO (Food and Agricultural Organization of the  
181 United States) soil texture datasets based on measurements following wet sieving, a technique  
182 that disperses (breaks up) the mineral aggregates found in the undisturbed parent soil into  
183 smaller particles (Chatenet et al., 1996). Furthermore, the samples that underpin these atlases

184 consider the first 10-15 cm of soil sediment, which is much deeper than the thin layer that is  
185 relevant to wind erosion and dust emission, and mineralogy is normally analysed after removing  
186 organic matter with hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), which can partially dissolve carbonate minerals.

187 The assumed relationship between mineralogy and soil type in these atlases neglects the role of  
188 geomorphology and sedimentology affecting the formation of different dust-emitting surface  
189 sediments, such as crusts, and paved sediments. This study provides a comprehensive analysis  
190 of the variability in particle size, mineralogical composition and Fe mode of occurrence of  
191 sediments collected across the Lower Drâa Valley, an inland drainage basin and prolific dust-  
192 source located in the north-western border of the Saharan desert in southern Morocco (Figure  
193 1). The data collection was carried out during a dust field campaign in September 2019 in the  
194 context of the FRontiers in dust minerAlOGical coMposition and its Effects upoN climaTe  
195 (FRAGMENT) project. Based on the analysis of the results a conceptual model is proposed that  
196 links formation processes of potential dust-emitting sediments to their particle size distribution  
197 (PSD) and mineralogy across the basin.

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## 199 **2. Methodology**

### 200 **2.1 The FRAGMENT field campaign and the study area**

201 The sediment samples analysed in this study were collected during a field campaign that took  
202 place in September 2019 in the Lower Drâa Valley, west of M'Hamid, between the Erg Chigaga  
203 and L'Bour (Figure 1a), a dry inland drainage basin where dust emission is frequent as evidenced  
204 by satellite data (Ginoux et al. 2012) (Figure 1b). The region lies where the Sahara Desert begins,  
205 to the south of the Atlas Mountain, near the Algerian border, in the Drâa River Basin. Preliminary  
206 results from the Earth Surface Mineral Dust Source Investigation, EMIT, (Green et al., 2020) show  
207 the presence of a complex regional mineralogy with fine-grained goethite, hematite (with  
208 substantial nano-sized hematite), gypsum sulphate salts in the lowlands (depressions) and  
209 Illite/muscovite, with local outcrops of carbonates in the study area (Figure 1c). The EMIT  
210 mineral maps show that the study area is representative of the larger area.

211 The campaign was conducted in the framework of the FRAGMENT project, in which distinct  
212 desert dust hotspot regions are being characterised to better understand the size-resolved dust  
213 emission and composition for different meteorological and soil conditions. The aim of  
214 FRAGMENT is to better understand dust emission, its mineralogical composition and the effects  
215 of dust upon climate, by combining field measurements, laboratory analyses, remote and in situ  
216 spectroscopy, theory and modelling. FRAGMENT field campaigns consist of intensive sediment  
217 sampling and meteorological and airborne dust measurements in one specific location, along  
218 with sediment sampling across the broader basin. The intensive meteorological and airborne  
219 dust measurements were performed in the dry lake L'Bour and are analysed in e. g., González-  
220 Florez et al., 2022; Panta et al., 2022; Yus et al., in prep. Here the focus is on the sediment  
221 sampling across the basin.

222 The study area records very low annual precipitation (80 mm) and extremely variable droughts  
223 interrupted by extreme floodings (Berger et al., 2021). The Drâa River was anthropogenically  
224 dried in this area mostly due to the construction of El Mansour Eddahbi dam in 1972 (near  
225 Ouarzazate). The Jbel Hassan Brahim range reaches the highest altitude in the area (840 m.a.s.l.),  
226 while the Drâa River is the lowest point (570 m.a.s.l.). The study region corresponds to a low

227 relief alluvial system, unarmored and unincised according to Bullard et al. (2011). Rains are  
228 scarce, but sometimes they concentrate in the mountains (high-lands) and even more  
229 sporadically they can directly affect the area during convective storms, creating flash floods with  
230 a high sediment load canalised by torrents or wadis, such as wadi Latache (high-lands) (Figure  
231 1a), which flood flat areas. In specific areas across the basin, highly sediment-loaded waters can  
232 be shortly ponded on the way to Drâa River in small depressions, such as dry Lake Iriki, Erg Smar  
233 or L'Bour (low-lands) (Figure 1a), among other areas, along the floodplain. Dunes are  
234 concentrated in small flat areas, near depressions, where, after wind erosion, sediment can be  
235 dragged and be entrapped by the very scarce and low vegetation.

## 236 **2.2 Sediment sampling**

237 The sampled sediments include paleo-sediments (hereafter named sediments), paved  
238 sediments, crusts, and dunes, according to the classification by Watt & Valentin (1992) and  
239 Valentin & Bresson (1992). Paved sediments result from cyclic drying and aeolian erosion of the  
240 surface of paleo-sediments and range from 0.5 to 2 cm of vertical depth. Crusts ranged from 0.1  
241 to 2 cm of vertical depth and two types are differentiated: i) thin depositional crusts formed as  
242 result of the deposition of sediments from running water during floods, and ii) thicker  
243 sedimentation crusts resulting from the sedimentation and drying of highly sediment-loaded  
244 waters in ponded areas of different sizes. The difference between paved sediments and crusts  
245 is mostly the period of formation. The former can date up to thousands of years ago, while the  
246 latter was formed recently. However, crust might have finer sediments because these are  
247 formed by ponding. Sediments are below the crusts (not exposed to the atmosphere) and dunes  
248 are aeolian deposits. A 50 cm<sup>2</sup> inox steel shovel was used to sample surfaces (first top cm),  
249 sediments (below surface, from 1 to 5 cm in the vertical depth) and dunes (from surface to 5  
250 cm). Coordinates, type of sample, surroundings description, and any other important  
251 information were registered. A total of 42 sediment samples were obtained, including crusts  
252 (12), dunes (12), paved sediments (11) and sediments (7) (Figure 2) from different locations in  
253 the Drâa River Basin (Figure S1). These were considered representative because the study  
254 focuses on sediments (not deposited dust) and one basin.

## 255 **2.3 Sample treatment**

256 To analyse mineral-size fractionation (<10 and 10-63 µm), a fully dispersed size fractionation  
257 using MilliQ-grade water and shaking the samples was applied prior to separation for 12-24 h.  
258 First, samples were subjected to 250, 63 and 10 µm sieves to obtain the <63 and <10 µm  
259 fractions. Due to availability, the smallest opening size of the sieve was 10 µm. Sonic sieving was  
260 applied for 60 s at maximum sustainable power for 3 min in every sieve. Finally, subsequent  
261 drying at 80 °C was applied to recover the solid fraction from the suspension.

## 262 **2.4 Analysis**

### 263 **2.4.1 Particle size distribution and texture**

264 The particle size analysis was carried out for fully (natural aggregates totally dispersed) and  
265 minimally (natural aggregates minimally dispersed) dispersed PSD to obtain the fully dispersed  
266 particle size distribution (FDPSD) and the minimally dispersed particle size distribution (MDPSD)  
267 to evaluate (i) how aggregates and particles occur in natural conditions (MDPSD) and (ii) the  
268 distribution of single particles that form the aggregates (FDPSD). The MDPSDs were obtained

269 with laser diffraction using a Malvern Mastersizer 2000 Scirocco accessory (hereinafter,  
270 Scirocco) for minimally dispersed conditions. In this case, samples of 0.3-0.5 g of the fraction <2  
271 mm were introduced into the Scirocco vibration plate with a 2 mm aperture and 5 s measuring  
272 time. FDPDs were determined using the Malvern Mastersizer 2000 Hydro G accessory  
273 (hereinafter, Hydro) with a water suspension and ultrasound assistance for totally dispersed  
274 conditions. In that case, the samples were pre-treated following the method by Sperazza et al.  
275 (2004). The suspension was introduced into the Hydro's sample container, pumping at 1750 rpm  
276 and stirring at 500 rpm. Results were obtained in both cases using the Fraunhofer method (Etzler  
277 et al., 1997).

278 To investigate the possible occurrence of vertical segregation of the PSD (top layers are the ones  
279 that are emitting dust), 7 crust and 5 paved sediment samples were selected for vertically-  
280 resolved PSD analyses. To this end, 3 sub-samples were extracted from each sample (top,  
281 middle, and bottom sections) by scratching the surface with a cutter from top to bottom and  
282 were analysed separately. The thickness of these crusts varied between 4 to 8 mm.

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

#### 287 2.4.2 Mineralogical composition

288 Quantitative mineralogical analyses of bulk sediment samples and segregated size fractions  
289 were carried out by means of powder X-Ray Diffraction (XRD), using a Bruker D8 A25 Advance,  
290 with Cu  $K_{\alpha 1}$  radiation of 1.5405 Å wavelength, a Bragg Brentano geometry and a LynxEyeXE  
291 detector. Analysis was performed at 40 mA and 40 kV with a range of angles from 4 to 60° and  
292 angle steps of 0.019° and 10 Hz for 1 h/sample. The mineral identification was made with the  
293 EVA software package by Bruker. For quantitative analyses the method of the internal reference  
294 material by Chung (1974) was applied, with quartz as the internal reference. The ratios of  
295 intensities of the different minerals versus quartz were obtained by preparing and analysing  
296 binary mixtures of the specific minerals and quartz. The accuracy of the XRD quantitative  
297 approach was tested by analysing 16 mixtures of reference materials with known concentrations  
298 of minerals. Figure S2 summarises major results, which yield relative standard deviations versus  
299 the known contents of quartz (13 % of error), albite/anorthite (10 %), calcite (31 %), dolomite  
300 (14 %), mica/illite (29 %), kaolinite (11 %), gypsum (27 %), anhydrite (19 %), goethite (42 %),  
301 hematite (50 %).

