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Ultraviolet-Visible Diffuse Reflectance Spectroscopy (UV-Vis DRS), a rapid and non-destructive analytical tool for the identification of Saharan dust events in Particulate Matter filters

This is the final peer-reviewed author's accepted manuscript (postprint) of the following publication:

Published Version:

Availability:

This version is available at: <https://hdl.handle.net/11585/813573> since: 2021-03-09

Published:

DOI: <http://doi.org/10.1016/j.atmosenv.2021.118297>

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The final published version is available online at:
<https://doi.org/10.1016/j.atmosenv.2021.118297>.

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1 **TITLE**

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3 Ultraviolet-Visible Diffuse Reflectance Spectroscopy (UV-Vis DRS), a rapid and non-destructive
4 analytical tool for the identification of Saharan dust events in Particulate Matter filters

5

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31 **ABSTRACT**

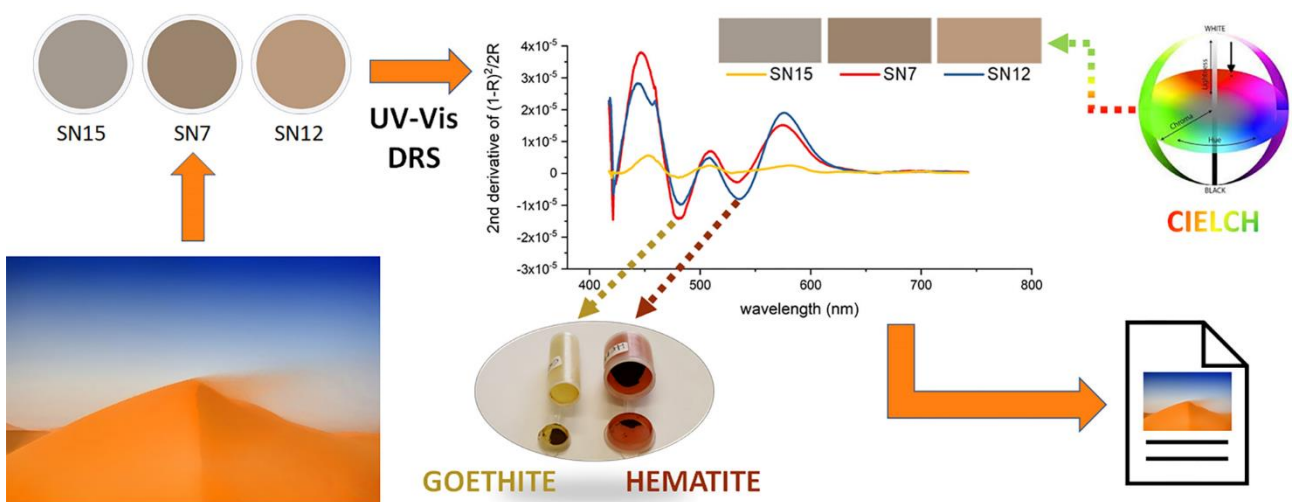
32

33 Mineral dust represents one of the main components of particulate matter (PM) in the Mediterranean
34 area. The rapid identification of Saharan dust events in PM samples is desirable and required for
35 several reasons, including their role in direct effect on climate by radiative forcing as well as their
36 adverse effects on human health.

37 For this purpose, the feasibility of UV-Vis Diffuse Reflectance Spectroscopy (UV-Vis DRS)
38 is described as a rapid, inexpensive and non-destructive method of analysis of PM filters. The method
39 developed allows to parameterize the PM filter colors and to obtain semi-quantitative data related to
40 iron oxide minerals, mainly hematite and goethite, two of the most representative minerals of Saharan
41 dust in the Mediterranean area. The obtained results were validated based on the correlation between
42 the spectrophotometric data of iron oxides from the membranes with the quantitative assessment of
43 the concentration of iron by Particle Induced X-ray Emission (PIXE). Moreover, colorimetric
44 parameterization allows setting up a classification approach for filters with potential for a posteriori
45 use of this data in the study of the optical behavior of aerosol particles in the air.

46 In this work, it is demonstrated how, as the concentration of iron mineral oxides and especially of
47 hematite increases, the extent of redness color in PM filters grows up. Therefore, this technique can
48 be extremely useful for a rapid, cheap and unambiguous identification of Saharan dust events in PM
49 filters. The diagnosis of Saharan dust events was performed on PM₁₀ filters with a strong mineral dust
50 component and demonstrated with the residence time analysis of back-trajectory ensembles, proving
51 the reliability of this non-destructive methodology.