302 For an in-depth evaluation of clay mineralogy, XRD analyses of oriented aggregates following  
303 the procedure by Thorez (1976) were carried out for the same six samples of the texture. The  
304 samples were treated for air drying (AO), glycolation (AG) and heating (AC). Mica/illite,  
305 chlorite/kaolinite, palygorskite and smectite were found in all the samples, as evidenced from  
306 the bulk XRD analysis. Calcite and dolomite were dissolved by acidifying soil suspension with a  
307 strong acid as HCl and the excess used to quantify stoichiometrically the content of carbonates  
308 using the method proposed by Horváth et al. (2005) also for the same six samples of the clay-  
309 oriented aggregates and texture.

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

### 313 2.4.3 Mode of occurrence of Fe

314 Fe is a key ingredient to climatic and biological processes affected by dust. For instance, the  
315 amount, mixing state and size of Fe-oxy/hydroxides determine the degree of absorption of solar  
316 radiation by dust (Engelbrecht et al., 2016) and the potential solubility of the dust deposited into  
317 the ocean (Shi et al., 2012). However, the XRD semiquantitative analysis for Fe-oxy/hydroxides  
318 are affected by large uncertainties due to the low concentrations (increasing relative errors) and  
319 is not sensitive to nano-Fe-oxides (Shi et al., 2012). XRD analyses were complemented by  
320 quantifying the levels and mode of occurrence of Fe in the bulk samples as described in Shi et  
321 al. (2009). The method is based on a sequential extraction protocol to obtain the proportions of  
322 readily exchangeable (adsorbed) Fe ions and Fe in nano-Fe-oxides (FeA) and the amount of Fe  
323 in crystalline Fe-oxides, mainly from hematite and goethite (FeD) in the samples. 30 mg of  
324 Arizona Test Dust (ATD; ISO 12103-1, A1 Ultrafine Test Dust; Powder Technology Inc.) were used  
325 to test the accuracy of the method and extractions were done with 15 ml of extractant solution.  
326 For total Fe content (FeT), a two-step wet acid digestion method developed by Querol et al.  
327 (1993, 1997) and a coal fly ash (1633b) standard sample were used to test accuracy. The 1633b  
328 gave 7.5 % with a standard deviation of 0.14 % for total Fe (reference content of 7.8 % of Fe),  
329 while ATD gave 0.076 % with a standard deviation of 0.002 % of FeA and 0.49 % with a standard  
330 deviation of 0.07 % for FeD + FeA (reference content of 0.067 % of FeA and 0.41 % of FeD).  
331 Furthermore, by subtraction, the contents of structural Fe ( $FeS = FeT - (FeA + FeD)$ ) were  
332 obtained, corresponding to the Fe fraction as elemental Fe into the structure of minerals other  
333 than Fe-oxides, such as illite or other Fe-bearing minerals. Furthermore, the FeD contents were  
334 converted stoichiometrically to hematite ( $Fe_2O_3$ ) and goethite ( $FeO(OH)$ ) by using the  
335 hematite/goethite proportions from XRD.

### 336 2.4.5 Electron microscopy of crust and paved sediment sections

337 The PSD, mineralogy and morphology of crust and paved sediments can vary along the vertical  
338 profile, especially in crusts where progressive sedimentation and subsequent evaporation leads  
339 to inter-layering of sediments with different properties. For that purpose, crust and paved  
340 sediment sections were impregnated with epoxy resin, cut, and polished with diamond paste  
341 for microscopy analysis. The polished samples were coated with graphite before analysis with a  
342 JEOM JSM-7001F SEM-EDX Scanning Electron Microscope (SEM).

## 343 **3. Results and discussion**

### 344 **3.1 Regional variability**

#### 345 3.1.1 Particle size distribution

346 The PSDs of the samples collected across the basin were analysed to detect possible trends or  
347 size segregation patterns from high- to low-lands for the different types of sediment. The mean  
348 median diameter values of each group of sediments provided in this section represent the mean  
349 and standard deviation of the median diameters. Because the PSDs are generally bi-modal, other  
350 PSD metrics can be found in Table 1, including the maximum, minimum and mean of the median



351 diameters for different types of sediments, location, PSD type (MDPSD and FDPS), and size  
352 fraction (full range, <63  $\mu\text{m}$  and >63 to < 2000  $\mu\text{m}$ ).

353 MDPSDs, excluding dune samples, show a major mode centred around 100  $\mu\text{m}$  in diameter and  
354 a secondary one between 2 to 20  $\mu\text{m}$  (Figure 3a; Table 1). FDPSD's also show two modes at 5  
355 and 100  $\mu\text{m}$  (Figure 3b; Table 1). The MDPSDs and FDPSDs of dune samples are very similar with  
356 a main mode centred around 150  $\mu\text{m}$  and a secondary small one at 5  $\mu\text{m}$  (30 times lower) (Figure  
357 3c and d). Crust samples show the largest fine (0-5  $\mu\text{m}$ ) fraction in MDPSD, followed by paved  
358 sediments and sediments (Figure 3e). FDPSDs show a similar trend but with a larger proportion  
359 of fine particles compared to MDPSD (Figure 3f).

360 The mean median diameter of the MDPSDs (Figure 4a), excluding dune samples, is  $88\pm 63$   $\mu\text{m}$ ;  
361 and that of the FDPSDs, is  $27\pm 51$   $\mu\text{m}$  (Figure 4a). Therefore, aggregates are about 3 times coarser  
362 than individual mineral particles. As expected, dunes were coarser than other types of  
363 sediments, with a mean median diameter of  $219\pm 70$   $\mu\text{m}$  of the FDPSDs, which is very similar to  
364 that of the MDPSDs (Figure 4b). The mean median diameters of MDPSDs are  $70\pm 48$ ,  $74\pm 45$  and  
365  $113\pm 79$   $\mu\text{m}$  for sediments, paved sediments and crusts, respectively (Figure 4c); whereas the  
366 mean diameters of FDPSDs are  $19\pm 11$ ,  $21\pm 26$  and  $37\pm 77$   $\mu\text{m}$  for sediments, paved sediments  
367 and crusts respectively, about 3 to 4 times finer (Figure 4d).

368 The spatial variation of the mean diameter of the FDPSDs (Figure 5) shows coarser crusts (>40  
369  $\mu\text{m}$ ) close to the high-land areas, and finer crusts (<40  $\mu\text{m}$ ) near the Drâa River, likely due to  
370 flooding (causing transport and deposition of fine sediments, especially in the low-lands) caused  
371 during scarce and intensive rains. For paved sediments, sediments and dunes, spatial PSD trends  
372 were not evident, with mean median diameters ranging from 10 to 120, 10 to 40 and 120 to 300  
373  $\mu\text{m}$ , respectively, randomly located across the basin (Table 1).

374 According to the size classification by Valentin & Bresson (1992) and using the FDPSD data  
375 (Figure S3), dune samples can be classified as sand, loamy sand, and sandy loam; sediments as  
376 silt loam and loam; paved sediments as sandy loam, loam and silt loam; and crusts as sandy  
377 loam, loam, silty clay loam and silt loam. As shown in Figure S3 and due the higher transport  
378 potential of clays during rain episodes, and their accumulation during ponding, crusts tend to be  
379 further enriched in clay fractions, especially in low-lands, compared to paved sediments and  
380 sediment samples (see section 3.4).

### 381 3.1.2 Mineralogical composition

382 Here the mineralogy of samples collected across the basin is described to detect possible trends  
383 or mineral segregation patterns from high- to low-lands for the different types of sediment. The  
384 mineralogical composition (mass % composition of the bulk sample) of dunes, crusts, paved  
385 sediments and sediment samples is summarised in Table 2. Dunes show a homogeneous  
386 mineralogy across the study area, with mineral abundances of  $74\pm 9.7$  % quartz,  $5.8\pm 2.9$  %  
387 calcite,  $6.7\pm 3.6$  % microcline,  $6.9\pm 3.1$  % albite/anorthite,  $4.1\pm 2.3$  % clay minerals,  $1.0\pm 1.4$  %  
388 dolomite,  $0.38\pm 0.26$  % goethite and  $0.12\pm 0.11$  % hematite and trace amounts of halite and  
389 gypsum (<0.1 %) (Figure 6). In comparison to dunes, crusts are depleted in quartz ( $48\pm 11$  %) and  
390 feldspars ( $5.0\pm 2.1$  % albite/anorthite and  $4.4\pm 3.1$  % microcline), and enriched in clay minerals  
391 ( $17\pm 8.0$  %), calcite ( $19\pm 8.0$  %), dolomite ( $3.0\pm 1.3$  %) and Fe-oxides ( $0.24\pm 0.28$  % hematite and  
392  $0.42\pm 0.56$  % goethite) (Figure 6). The content of gypsum ( $0.23\pm 0.56$  %) and halite ( $2.9\pm 5.1$  %) is  
393 higher than in dune samples, but variability is large because it depends on the exact point of

394 crust sampling. Paved sediments have a similar mineralogy than crusts, for quartz ( $51\pm 8.7\%$ ),  
395 calcite ( $17\pm 4.9\%$ ), clay minerals ( $16\pm 7.3\%$ ), albite/anorthite ( $6.3\pm 1.8\%$ ), microcline ( $4.5\pm 2.5\%$ ),  
396 dolomite ( $3.5\pm 0.79\%$ ), hematite ( $0.34\pm 0.25\%$ ), and goethite ( $0.38\pm 0.38\%$ ), but with lower  
397 content of gypsum ( $<0.1\%$ ) (Figure 6). Sediments are also similar to paved sediments and crusts  
398 with a mean quartz content ( $55\pm 11\%$ ), calcite ( $17\pm 4.6\%$ ), clay minerals ( $14\pm 6.8\%$ ),  
399 albite/anorthite ( $5.8\pm 1.5\%$ ), microcline ( $3.7\pm 1.6\%$ ), dolomite ( $3.4\pm 1.3\%$ ), hematite ( $0.28\pm 0.37$   
400 %) and goethite ( $0.37\pm 0.32\%$ ). Trace amounts of gypsum ( $<0.1\%$ ) and halite ( $0.32\pm 0.55\%$ ) were  
401 also found in sediments (Figure 6).

402 In comparison with the bulk sediment, the fully dispersed silt fraction ( $10\text{--}63\ \mu\text{m}$ ) shows a lower  
403 amount of quartz ( $35\pm 6.4\%$ ) and feldspars ( $7.4\pm 2.5\%$ ), a higher content of carbonates ( $25\pm 5.2$   
404 %), clays ( $22\pm 10\%$ ) and hematite ( $1.07\pm 0.38\%$ ) and a similar content of goethite ( $0.61\pm 0.32\%$ ).  
405 In the fully dispersed  $<10\ \mu\text{m}$  sieved fraction, the amount of quartz ( $23\pm 5.2\%$ ) and feldspars  
406 ( $4.7\pm 1.1\%$ ) is two times lower than in the bulk sediments. The fraction of carbonates remains  
407 similar ( $21\pm 9.0\%$ ) and the content of clays increases substantially ( $38\pm 9.8\%$ ) compared to the  
408 bulk and silt-size mineralogy. The Fe-oxide content increases by about a factor two for both  
409 hematite ( $2.2\pm 2.0\%$ ) and goethite ( $1.8\pm 1.2\%$ ). Table 3 compares the mineralogical results in the  
410 clay and silt size ranges, both with the fully dispersed separation and the pipette methods,  
411 against the corresponding values provided by the available global mineralogical atlases of  
412 Claquin et al. (1999) and Journet et al. (2014), which assume the sample locations to be either  
413 fluvisols or yermosols in terms of soil type. In the silt-size fraction, similar contents of quartz,  
414 total clay, mica/illite, chlorite+kaolinite, calcite and Fe-oxides, were found, but 3 times less  
415 feldspars and 5 times more dolomite. Compared to the clay-size fraction in the atlases, the  $<10$   
416  $\mu\text{m}$  fraction in this study shows larger content of quartz and feldspars (by factor of 2 to 4), a 30  
417 % lower total clay content and similar contents of calcite and Fe-oxides, which can only be partly  
418 explained by the difference in the size fraction considered ( $<10\ \mu\text{m}$  vs  $<2\ \mu\text{m}$ ) as shown by the  
419 results obtained with the pipette method. Because kaolinite and chlorite have coincident  
420 spacing at  $7\ \text{\AA}$  in the XRD spectra, in current atlases these minerals may be confounded, whereas  
421 in this study chlorite was quantified separately by identifying other minor peaks in the spectra.  
422 This is relevant as both minerals are very different in terms of chemical composition. In this  
423 study, minor concentrations of dolomite and traces of smectite and palygorskite, were also  
424 detected. The large differences in the silt-size feldspar content may be largely due to the lack of  
425 data and coarse assumptions used in current atlases.

426 Table S1 in the supplemental material compares the silt+clay and sand proportions and the  
427 mineral contents of the crusts from this study in Morocco with those from deposited dust in  
428 different arid regions of the world. The FD-PSD data from this study evidences that 72% of the  
429 particles in the crusts fall in the clay+silt fraction ( $<63\ \mu\text{m}$ ), while 28% in the sand size-range.  
430 This is close to the average value (74 and 26%, respectively) calculated from the existing studies  
431 on deposited dust. Concerning the mineralogy, the crusts of this study are enriched in clays and  
432 depleted in carbonate minerals and feldspars compared with the average of the mineralogy of  
433 deposited dust (Table S1).

434 In this analysis of trends in mineralogy from the high-lands to the low-lands, all sample types  
435 except dunes, were considered. The low-lands, such as L'Bour and Erg Smar, are enriched in clay  
436 minerals ( $17\pm 9.6$  and  $14\pm 3.4\%$ , respectively) compared to the high-lands ( $9.1\pm 0.97\%$ ) (Figure  
437 6). Mica/illite is the most common clay mineral reaching mean contents of  $9.1\pm 4.8$ ,  $8.1\pm 2.0$  in

438 Erg Smar and L'Bour, respectively, and  $5.0\pm0.70$  % in the high-lands. Kaolinite reaches  $7.2\pm5.4$ ,  
439  $4.9\pm2.1$  and  $3.5\pm0.30$  % and clinocllore  $1.7\pm1.8$ ,  $1.3\pm0.67$  and  $0.49\pm0.38$  %, respectively.  
440 Smectite and palygorskite were detected only in trace amounts ( $<0.1$  %) in most samples, with  
441 only palygorskite at Erg Smar and high-lands reaching a mean content of  $0.34\pm0.58$  and  
442  $0.15\pm0.06$  %, respectively. The same trend is found for calcite ( $24\pm13$ ,  $16\pm3.1$  and  $11\pm2.7$  %, Erg  
443 Smar, L'Bour and high-lands), dolomite ( $5.0\pm5.1$ ,  $3.6\pm0.51$  and  $1.7\pm0.50$  %, at Erg Smar, L'Bour  
444 and high-lands) and Fe-oxides ( $0.78\pm1.4$ ,  $0.37\pm0.43$  and  $0.08\pm0.04$  % for hematite at Erg Smar,  
445 L'Bour and high-lands and  $0.42\pm0.51$ ,  $0.39\pm0.35$  and  $0.32\pm0.21$  % for goethite at Erg Smar, L'Bour  
446 and high-lands) being steeper for hematite than goethite (Figure 6). Quartz follows an opposite  
447 trend, increasing towards the high-lands ( $42\pm18$ ,  $53\pm5.0$  and  $61\pm5.4$  %, at Erg Smar, L'Bour and  
448 high-lands, respectively) (Figure 6). Albite/anorthite and microcline do not show a clear trend,  
449 with  $5.5\pm2.3$ ,  $5.9\pm1.8$  and  $5.4\pm1.2$  % at Erg Smar, L'Bour and high-lands, and  $3.4\pm2.4$ ,  $5.0\pm3.4$  and  
450  $4.6\pm1.7$  %, respectively (Figure 6). Salt concentrations peak randomly and depend on very local  
451 scale conditions, being higher at concave areas where ponding is favoured (see section 3.4). The  
452 mean content of halite is  $1.0\pm2.2$ ,  $<0.1$  and  $4.0\pm7.7$  % at Erg Smar, L'Bour and high-lands and  
453 that of gypsum is  $0.18\pm0.35$ ,  $<0.1$  and  $0.15\pm0.92$  %, respectively (Figure 6).

454 A soft crust occurred on the surface of several dunes (Figure 2). The PSD and mineralogical  
455 analysis of the crust and the underlying sands did not reveal significant differences. Pye & Tsoar  
456 (2015) reported that surface hardening of dunes is due to the scavenging and deposition of clays  
457 from suspended dust in light rains and by cementation of sand particles (meniscus) by  
458 precipitation of carbonates and silica in the retained interstitial pore water. In both cases the  
459 potential variability caused by the slight increase of this clay and carbonate/silica cementation  
460 is obscured by variations in the bulk mineralogy.

### 461 **3.2 Vertical segregation in crust and paved sediments**

462 The examination of thin vertical cross-sections provides insight into how particle size and  
463 mineral composition vary within the top few  $\mu\text{m}$  or mm of the surface. These differences are  
464 relevant to the mineralogy and PSD of newly emitted dust.

465 The MDPSDs of crust sections (top, middle and bottom) are very similar, with two modes of  
466 occurrence at 5-7 and 200  $\mu\text{m}$  (Figure S4a). Yet, while the FDPDSs show similar two modes at 1-  
467 5 and 100  $\mu\text{m}$  for the top and middle sections and a second mode at 300  $\mu\text{m}$  for the bottom  
468 section (Figure S4b). The MDPSD mean median diameter of the 7 crust profiles reach  $25\pm25$ ,  
469  $54\pm80$  and  $25\pm26$   $\mu\text{m}$  for the top, middle and bottom sections, respectively, while FDPDS means  
470 are  $9.4\pm9.4$  and  $11\pm9.5$   $\mu\text{m}$  in the top and middle sections and  $94\pm145$   $\mu\text{m}$  in the bottom one  
471 (Figure S4c and d). Therefore, during the initial stages of ponding, coarser particles are deposited  
472 first while finer particles remain suspended (see section 3.4) in the later stages before  
473 evaporation of the water. Even some oxides, carbonates and salts may precipitate in the top  
474 layers of the crust as water evaporates and the ionic strength increases.