52



53

54

55 **KEYWORDS**

56 Saharan dust, diffuse reflectance spectroscopy, iron oxide minerals, colorimetry, chemometrics,
57 residence time analysis

58 **HIGHLIGHTS**

59

- 60 - Rapid and non-destructive technique for particulate matter filter analysis
- 61 - Quick identification of filter colour
- 62 - Semi-quantitative analysis of iron oxide minerals
- 63 - Fast detection of Saharan dust events
- 64 - Determination of particular events and/or emission sources of particulate matter

65

66 **1. INTRODUCTION**

67 Mineral dust uplifted into the atmosphere by the wind in arid and semi-arid regions of North Africa
68 are often transported across thousands of kilometers including both the northernmost European
69 territory and/or the American continent (e.g., Middleton & Goudie, 2002). Overall the Mediterranean
70 basin owing to proximity and average circulation patterns, is directly and widely affected by these
71 phenomena throughout the year with events whose intensity and frequency are object of extensive
72 research (Brattich et al., 2015a; Brattich et al., 2015b; Cabello et al., 2016; Cuevas et al., 2017; Cusack
73 et al., 2012; Israelevich et al., 2012; Riccio et al., 2009; Tositti et al., 2014). The frequency and
74 intensity of Saharan Dust outbreaks are presently reported as increasing due to the effect of global
75 warming (Middleton & Goudie, 2002; Soleimani et al., 2020). These events can contribute to an
76 increase of PM₁₀ levels above the limits allowed by the air quality regulations drawing attention on
77 health threatens as well as on their correct management (Diapouli et al., 2017; Krasnov et al., 2014;
78 Matassoni et al., 2011; Nava et al., 2012; Querol et al., 2019). In particular, the European Air Quality
79 Directive 2008/50/EC establishes that PM₁₀ (i.e. PM with an aerodynamic diameter less than 10 µm)
80 daily mean value may not exceed 50 µg/m³ more than 35 times in a year and that the PM₁₀ annual
81 mean value may not exceed 40 µg/m³ (EU, 2008). similarly, the World Health Organization (WHO)
82 recommends a PM₁₀ daily mean value less than 50 µg/m³ but with a considerably lower PM₁₀ annual
83 mean value of 20 µg/m³ (WHO, 2006). The European legislation allows subtracting the contribution
84 associated to natural episodes and therefore the quantification of the African dust contribution is

85 relevant in the air quality field (EEA, 2012). The relevance of this quantification is not limited to
86 legislation, however.

87 Saharan dust plays a significant role in the climate system by affecting the radiative balance of the
88 planet. Dust particles heavily modify the optical properties of the troposphere by the so-called direct
89 effect (e.g., Chin et al., 2009; Ginoux, 2017; IDSO, 1981; Littmann & Steinrücke, 1989; Sokolik &
90 Toon, 1999), while recent research has shown it plays also a fundamental role in cloud processing
91 and nucleation, i.e. by indirect effect (see for example Reicher et al., 2019). Furthermore, mineral
92 dust strongly influences the atmospheric reactivity through complex surface chemical reactions
93 (Usher et al., 2003). Mineral particles may also affect the oxidant capacity of the troposphere by
94 catalyzing ozone destruction, an important pollutant and reactive greenhouse gas (Bonasoni et al.,
95 2003; Dickerson et al., 1997; Prospero et al., 1995). Moreover, iron contained in Saharan dust plumes
96 can settle on oceanic surfaces and be a nutrient for marine phytoplankton, with beneficial results for
97 oligotrophic aquatic systems but potentially damaging the eutrophic ones (e.g., Bristow et al., 2010;
98 Molinaroli & Masiol, 2006). However, Saharan dust has been often associated with a coral decline in
99 the Caribbean region, suggesting that either mineral or microbiological components of Saharan dust
100 may reveal detrimental to especially fragile ecosystems (Garrison et al., 2003; Shinn et al., 2000).
101 This is in agreement with further, not negligible, implications linking Saharan dust transport with an
102 increase in the mortality rate and adverse health effects on the Mediterranean population (Karanasiou
103 et al., 2012; Querol et al., 2019; Stafoggia et al., 2016).

104 Common procedures to identify dust outbreaks use a combination of back-trajectory analysis, satellite
105 retrievals, and the output of dust prediction models. Collectively they provide a reasonable degree of
106 evidence though each of these tools has limitations: a back-trajectory travelling over North Africa is
107 not always associated to dust advection; satellite retrievals are limited by cloud coverage and transit
108 time; uncertainties in dust model estimates remain, due to incomplete representation of several
109 processes. All of this information is frequently combined with PM₁₀ levels or columnar aerosol

110 properties at the study site. These levels are compared with local threshold values or background
111 levels obtained by their own time series (Barnaba et al., 2017) or with PM₁₀ concentrations at a close
112 regional background site (EC Commission, 2011; Escudero et al., 2007). Saharan mineral dust
113 consists mainly of silicates, aluminum oxides, carbonates, gypsum, and iron oxides, with a specific
114 composition that depends on the geological material from its lifting place (e.g., Linke et al., 2006).
115 Chemical speciation analysis is largely used to characterize aerosol composition as a function of its
116 sources. Mineral dust is successfully identified with all the basic techniques devoted to elemental
117 inorganic analysis; X-ray emission techniques such as Particle Induced X-ray Emission (PIXE) and
118 X-ray fluorescence (XRF) are the most efficient and non-destructive ones since they allow a prompt
119 detection of the abundant geochemical components, without any demanding and costly chemical
120 processing in advance. Crustal elements like silicon (Si), aluminum (Al), titanium (Ti), calcium (Ca),
121 and iron (Fe) are successfully used to identify Saharan dust events, as reported by, e.g., Alastuey et
122 al., 2016; Formenti et al., 2010; Marconi et al., 2014; Nava et al., 2012; Rodríguez et al., 2020. In
123 most cases, elemental analysis is complemented by ion chromatography wherein the most informative
124 species associated with mineral dust is calcium ion (Ca²⁺) (Escudero et al., 2005; Flentje et al., 2015;
125 Putaud et al., 2004). This approach can be integrated by the detection of mineral species such as
126 quartz, feldspars, illite, smectite, kaolinite, chlorite, vermiculite, mica, calcite, gypsum, hematite and
127 goethite (Caquineau et al., 2002; Journet et al., 2014) requiring X-Ray Diffraction (XRD) (Menéndez
128 et al., 2007; Shao et al., 2007), and Scanning Electron Microscopy (SEM) (Menéndez et al., 2007;
129 Remoundaki et al., 2011). However, many of these analytical techniques are expensive, time
130 consuming, and require sophisticated instrumental facilities and highly skilled personnel.

131 In this article, a procedure based exclusively on Ultraviolet-Visible Diffuse Reflectance Spectroscopy
132 (UV-Vis DRS) (Torrent & Barrón, 2008) is proposed as a simple, cheap, efficient, rapid, and non-
133 destructive analysis of particulate samples collected on a membrane for Saharan Dust transport
134 diagnosis. This method is based on iron oxides, which account for approximately 2 to 7 % in weight

135 of the total amount of mineral dust (Alfaro et al., 2004; Formenti et al., 2008; Goudie et al., 2006) in
136 atmospheric aerosol and mainly consist of goethite (predominant, 52-78% of the total iron oxides)
137 and hematite (22-48 % of the total iron oxides) (Formenti et al., 2014; Lafon et al., 2006; Shi et al.,
138 2012). Since these minerals are important markers of mineral dust (Formenti et al., 2014; Lafon et
139 al., 2006; Shi et al., 2012), they can be used as proxies for Saharan dust events in locations
140 characterized by a low background of iron oxides from other more common sources, i.e. the Earth's
141 crust especially by soil resuspension or technogenic ones such as metal works. Hematite and goethite
142 are typically characterized by intense colors, which can impart a yellowish to red color to atmospheric
143 dust particles (Arimoto et al., 2002). The proposed methodology is characterized by: (a) color
144 parametrization and (b) iron mineral oxides semi-quantification of the analyzed PM filters, followed
145 by (c) chemometric tools. It can be integrated into routine sampling when no further speciation
146 analysis is needed. This procedure does not intend to replace the existing speciation methods, but to
147 support them in order to enhance and corroborate the identification of Saharan Dust transport
148 episodes.

149

150 **2. MATERIALS AND METHODS**

151 The Materials and Methods section is organized as follows:

152 (a) Subsection 2.1 describes the PM₁₀ samples used for the application and characterization of the
153 UV-Vis DRS methodology reported in this work;

154 (b) Subsection 2.2 describes exhaustively the method used, paying particular attention to the
155 instrumental configuration and analysis method of PM filters (subsection 2.2.1), how it is possible to
156 parameterize the colour of the analyzed filters (subsection 2.2.2) and obtain semi-quantitative data of
157 iron oxide minerals from sample reflectance spectra (subsection 2.2.3);

158 (c) Subsection 2.3 presents the validation of the proposed methodology. After checking the main
159 results of the proposed methodology (subsection 2.3.1), chemometric methods are used to identify
160 the PM filters that have been subjected to a Saharan dust transport event (subsection 2.3.2.1), and the
161 diagnosis obtained are assessed and confirmed by a residence time analysis of back-trajectory
162 ensembles (subsection 2.3.2.2).

163 **2.1 Particulate matter samples**

164 PM₁₀ samples analyzed in this work were collected at Sierra Nevada a high altitude site (37.096 N, -
165 3.387 W, 2550 m a.s.l.) in Southern Spain, within the framework of the Spanish national project
166 FRESA (Impact of dust-laden African air masses and of stratospheric air masses in the Iberian
167 Peninsula. Role of the Atlas Mountains, Ref: CGL2015-70741-R). The sampling station is located in
168 an area scarcely influenced by traffic and other anthropogenic sources, but strongly impacted by
169 Saharan dust incursion events due to its proximity to North Africa (Figure 1).

170 PM sampling was carried out using a high volume PM₁₀ sampler (CAV-A/mb, MCV S.A.) on a
171 weekly basis (168 h at 30 m³/h) using quartz filters (Ø 15 cm, Whatman QM-A quartz filters) from 8
172 June to 11 October 2016. Nineteen weekly PM₁₀ samples (labeled as SN1-SN19) were overall
173 obtained and processed along with three field blanks (labeled as B1-B3).