475 No vertical PSD segregation is observed in paved sediments, but some top sections analysed  
476 show enrichment in coarser fractions in FDPDS (the median diameter increases from bottom  
477 and middle sections ( $14\pm6.8$  and  $12\pm5.8$   $\mu\text{m}$ ) to the top section ( $23\pm28$   $\mu\text{m}$ )), likely due to  
478 preferential erosion of finer fractions through sandblasting (see section 3.4).

479 The mean levels of quartz and feldspars are enriched in the bottom sections of the crusts ( $46\pm 17$   
480 and  $8.7\pm 4.6$  %, respectively) compared to the middle ( $38\pm 11$  and  $8.3\pm 2.5$  %) and top sections  
481 ( $41\pm 12$  and  $6.9\pm 2.2$  %) due to the higher quartz content of the coarse fraction that is deposited  
482 first (see section 3.4). The content of clay minerals, salts and Fe-oxides is similar in the top  
483 ( $20\pm 7.2$ ,  $<0.1$  and  $3.3\pm 1.9$  %, respectively), middle ( $21\pm 5.0$ ,  $<0.1$  and  $2.8\pm 1.6$  %), and bottom  
484 sections ( $19\pm 9.1$ ,  $<0.1$  and  $1.9\pm 1.0$  %). Carbonate minerals are relatively homogeneous, but  
485 slightly enriched in the middle and top sections ( $29\pm 9.7$  and  $28\pm 7.9$  %, respectively) compared  
486 to the bottom section ( $24\pm 8.4$  %). This can arise from both detrital carbonate particles and  
487 precipitation from high ionic strength waters that are ponded and dried in the low-lands.

488 The mineral composition of the paved sediment profiles differs slightly from that of crust  
489 profiles. This is because the latter are affected by particle segregation during transport and  
490 subsequent sedimentation. The top section of the paved sediment profiles has more quartz than  
491 in the middle and bottom sections ( $44\pm 8.1$ ,  $38\pm 5.7$  and  $40\pm 9.8$  %, respectively), whereas  
492 feldspars decrease from the bottom and middle to the top sections ( $9.1\pm 4.2$ ,  $9.3\pm 2.2$  and  $6.9\pm 2.7$   
493 %). Carbonates and clay are relatively homogeneous ( $26\pm 4.9$ ,  $26\pm 2.0$  and  $25\pm 4.2$  % for  
494 carbonates, and  $22\pm 8.4$ ,  $23\pm 9.2$  and  $25\pm 4.9$  % for clays, respectively). The slight depletion of  
495 minerals in the top section may be due to sandblasting, which tends to erode the fine fraction  
496 of the surface over time (see section 3.4). Fe-oxides are more present in the top section than in  
497 the middle and bottom sections ( $2.1\pm 0.47$ ,  $2.0\pm 0.38$  and  $1.7\pm 0.27$  %, respectively) and the  
498 presence of salts is very low ( $<0.1$  % for all sections).

### 499 **3.3 Mode of occurrence of Fe**

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

503 The mean FeT content of bulk crusts, paved sediments and sediments was found to be  $3.6\pm 0.71$ ,  
504  $3.4\pm 0.47$ , and  $3.2\pm 0.44$  %, respectively, while bulk dunes had a much lower FeT content  
505 ( $2.0\pm 0.33$  %). Fe-speciation studies reveal that FeS percentage from FeT (FeS/FeT) is the  
506 prevailing Fe mode of occurrence ( $67\pm 2.4$ ,  $69\pm 3.0$ ,  $68\pm 2.7$  and  $73\pm 5.9$  % in crusts, paved  
507 sediments, sediments and dunes, respectively), followed by FeD percentage from FeT (FeD/FeT)  
508 ( $31\pm 2.3$ ,  $29\pm 3.0$ ,  $30\pm 3.0$ ,  $26\pm 5.8$  %), and FeA percentage from FeT (FeA/FeT) ( $1.9\pm 0.55$ ,  $1.7\pm 0.56$ ,  
509  $1.4\pm 0.55$  and  $1.0\pm 0.54$  %). These results show that FeT is very similar between crusts, paved  
510 sediments and sediments while FeT in dunes is depleted by almost 50 %. Compared to Shi et al.  
511 (2011) samples from northwestern Africa, the samples of this study are depleted in total iron  
512 ( $4.7$  % FeT from Shi et al. (2011)), quite similar in FeS ( $67$  % from Shi et al. (2011)), similar in FeD  
513 ( $33$  % from Shi et al. (2011)) and much higher in FeA ( $0.43$  % from Shi et al. (2011)).

514 The mean FeT content in the basin is similar in Erg Smar ( $3.6\pm 0.27$  %) and L'Bour ( $3.2\pm 0.66$  %)  
515 compared to high-lands ( $3.0\pm 0.24$  %). The ratio FeA/FeT was slightly higher at Erg Smar ( $1.9\pm 0.53$   
516 %) but similar at L'Bour and high-lands ( $1.3\pm 0.44$  and  $1.5\pm 0.47$  %, respectively). This is probably  
517 due to the preferential accumulation of exchangeable and nano-Fe-Oxides (FeA) in the low-  
518 lands, where flooding results in red-water ponds and red surfaces after drying. Subsequently,  
519 highly concentrated ionic Fe is trapped in the last stages of ponding, and nano-Fe-oxides may

520 precipitate during drying. Once the ponded is dried, the crusts of the low-lands tend to have a  
521 reddish patina (see section 3.4). However, a slightly higher mean FeD/FeT of  $33\pm 2.4$  % is  
522 obtained in the high-lands compared to  $31\pm 2.7$  and  $29\pm 2.4$  % at L’Bour and Erg Smar,  
523 respectively. The FeS/FeT mean content is slightly lower at the high-lands ( $65\pm 2.5$  %) compared  
524 to Erg Smar and L’Bour ( $69\pm 2.6$  and  $68\pm 2.6$  %, respectively).

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

529 The proportions of FeD + FeA are higher in crusts, probably due to preferential transport of non-  
530 FeS to the low-lands and the trapping of Fe ions (FeA) by clay adsorption during ponding, and  
531 the formation of nanosized Ferrihydrite ( $\text{Fe}_{4-5}(\text{OH},\text{O})_{12}$ ). This readily exchangeable Fe has very low  
532 impact on radiative forcing but a high impact in Fe fertilisation of oceans during dust events  
533 (Gobler et al., 2001), as ionic Fe adsorbed by clays and nano-Fe-oxides are easily released in  
534 water solutions. The correlation of FeS, FeD and FeA with FeT is linear, with coefficients of  
535 determination ( $R^2$ ) reaching 0.96, 0.89 and 0.67 for FeS, FeD and FeA respectively (Figure S5).  
536 Thus, when increasing total Fe content all modes of occurrence of Fe increase, but the increase  
537 is preferentially driven by FeS, while it seems that the basin FeA segregation causes a lower  
538 correlation with FeT.

### 539 **3.4 Conceptual model for particle size and mineralogy fractionation in crusts and paved** 540 **sediments**

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

547 In the low-lands, floodwaters carrying fine sediments flood extensive flat areas, such as Erg Smar  
548 or Iriki lake. Prospero et al. (2002), Bullard et al (2011) and Ginoux et al (2012), among others,  
549 have shown that dust emissions originate from relatively small and localised areas where  
550 sediments are supplied by floodwaters, and that the occurrence of dust emissions from these  
551 areas may be partly due to the occurrence or absence of floodings. During ponding in low-lands,  
552 coarser particles deposit first and form a high sand-rich bottom layer of the crust (as described  
553 in section 3.2) (Figure 7a & 8a). Subsequently, the clay fraction deposits on top of the bottom  
554 layer until total dryness (Figure 7a & 8a) forming a second clay-rich fraction layer in the crust.  
555 However, the particle size in crust surfaces is heterogeneous (Figure S6 & S7), which can result  
556 in erodible dust-emitting sediment (heterogeneity enhances sandblasting). The finer and more  
557 easily exchangeable FeA fraction remains in suspension until the last drying stages on the most  
558 superficial layer of the crust, during drying out of the remaining ponds (as described in section  
559 3.3) (Figure 7a & 8a). During this ponding, dissolved Fe ions interact with clays in such a way that  
560 they can be adsorbed on clay surfaces according to the ionic composition of the waters (as

561 described in section 3.3) (Echeverría et al., 1998). This typical ion adsorption by clays is higher  
562 for montmorillonite than for other clays but the content of montmorillonite is low compared to  
563 illite. In this study a high correlation is obtained for FeA and illite contents (Figure S8).  
564 Furthermore, crusts contain a higher proportion of hematite(oxide)/goethite(hydroxide) in the  
565 FeD, due to the weathering with water during transport and ponding and precipitation of nano-  
566 Fe-oxides during drying.