174 **2.2 The UV-Vis DRS methodology**

175 *2.2.1 Sample preparation and UV-Vis DRS analysis*

176 UV-Vis Diffuse Reflectance Spectroscopy is a widely used, basic spectrophotometric technique for
177 the analysis of powders and surfaces, requiring a negligible sample preparation (Torrent & Barrón,
178 2008). It is based on the surface dispersion of a fraction of the UV-Vis incident radiation on it. A UV-
179 Vis collimated light beam is directed with a certain angle onto the sample and, as a result, an ensemble
180 of optical processes leads to radiation reflection by the sample surface on the whole overlying

181 hemisphere. Generally, the radiation reflected by a sample can be considered as the sum of two
182 components: regular (or specular) and diffuse (or nondirectional) reflectance (e.g., Torrent & Barrón,
183 2008). Regular reflectance occurs when incident radiation hits an ideally smooth and planar surface
184 (i.e. without roughness) of the sample, and it is then reflected at an angle equal to the angle of
185 incidence (Fresnel law). Instead, diffuse reflectance is a combination of several optical phenomena,
186 such as multiple reflections, scattering and refraction, which disperse the radiation at all of the angles
187 of the hemisphere of origin of the incident radiation (Blitz, 1998). Diffuse reflectance is the most
188 informative component as it regards the physico-chemical properties and color of the surface (Sellitto
189 et al., 2008).

190 Diffuse reflectance spectroscopy of PM filters is conducted with an ordinary UV-Vis
191 spectrophotometer (in this work, a Perkin Elmer Lambda 35 UV-Vis Spectrophotometer was used)
192 equipped with a suitable accessory, known as integrating sphere, which allows the measurement of
193 all the reflection produced by the sample, which is diffused in the inner walls of the integrating sphere.
194 The analysis does not include any sample chemical preparation: to the scope a squared sample portion
195 of 1.8 cm x 1.8 cm is placed inside a flat sample holder, designed to position the sample for beam
196 irradiation at a 0° incidence angle, and lodged over the reflectance sample port of the integrating
197 sphere, a 50 mm diameter Labsphere RSA-PE-20 (Labsphere, United States). With the 0° sample
198 holder in place, any specular component of reflection from the sample is excluded from measurement,
199 since this component is directed out of the sphere through the transmittance sample entrance port.
200 This integrating sphere configuration is named 0°/diffuse (0°/d) and Figure 2 shows its operating
201 scheme. Square sample pieces were carefully cut by means of a square die-cutting tool with a side of
202 1.8 cm. The UV-Vis DRS analysis of the latter is non-destructive since the side containing the
203 particulate material is analyzed as it is, without any treatment and contact with the instrument. Firstly,
204 a Spectralon white standard (USRS-99-010-EPV, Labsphere, United States) was analyzed as a
205 reference and the instrumental autozero was performed. Then, one aliquot from each field blanks (B1-

206 B3) was prepared and B1 portion was used for the background correction. Subsequently, the analysis
207 of the remaining blank portions (B2 and B3) and the PM sampled filters was carried out. In particular,
208 in addition to the two blank portions, three different portions of each PM sampled filter were prepared
209 and analyzed in order to compute average values and standard deviations of each sample outcome
210 and, therefore, account for possible filter anisotropy. In this way, a total of 59 analyses were
211 completed for this work. Therefore, the percentage reflectance (% R), i.e. the ratio between the
212 intensity of the radiation reflected by the sample and the intensity of the total radiation reflected by a
213 white diffuse reflectance standard, was determined for each of the analyzed portions. In particular,
214 this parameter was measured as a function of the wavelength λ of the UV-Vis incident radiation (i.e.,
215 reflectance spectra were obtained), based on the following instrumental parameters: λ range = 780 -
216 380 nm; resolution = 0.3 nm; scan speed = 480 nm/min, smoothing = 2 nm, and slit = 2 nm.

217 *2.2.2 Color parametrization*

218 Sample color can be obtained by its UV-Vis reflectance spectrum. Indeed, sample color strongly
219 depends on its diffuse reflection: an object irradiated by a light source disperses part of the incident
220 radiation by diffuse reflection, which is subsequently collected by the eyes of an observer which in
221 turn act as transducers, converting the light signal into appropriate electrical impulses for the brain.
222 Ultimately, these impulses are integrated and processed by the latter, which generates the color
223 perception for the observer (Kremers et al., 2016). Therefore, color is an extremely complex and
224 subjective entity, as it is not a specific feature of the object itself but depends on many variables such
225 as the light source, the optical behavior of the object, the observer's eyes and brain, etc. Since 1931,
226 the International Commission on Illumination (CIE) has outlined guidelines to standardize color
227 perception, through the standard definition of three elements: light sources, observers, and
228 colorimetric spaces. The latter are mathematical models able to define the color of an object in a
229 rigorous manner (Ibraheem et al., 2012). One of the currently most used colorimetric space is the CIE
230 $L^*a^*b^*$ (CIELAB) (ISO-CIE 11664-4-2019), which uses three cartesian components to uniquely