567 After the pond drying, the continuous heating of the clay rich surface layer causes the hardening  
568 of the crust and mud-cracking, giving a 'ceramic-like' compactness to the thick crusts in the low-  
569 lands, usually with a reddish colour induced by the Fe-oxides (Figure S6a). Complete drying  
570 causes mud cracks due to loss of volume, breaking the crust into polygonal pieces, whose  
571 thickness and area depend on the amount of clay deposited. Furthermore, these concave mud-  
572 crust pieces resulting from the cracking usually have a grey-colour patch in the middle due to  
573 the superficial precipitation of salts, which together with carbonates accumulate by capillarity  
574 (see section 3.1.2) (Figure S6b). This capillary ascension and precipitation of salts (the latter  
575 being an expansive process) causes sponging and breaking of the surface layers. Thus, a third  
576 (top) layer is formed in the crusts of the low-lands, which is very easily eroded by wind because  
577 of the spongy structure and enriched in clay and readily exchangeable Fe. In some cases, in Erg  
578 Smar, an additional breaking and sponging of the third (upper layer) due to expansive clays, was  
579 observed. Both the ceramic-like compactness and the cementing of salts give the fine-clay rich  
580 crusts in the low-lands a compact pattern with coarser MDPSD compared with the high-lands  
581 where ponding is limited and very thin crusts occur. This could explain why the crusts from the  
582 low-lands have finer FDPDs and coarser MDPSDs compared to the high-lands (see section  
583 3.1.1). Also, wind erosion of the few top millimetres of these crusts may result in dust with higher  
584 contents of clay, Fe-oxide and salts compared to a 15 cm sediment profile.

585 In the high-lands, washout erosion occurs during rainfall, leading to the formation very thin  
586 crusts in reduced areas. This results in sources of dust made of very thin crusts and fields of  
587 stony surfaces with lower emission rates compared to the low-lands (Bullard et al., 2011). As  
588 illustrated in Figure S7 the surfaces of paved sediments and their thin crusts might resemble  
589 crusts profiles, but with the top section depleted on clay minerals due to preferential erosion  
590 over time, and with a very thin layer (a few micrometres) of clay minerals from the previous  
591 intact formed crust after flooding or running water. The top paved sediments are more compact,  
592 finer and have homogeneous distribution of the particle than crusts, which makes them less  
593 erodible and less likely to emit dust compared to crusts (which have heterogeneous particle-  
594 size, see section 3.2).

#### 595 **4. Conclusions**

596 This study analysed the particle size and mineralogy of dust-emitting sediments in the region of  
597 Drâa basin in Northern Africa, at the north-western fringe of the Sahara. The study aimed to  
598 compare these patterns for different types of dust-emitting sediments and their variations  
599 across the basin. The results are consistent with the conceptual models of dust emission sources  
600 in desert areas of Prospero et al. (2002) and Bullard et al. (2011), which predict higher dust  
601 emissions in the low-lands than in the high-lands. The study shows a clear size and mineralogical  
602 fractionation between paleo-sediments and low-land dust-emitting sediments, indicating that

603 collecting samples of parent paleo-sediments for particle size and mineralogy may not fully  
604 represent the dust emission hotspots.

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

620 The results of this study show that modeling mineral-speciated dust emission requires  
621 understanding of the mineralogical and particle size fractionation of accumulated sediments  
622 across inland enclosed basins. Large areas may act as sediment suppliers, while reduced areas  
623 may act as dust emitters with differences in sediment composition. Models that represent  
624 mineral-speciated dust emission and transport should be developed to properly account for  
625 these factors.

626 The results have also shown that global atlases overestimate the clay mineral content and  
627 underestimate that of quartz, feldspars, and Fe-oxides. Quartz and feldspars are overestimated  
628 and clay minerals underestimated in the silt-size fractions. Kaolinite-chlorite are not  
629 differentiated, while this study finds major differences. The classical procedure loses salts during  
630 fractionation, and Fe-oxides are detected mainly by colour without precision. This study detects  
631 dolomite, palygorskite, and smectite, and provides more precision for Fe-oxides, with the mode  
632 of occurrence of Fe in different types of samples and locations. However, the study was unable  
633 to obtain a sample below 10  $\mu\text{m}$  without losing salts in the process.

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

641 The large dam built in the Drâa River has caused the drying of this part of the basin, a reduction  
642 of vegetation and probably increased dust emissions. The region exemplifies how anthropogenic  
643 activities can promote wind erosion and represents a unique location for research on the topic.

644 Future studies may indeed explore many other aspects related to sedimentology, mineralogy,  
645 wind erosion, dust emission and anthropogenic impacts, including the study of the introduction  
646 of native plants and green belts to reduce wind erosion as has already been done in other  
647 regions (Al-Dousari et al. 2020).

648

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#### 670 **Credit authorship contribution statement**

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

677

#### 678 **Declaration of competing interest**

679 Some authors are members of the editorial board of journal ACP. The peer-review process was  
680 guided by an independent editor, and the authors have also no other competing interests to  
681 declare.

682

## 683 **5. References**

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940 **Figure captions**

941 **Figure 1.** a) Location of the study area (exact location of data measurement “star”: 29°49’30”N,  
942 5°52’25”W), near M’Hamid el Ghizlane, into the Drâa basin in S Morocco. Base layer from  
943 world imagery of Google Earth Pro v:7.3.6.9345. b) Frequency of occurrence (%) of dust  
944 optical depth above 0.2 in September, October and November between 2003 and 2016  
945 derived from MODIS Deep Blue. c) EMIT scenes  
946 emit20220903t082303\_o24606\_s001\_l2a\_rfl\_b0106\_v01 and  
947 emit20230206t101334\_o03707\_s000\_l2a\_rfl\_b0106\_v01 at 60 meters per pixel show the  
948 diversity of Fe<sup>2+</sup> and Fe<sup>3+</sup> bearing minerals (left) and the EMIT 8 phyllosilicates, carbonates,  
949 and sulphates (right). The mineral maps were produced by tetracorder 5.27c1 (Clark, 2023).  
950 There is some mapped mineralogy difference at the scene boundaries, possibly due to the  
951 changing viewing geometry, and variation in atmospheric removal between the two scenes.  
952 Cirrus clouds in the scene on the right may also be impacting derived mineralogy.

953 **Figure 2.** Images of samples collected during a field campaign near M’Hamid el Ghizlane, into  
954 the Draa Basin, S Morocco.

955 **Figure 3.** Median minimally and fully dispersed PSDs of crusts, sediments, paved sediments and  
956 dunes. (a) MDPSDs and (b) FDPDs combined from crust, sediment and paved sediment  
957 samples; (c) and (d) are MDPSDs and FDPDs for dune samples; (e) and (f) are MDPSDs and  
958 FDPDs differentiated by type of sample.

959 **Figure 4.** Boxplot of median particle-size diameters in  $\mu\text{m}$  including both fully and minimally  
960 dispersed analysis (a) for all samples combined excluding dunes and (b) for dune samples  
961 only. Also particle-size diameter in  $\mu\text{m}$  for crusts, sediment and paved sediment for (c)  
962 minimally dispersed and (d) fully dispersed results. Means median diameters for each  
963 sediment type are shown with crosses.

964 **Figure 5.** Spatial variation map with crust fully dispersed mean median particle diameter.

965 **Figure 6.** Mean mineral group content of dune, crust, paved sediment and sediment samples,  
966 and also at Erg Smar, L’ Bour and High-lands. Solid lines mark the mean content of all the  
967 samples (excluding dune samples). The dashed line divides between type and location of  
968 the samples.

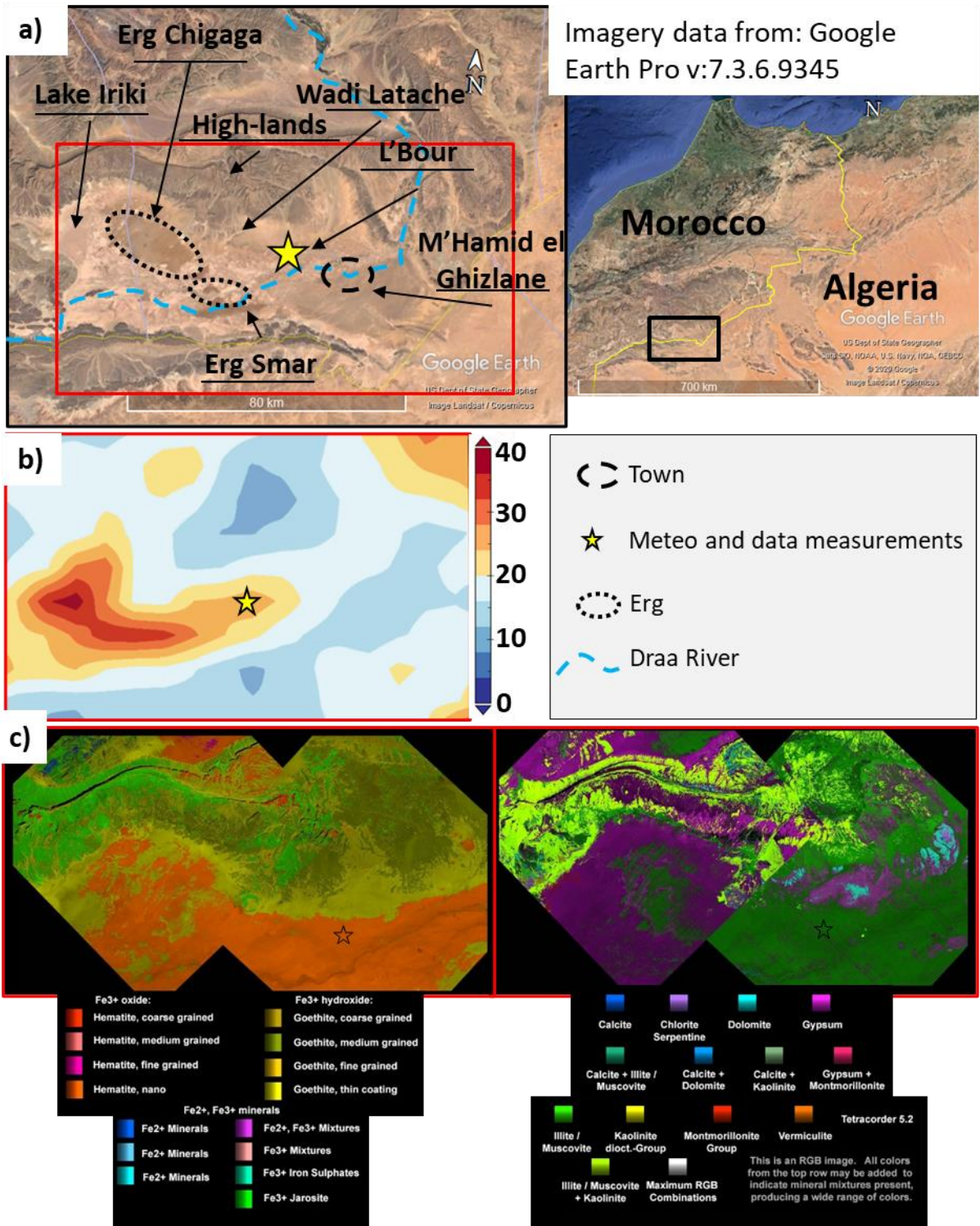
969 **Figure 7.** Schematic model of sedimentation and deposition processes in the study site from  
970 high-lands to low-lands for a) crusts and for b) paved sediments.