231 define color sample: L^* , that indicates the CIELAB lightness in the range 0 (pure black) to 100 (pure
232 white); a^* , that indicates the CIELAB redness-greenness coordinate; and b^* , that indicates the
233 CIELAB yellowness-blueness coordinate. This colorimetric space can also be defined in polar
234 coordinates, thus obtaining the CIE $L^*C_{ab}^*h_{ab}^\circ$ space (CIELCH) (ISO-CIE 11664-4-2019), wherein:
235 L^* always indicates the CIELAB lightness; C_{ab}^* represents the CIELAB chroma, a measure of the
236 color intensity, defining how much a certain color shade is "contaminated" by gray; and h_{ab}° indicates
237 the CIELAB hue angle, whose value is expressed in degrees and describes the color tone. In
238 particular, the 0° angle represents the red color. Because of their easy interpretability, in this work
239 the mathematical definition of the colors of the analyzed portion samples was carried out employing
240 the CIELAB and CIELCH spaces, starting from the reflectance spectra obtained and using the Color
241 2.01 software (Perkin Elmer Ltd, United Kingdom). Standard Illuminant D65 was set up as a
242 representation of solar light source, according to CIE (ISO-CIE 11664-2-2020), and an observer angle
243 of 10° was set up because provide the best average spectral response in human observers (ISO-CIE
244 11664-1-2019). Average values and the standard deviations of the colorimetric parameters (L^* , a^* , b^* ,
245 C_{ab}^* , and h_{ab}°) were calculated for the blank filter (B) and the 19 samples (SN1,...SN19) starting from
246 the analyzed portions for each sample. The CIELAB average data were used for color visualization
247 through the online tool nix Color Sensor (<https://www.nixsensor.com/free-color-converter/>). In this
248 step, illuminant and reference (observer) angle have been set as D65 and 10° , respectively.

249 *2.2.3 Semi-quantification of iron oxide minerals*

250 Diffuse reflectance measurements are extremely useful for the individuation and quantification of the
251 most important sample pigments. Indeed, they exhibit attenuation in reflectance spectra due to their
252 light-absorption in specific UV-Vis wavelength ranges. Iron oxide minerals absorption is associated
253 with their electronic transitions within the $3d^5$ shell of Fe^{3+} ion triggered by UV-Vis radiation
254 (Scheinost et al., 1998) and, consequently, reflectance spectra can be useful for the assessment of
255 these mineral species. While past work on PM membranes was mainly based on the first derivative

256 of the UV-Vis reflectance (e.g., Arimoto et al., 2002; Shen et al., 2006), in this work we compute the
257 second derivative of the Kubelka-Munk (K-M) function spectra. This method has been widely used
258 for the assessment of iron oxides in soil samples (Barrón & Torrent, 1986; Fernandez et al., 1992;
259 Sellitto et al., 2009; Szalai et al., 2013) and less frequently for PM filters analysis (Lafon et al., 2006),
260 while the approach was used in several spectrophotometric applications for the most absorbing
261 aerosol component, i.e. soot (see for example Pandey et al., 2019; Petzold et al., 2004). The
262 calculation of the second derivative was performed using the Savitzky-Golay filter (Schafer, 2011),
263 an averaging algorithm that fits a polynomial to the data points and allows the calculation of a
264 derivative of this function. It was computed choosing a polynomial order of 4, i.e. using a fourth-
265 degree equation fit of the data points and a number of smoothing points equal to 251. Processed
266 spectra thus obtained clearly presents significant peaks due to the absorption of iron oxide minerals.
267 The semi-quantitative data of iron oxide minerals are represented by the heights of these peaks, which
268 were determined by tracing the "baselines", subtracting them from the starting spectrum, and
269 quantifying the height of the resulting peak.

The conversion of the reflectance spectra into K-M spectra was carried out using the UV WinLab
2.85 Software (Perkin Elmer Ltd, United Kingdom), while the calculation of the Savitzky-Golay
second derivative was performed by the software The Unscrambler V10.4 (Camo, Oslo, Norway),
and the quantification of peak heights was carried out using the Peak Analyzer tool of the OriginPro
2018 software (Northampton, USA).

270 **2.3 Validation of the methodology**

271 *2.3.1 Validation of UV-Vis DRS results*

272 Sample color (subsection 2.2.2) was numerically defined by means of colorimetric parameters
273 described by CIE, and semi-quantitative information about iron oxide minerals (subsection 2.2.3)
274 were achieved by a suitable mathematical treatment of reflectance spectra. In order to validate the

275 semi-quantitative data of iron oxide minerals, they were compared with elemental iron concentration
276 data ($\mu\text{g cm}^{-2}$) obtained by Proton Induced X-ray Emission (PIXE) carried out on the same PM_{10}
277 filters using a Tandatron 3 MeV accelerator located at LABEC laboratory (Laboratorio di tecniche
278 nucleari per l'Ambiente e i Beni Culturali, [https://www.ionbeamcenters.eu/RADIATE-project-](https://www.ionbeamcenters.eu/RADIATE-project-partners/inf/)
279 [partners/inf/](https://www.ionbeamcenters.eu/RADIATE-project-partners/inf/)) of the INFN Section of Florence (Italy) (Lucarelli et al., 2014; Lucarelli et al., 2018).
280 Since the membranes used in this work are made of quartz instead of the typical teflon (PTFE) or
281 cellulose/nuclepore, more suitable for the PIXE technique, PIXE analysis required appropriate
282 spectral processing owing to the huge interference of silicates in the filter medium itself as described
283 in detail by Calzolari et al., 2006; Chiari et al., 2018; and Lucarelli et al., 2011. In brief, PM_{10} samples
284 were irradiated with a 3.0 MeV proton beam with a 5 nA current for 60 s, with no He flow, using a
285 scanning system allowing to analyse most of the sample area and average over possible non-
286 homogeneous deposits.