971 **Figure 8.** Dust emission conceptual model integrating particle-size distributions and mineralogy  
972 of dust hotspots sediments. a) Refers to the conceptual thickness and particle-size  
973 distributions along the basin, b) to the particle-size distribution and segregation of  
974 mineralogy and c) to the dust emission quantity expected depending on the place in the  
975 basin.

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Figure 1.

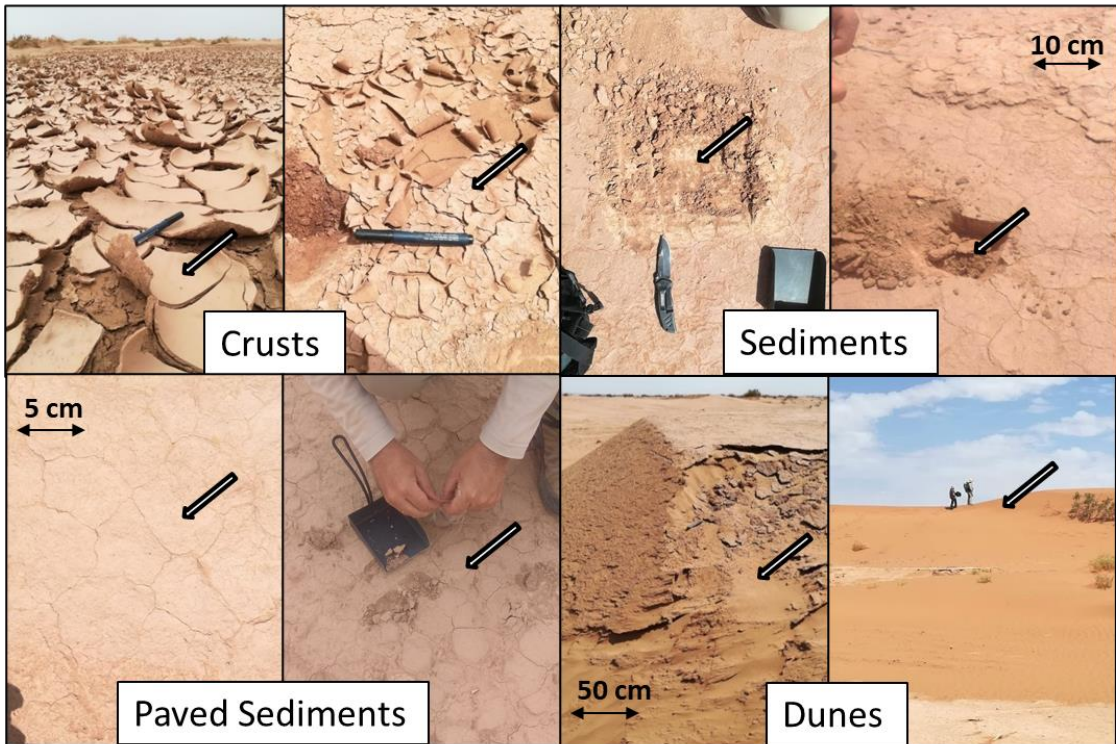
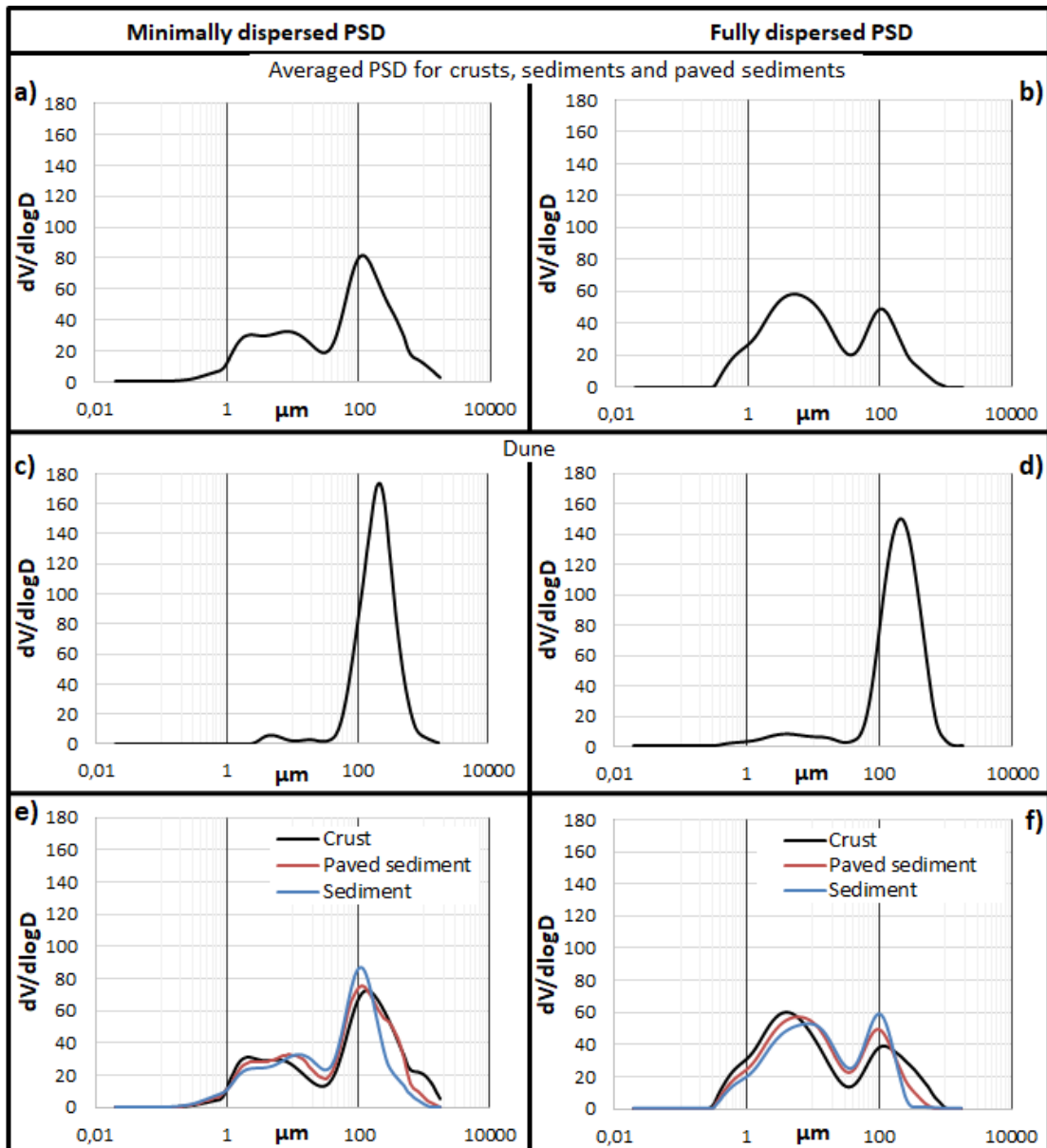


Figure 2.