Spearman correlation analysis (Akoglu, 2018) was then performed between the UV-Vis DRS
outcomes (color parameters and semi-quantitative data of iron oxide minerals) with elemental iron
concentration calculated by PIXE and PM_{10} obtained by gravimetry, both expressed in $\mu\text{g cm}^{-2}$ for
dimensional consistency. This allowed assessing the relationship between these variables and the
effectiveness of the spectrophotometric outcomes in identifying Saharan Dust events.

287 *2.3.2 Diagnosis and validation of Saharan mineral dust events*

288 *2.3.2.1 Diagnosis based on PM filters*

289 The ultimate goal of this work is to sort out quickly but safely the PM filters exposed to Saharan Dust
290 transport events from the others.

291 Therefore, Ward's cluster analysis (Ward, 1963) using squared Euclidean distance was employed for
292 the detection of two sample clusters, respectively a cluster indicating the samples subjected to Saharan
293 dust transport events and another cluster for all the other sample cases. After standardization by

294 autoscaling (van den Berg et al., 2006), the colorimetric parameters of CIELCH model (subsection
295 2.2.2) and the semi-quantitative data of iron oxide minerals (subsection 2.2.3) were used as starting
296 variables. The statistical analysis was carried out by means of the software Statistica V.10 (StatSoft
297 Inc., Tulsa, USA).

298 The two clusters obtained were subsequently compared with PM₁₀ data normalized in air sampled
299 volume ($\mu\text{g}/\text{m}^3$) in order to associate or exclude, in a binary way, the occurrence of a Saharan dust
300 incursion.

301 2.3.2.2. Back-trajectory ensembles and residence time analysis

302 A residence time analysis of back-trajectory ensembles (Lin, 2012; Lupu & Maenhaut, 2002; Stohl,
303 1998) was carried out to assess and confirm the diagnosis made by UV-Vis DRS.

304 Ensembles of back-trajectories were calculated using the NOAA Hybrid Single Particle Lagrangian
305 Integrated Trajectory (HYSPLIT_4) model (Rolph et al., 2017; Stein et al., 2015). Meteorological
306 data from the ERA-Interim reanalysis (Dee et al., 2011) was used as input for the trajectory
307 calculations with data interpolated into a 0.5-degree grid, to make use of the 0.5-degree model terrain
308 available in the Hysplit model, and 27 pressure levels from 1000 to 100 hPa. The ensemble is
309 generated by offsetting the meteorological database by one grid point in the horizontal and 0.01 sigma
310 units in the vertical, resulting in 27 back- trajectories (Draxler, 2003). 96-hour kinematic back-
311 trajectory ensembles were calculated starting 200 m over the sampling site at 00, 06, 12, and 18 UTC
312 in the period 8 June – 11 October 2016.

313 The trajectory ensembles were grouped together according to the weekly PM sampling period. After
314 that, a residence time analysis was performed by counting the number of trajectory endpoints over
315 five broad regions: Africa, America, Europe, Mediterranean Sea, and Atlantic Ocean for the
316 trajectories within each sampling period. Furthermore, two additional specific areas such as
317 Mauritania and the Atlantic Ocean at low altitude (below 800 m) were investigated. Subsequently,

318 the ratio between the total counts and the total number of endpoints was calculated in order to obtain
319 percentages of residence time for each investigated area corresponding to each PM filter (Ashbaugh
320 et al., 1985; Orza et al., 2013; Xu et al., 2006). All the trajectory calculations were performed with R
321 (R Core Team, 2019) scripts. Further support of the occurrence of African dust events was attained
322 with the Merged Dark Target/Deep Blue aerosol optical depth (AOD) Collection C6 product from
323 the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard the Aqua and Terra satellites.

324

325 **3. RESULTS AND DISCUSSION**

326 Results and discussion section is organized as follows:

327 (a) Subsection 3.1 reports the colorimetric parameters and the related digitized colours for the
328 analyzed PM filters, highlighting and discussing their differences.

329 (b) Subsection 3.2 details the semi-quantitative results of iron oxide minerals, and their
330 processing from raw reflectance spectra.

331 (c) Subsection 3.3 shows the correlation between the semi-quantitative data of the iron oxide
332 minerals, the elemental iron obtained by Proton Induced X-ray Emission (PIXE) analysis, and
333 the colorimetric parameters.

334 (d) Subsection 3.4 reveals the PM filters that have been subjected to a Saharan Dust transport
335 event and the validation of this diagnosis by residence time analysis.

336 **3.1 The color samples**

337 For each portion of the PM filter analyzed, the color samples were obtained by the procedure
338 described in subsection 2.2.2. The colorimetric parameters related to the CIELAB (L^* , a^* , b^*) and
339 CIELCH (L^* , C_{ab}^* , h_{ab}°) models and the subsequent color obtained for the blank filter (B) and for the
340 19 PM samples (SN1, ...SN19) are reported in Table 1.