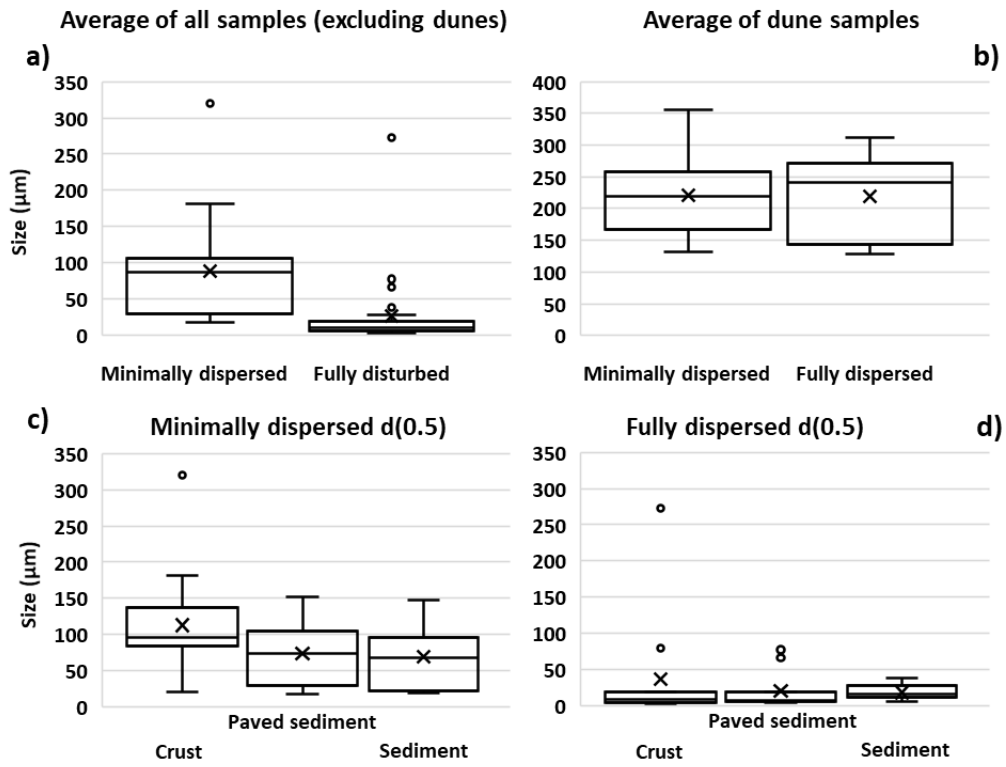
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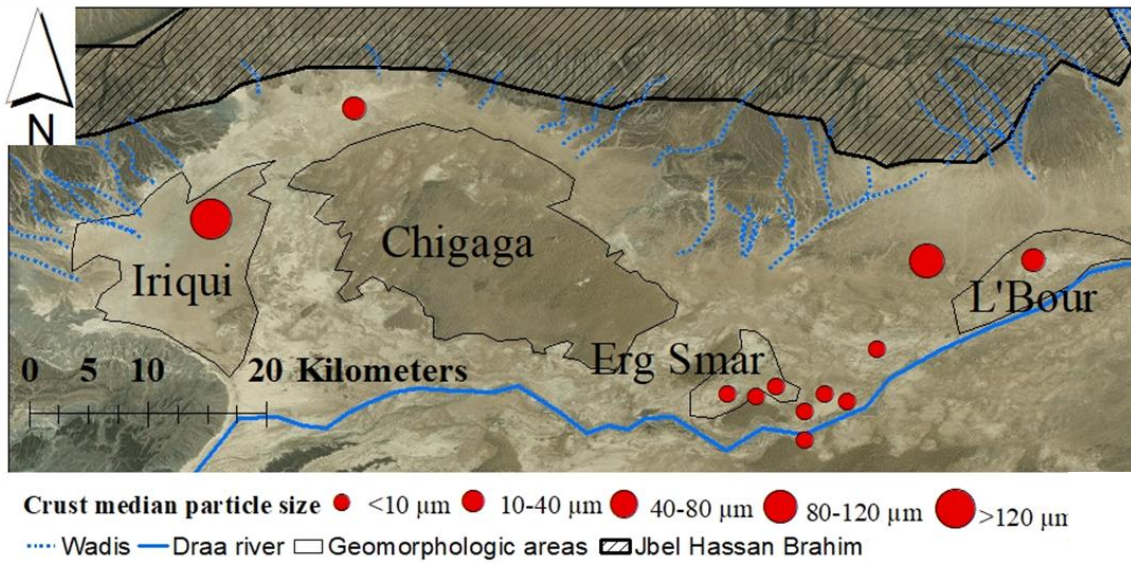


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Figure 3.



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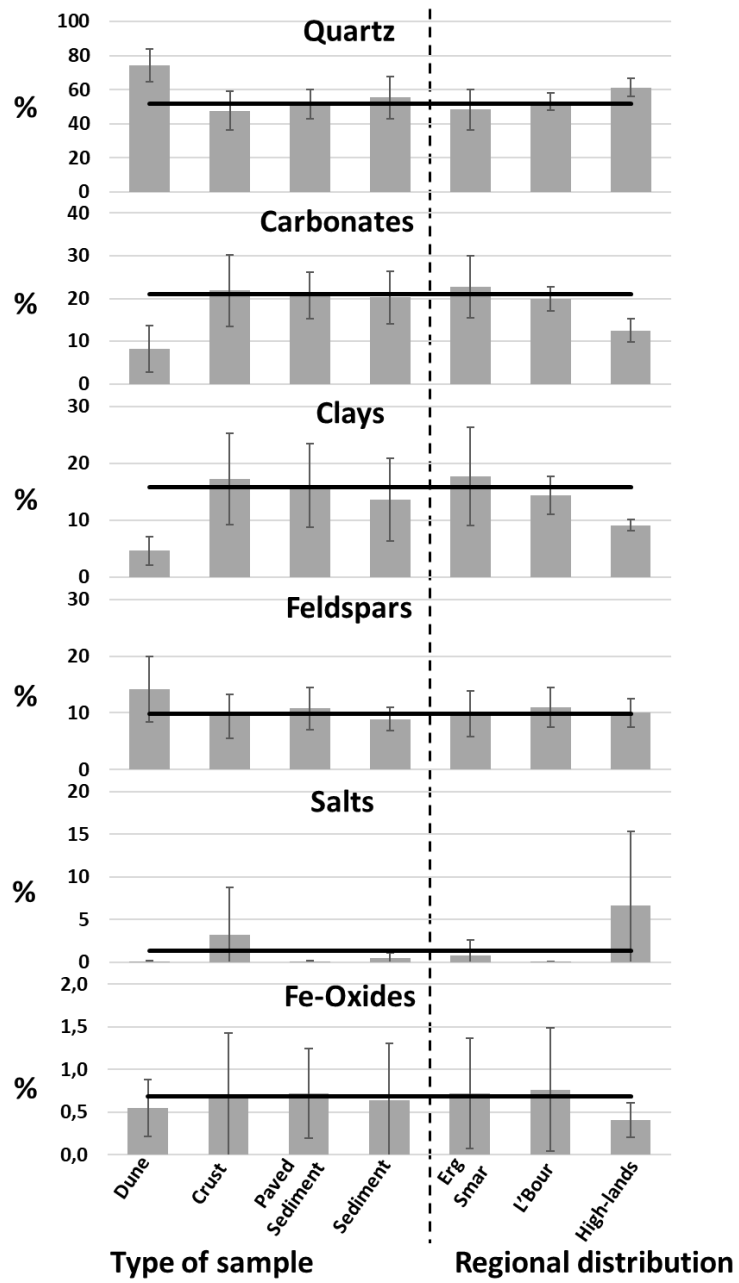
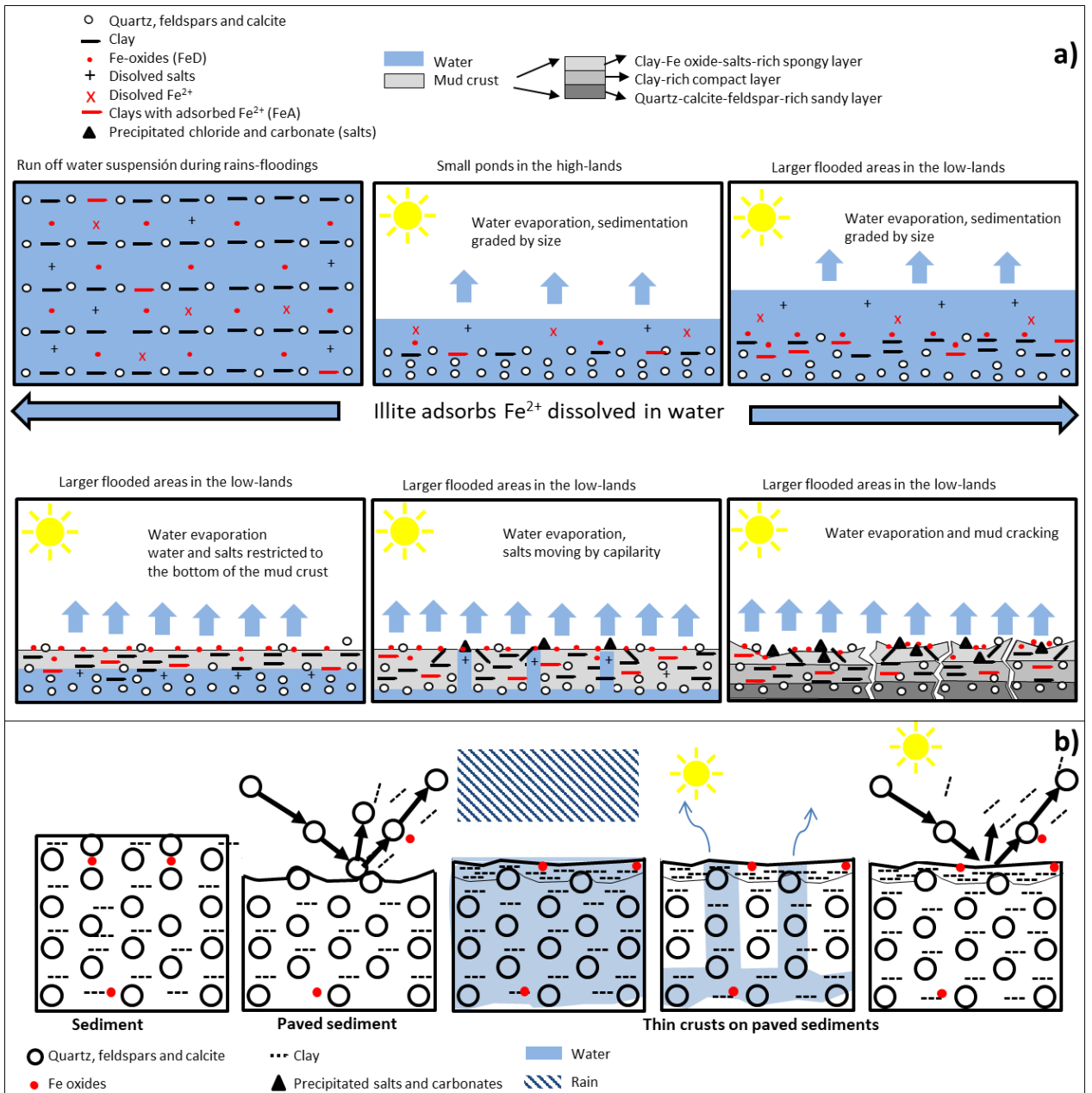


Figure 6.