341 The collected PM samples present different colors (from gray to red) as a function of the dominating
342 aerosol source during the respective sampling time interval. In order to examine how these samples
343 differ in color, a graphical representation based on chroma (C_{ab}^*) and hue (h_{ab}°) is shown in Figure 3.
344 First, in Figure 3 (a), it can be observed how the blank filter (B) remarkably differs from all the others
345 (SN1, ...SN19). In fact, the B filter is characterized by a whitish color due to the absence of particulate
346 material, while a greyish-yellowish-reddish color characterizes the sampled filters, as can be seen in
347 the last right column of Table 1. Sampled filters in Figure 3 (b) show a defined trend: the grayest
348 samples (like SN15) present a higher hue value and a lower chroma value than other filters while the
349 most reddish samples (like SN12) show a higher value of chroma and a lower value of hue. Instead,
350 the samples with intermediate values of chroma and hue (like SN7) exhibit a browner coloration than
351 the others. This observation is in agreement with the CIELCH color definition: a lower hue value
352 corresponds to a color tone more shifted towards red, while a higher chroma value corresponds to a
353 more marked color intensity (compared to the gray color that occurs at chroma values close to zero).
354 This demonstrates how the CIELCH color space adequately describes the colors of the analyzed PM
355 filters due to their high chromaticity. In fact, the higher efficiency of the CIELCH model as compared
356 with the CIELAB one in assessing the differences between the more chromatic colors, since C_{ab}^* and
357 h_{ab}° allow a better identification of the more saturated colors than a and b (Schloss et al., 2018).

358 **3.2 Iron oxide minerals**

359 As previously reported in subsection 2.2.3, the calculation of the second derivative of the Kubelka-
360 Munk function spectra was performed over the DRS spectra. This operation allows to identify
361 decreasing peaks due to light absorption by iron oxide minerals, such as akaganéite, feroxyhyte,
362 ferrihydrite, goethite, hematite, lepidocrocite, maghemite, and schwertmannite (Sherman & Waite,
363 1985; Torrent & Barrón, 2002), whose characteristics are reported in Figure 4.

364 As reported by Scheinost et al., 1998, only the decreasing peak at around 535 nm is specific for a
365 single mineral (hematite), while the other peaks at 420 nm and 480 nm are shared by several other
366 iron oxides. Therefore, the semi-quantitative nature of the spectral data of iron oxide minerals can be
367 deduced for each portion of the PM filter through an appropriate processing of their UV-Vis
368 reflectance spectra. For the sake of brevity/conciseness, from now on graphical plots present the
369 results for only three characteristic filters, namely SN15, SN7, and SN12 (Figure 5). The selection of
370 the filters is not arbitrary but relies on careful considerations based on the colors determined for these
371 filters, as previously reported in subsection 3.1.

372 Figure 5 (a) reports the reflectance spectra obtained from the instrumental analysis. As can be seen,
373 these raw spectra present a characteristic baseline due to scattering of the UV-Vis radiation with tiny
374 signal decreases. Samples SN7 and SN12 in particular present two bands at around 480 nm and 535
375 nm characterized by a decrease in percentage reflectance due to absorption by iron oxides (Gonçalves
376 et al., 2012; Torrent & Barrón, 2002), a spectral feature not present in the sample SN15. Though the
377 attenuation in the reflectance signal due to iron oxide minerals can be observed, the latter does not
378 allow for accurate spectral quantification. The derivative spectroscopy has been widely used to: (a)
379 correct for baseline effects in spectra to remove non-chemical effects, (b) enhance small spectral
380 detail, and (c) resolve overlapped band (Ojeda & Rojas, 2013). Therefore, the application of
381 derivative spectroscopy can be particularly useful to enhance these iron peaks in UV-Vis reflectance
382 spectra of PM filters and, subsequently, obtain their relative semi-quantitative data. As from Figure
383 5 (b), the processed spectra present considerably more marked decreasing peaks due to iron oxides,
384 compared to the starting reflectance spectra reported in Figure 5 (a). Furthermore, the intensities of
385 the original curves can be seen in the derivatives in order of intensity, and this is necessary for
386 performing quantitative analyses. As such, three principally decreasing peaks can be identified:
387 around 420, 480 and 535 nm, respectively.

An example is reported in Figure 5 (c). The 420 nm peak was discarded as significantly affected by the instrumental spike due to the lamp shift from Vis to UV, a detail that can be noticed in Figure 5 (a) at around 380 nm. Average values and the standard deviations of the peak heights thus obtained (h_480nm and h_530 nm) were calculated for the blank filter (B) and for the 19 samples (SN1, ...SN19) starting from the three analyzed portions for each sample, and were reported in Table 2.