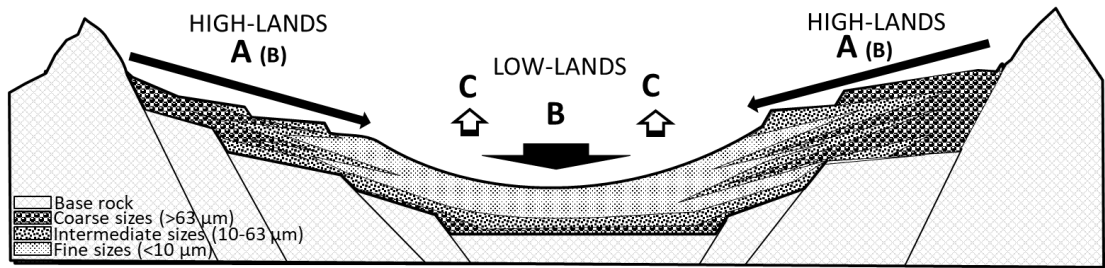
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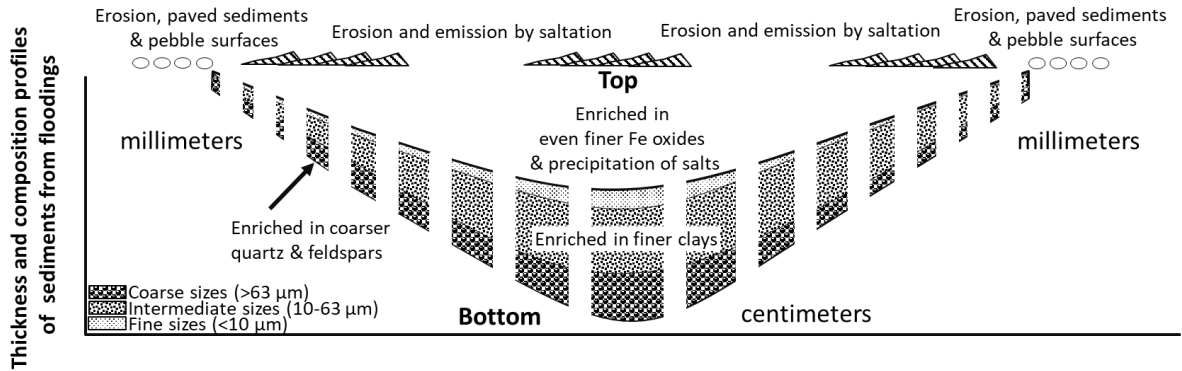
Figure 7.

**a. Macro-scale (basin) size and mineral segregation of sediments**

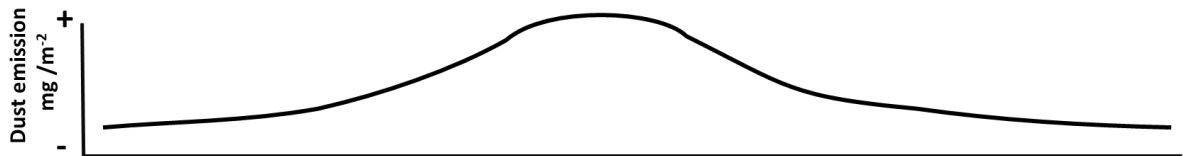


A: Washout, erosion and sporadic flooding with deposition of coarser sediments enriched in quartz and feldspars  
 B: Flooding and deposition of finer sediments enriched in clays and Fe oxides  
 C: Evaporation and deposition of fine clays and readily exchangeable Fe oxide, salt crystallization in upper 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**

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Figure 8.

Table 1. Full range, <63µm and >63 to 2000 µm mean diameter, standard deviation, min., max. and for Minimally dispersed particle-size distribution and fully dispersed particle-size distribution.

Surface Type	Location	Nº of samples	MDPSD		
			Full range	≤ 63 µm	>63 to 2000 µm
			Mean of medians ± sd [Min,Max]		
Dunes	Mean	12	221 ± 64 [132,355]	32 ± 9.3 [20,46]	252 ± 65 [142,364]
	High-Land	3	212 ± 27 [195,243]	45 ± 1.3 [44,46]	259 ± 22 [243,284]
	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]
Crusts	Mean	12	113 ± 79 [20, 320]	15 ± 3.7 [7.7,19]	308 ± 146 [146,635]
	High-Land	3	94 ± 5 [89,99]	18 ± 1.1 [17,19]	219 ± 28 [187,238]
	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]
Paved Sediments	Mean	11	74 ± 48 [19,152]	17 ± 6.7 [11,33]	237 ± 71 [146,387]
	High-Land	0	NA	NA	NA
	Erg Smar	8	68 ± 46 [19, 148]	17 ± 7.0 [11,33]	240 ± 43 [167,320]
	L'Bour	3	90 ± 61 [29,148]	18 ± 7.1 [13,26]	230 ± 137 [146,387]
Sediments	Mean	7	70 ± 45 [20,147]	18 ± 5.1 [15,29]	175 ± 58 [129,302]
	High-Land	1	97 ± NA [NA,NA]	18 ± NA [NA,NA]	149 ± NA [NA,NA]
	Erg Smar	2	115 ± 45 [83,147]	22 ± 11 [15,29]	229 ± 104 [155,302]
	L'Bour	4	40 ± 23 [20,68]	17 ± 0.79 [16,17]	155 ± 21 [129,178]
			FDPSD		
Dunes	Mean	12	219 ± 70 [128,312]	24 ± 13 [9.0,46]	247 ± 72 [145,355]
	High-Land	3	250 ± 73 [169,312]	41 ± 6.8 [33,46]	290 ± 77 [205,355]
	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]
Crusts	Mean	12	37 ± 77 [2.7,272]	9.8 ± 3.6 [3.6,16]	196 ± 76 [119,389]
	High-Land	3	124 ± 132 [20,272]	13 ± 1.1 [12,14]	251 ± 121 [162,389]
	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]
Paved Sediments	Mean	11	21 ± 26 [2.3,78]	13 ± 4.8 [8.2,21]	157 ± 36 [120,221]
	High-Land	0	NA	NA	NA
	Erg Smar	8	18 ± 24 [5.9,78]	12 ± 4.6 [8.2,21]	169 ± 34 [129,221]
	L'Bour	3	29 ± 33 [5.3,67]	14 ± 6.0 [8.3,20]	122 ± 2.2 [120,124]
Sediments	Mean	7	19 ± 11 [5.8,39]	14 ± 3.9 [7.7,19]	128 ± 9.6 [117,144]
	High-Land	1	12 ± NA [NA,NA]	9.9 ± NA [NA,NA]	133 ± NA [NA,NA]
	Erg Smar	2	22 ± 23 [5.8,39]	13 ± 8.1 [7.7,19]	126 ± 13 [117,135]
	L'Bour	4	19 ± 6.3 [13,28]	15 ± 1.3 [13,17]	128 ± 11 [122,144]

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	Feldspars		Carbonates		Clays					Salts		Iron Oxides	
			Mc	Ab	Cal	Dol	Sme	Mca	Kln	Chl	Plg	Hl	Gp	Hem	Gt
C	ES	55	2,6	4,8	20	3,3	<0.1	11	<0.1	1,2	<0.1	<0.1	<0.1	1,2	<0.1
C	ES	57	2,7	3,1	20	3,4	<0.1	5,2	<0.1	0,78	0,26	7,2	<0.1	0,87	<0.1
C	ES	36	2,2	10	21	2,7	<0.1	15	10,0	1,4	<0.1	<0.1	0,20	1,2	<0.1
C	ES	32	1,7	3,3	29	3,4	<0.1	10	17	2,2	0,20	<0.1	<0.1	0,24	1,3
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	<0.1
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	0,85
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	<0.1
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	0,40
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	0,18
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	0,17
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	0,60
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	0,65
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	<0.1
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	0,23
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	0,82
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	<0.1
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	0,60
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	0,90
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	0,50
PS	ES	44	3,0	5,7	15	3,1	<0.1	16	11	1,5	<0.1	<0.1	<0.1	0,35	0,64
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	<0.1
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	<0.1
PS	ES	40	5,3	4,7	20	4,3	<0.1	13	10	1,1	<0.1	<0.1	0,29	0,77	0,23
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	0,29
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	<0.1
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	0,33
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	0,69
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	0,64
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	0,53
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	0,38
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	0,37
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	0,32
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	0,65
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	0,32
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	0,61
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	0,21
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	0,50
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	0,37
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	0,69
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	0,29
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	0,41

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 µm and silt (10-63 µm) fractions mineralogy using fully dispersed separation. All content is in mass %.

	Carbonates				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

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.