388 **3.3 Validation of UV-Vis DRS results**

389 The similarity between the sample series of iron oxide minerals semi-quantitative data (h_480nm and
390 h_535nm) and elemental iron concentration (Fe) is reported in Figure 6. Figure 6 shows a comparable
391 trend between the semi-quantitative data of iron oxide minerals and elemental iron concentration,
392 validating the results and confirming the usefulness of the diffuse reflectance measurements in the
393 analysis of iron mineral oxides. Table 3 reports the Spearman correlation coefficients between
394 CIELCH parameters, iron oxide minerals semi-quantitative data, elemental iron, and PM₁₀. As can
395 be observed, iron (Fe) is significantly linearly correlated with PM₁₀ (+0.96), confirming that mineral
396 dust is one of the main components in the analyzed samples. A high positive correlation between the
397 height of the peaks at 480 nm and 535 nm and the concentration of iron is observed (respectively,
398 +0.84 and +0.86) confirming the similar behavior shown in Figure 6 and proving the reliability of the
399 semi-quantitative data on iron mineral oxides obtained from diffuse reflectance measurements.
400 Among the two semi-quantitative data of iron oxide minerals, h_535 nm appears as the most
401 significantly related to filter colors (+0.85 with chroma and -0.83 with hue). This result is extremely
402 interesting because, as previously described in subsection 3.2, this peak is specific to hematite while
403 the other peak (h_480nm) is associated with several other iron oxides, and especially goethite. An
404 increase in chroma and a decrease in hue leads to a higher degree in the redness of the PM filters (see
405 subsection 3.1), which is exactly the characteristic color of hematite (Rossman, 1996). As such, the
406 semi-quantitative data on iron oxide minerals (especially hematite) are strongly related to PM filter
407 colors (especially the red color), and both parameters are indicative of Saharan dust transport events.

408 **3.4 Diagnosis and validation of Saharan mineral dust events**

409 Figure 7 reports the dendrogram obtained from Ward's clustering analysis applied to the UV-Vis DRS
410 results (CIELCH parameters and iron oxide minerals semi-quantitative data), and where two main
411 clusters are identified. In particular, 15 PM samples belong to cluster 1, while the other 4 PM samples
412 belong to cluster 2. These clusters are compared with PM₁₀ data (normalized in air sampled volume,
413 $\mu\text{g}/\text{m}^3$) in Figure 8 (a). Indeed, the association of mineral dust transport events has already been
414 clearly related to significant increases in PM due to the considerable mass load of this component in
415 particulate material owing to the large fraction of the coarse particles (Krasnov et al., 2014; Matassoni
416 et al., 2011). In order to facilitate the Saharan Dust detection, the UV-Vis DRS results reported in the
417 previous subsections are reported again in Figure 8(b) and Figure 8(c). The residence time spent by
418 the air parcels over different areas before reaching the sampling site is also reported in Figure 9 for
419 each PM sample.

420 Cluster 1 (highlighted in red) clearly identifies the Saharan dust events while cluster 2 (highlighted
421 in green) identifies the non-Saharan dust events (Figure 8(a)). Indeed, the PM filters classified as
422 cluster 1 are largely characterized by a higher value of PM₁₀, confirming a significant increase in PM
423 mass load as a result of mineral dust contribution. This evidence is also supported by the color of the
424 filters and semi-quantitative data of iron oxide minerals, (see Figures 8 (b,c)), whereas samples
425 belonging to cluster 2 are more greyish owing to the lower concentration of iron mineral oxides, while
426 samples belonging to cluster 1 are more brownish/reddish and with higher concentrations of iron
427 mineral oxides. As a final not negligible data, these results agree with the outcome of the residence
428 time analysis depicted in Figure 9 (a). The residence time analysis highlights how SN2, SN15, and
429 SN19 samples belonging to cluster 2 are characterized by shorter residence times (less than 10%)
430 over Africa than other PM filters (higher than 10%), suggesting uplift and rapid transport of mineral
431 dust from the North-African desert without appreciable mixing with other aerosol sources.

432 Sample SN1 presents an exception being characterized by an elevated PM₁₀ value and a residence
433 time over Africa comparable to the samples belonging to the Saharan dust events cluster (cluster 1).
434 Its assignment to the category of non-Saharan dust events, carried out only by UV-Vis DRS, is linked
435 to its greyish color and to the low concentrations of iron oxide minerals. In order to justify this
436 outcome, an in-depth analysis was carried out by satellite images retrieved from NASA's Earth
437 Observing System Data and Information System (EOSDIS) (Behnke et al., 2019), shown in Figure
438 10. Whitish dust from the dried surface of Chott el-Jerid ephemeral lake (Figure 10(a)), was uplifted
439 by the wind on 05/06/2016 (Figure 10(b)) and reached the receptor site on 08/06/2016 (from Figure
440 10(c) to Figure 10(f)), that is the first day of SN1 filter sampling period. Therefore, although the SN1
441 sample is significantly impacted by this Saharan dust transport event, its chemical-mineralogical
442 composition is unusual compared to other PM samples due to the deficiency of iron oxide minerals,
443 as evidenced by the grayish color of the examined filter.

444 After the classification of the Saharan dust events in the analyzed filters, some particular
445 considerations can be drawn for some of the other samples, i.e. SN12 and SN19. SN12 is one of the
446 PM filters more impacted by a Saharan dust events, as from its high PM₁₀ concentration and its
447 pronounced reddish color, as previously described in subsection 3.1. The latter observation may be
448 explained by the higher value of residence time over Mauritania (> 4%), as reported in Figure 9 (b),
449 whose area is known to be an important source of reddish hematite (Journet et al., 2014; Schlueter,
450 2006; Waele et al., 2019). As previously assessed, sample SN19 is clearly a PM sample not affected
451 by Saharan dust transport. This filter presents the “whitest” color amongst the samples analysed, as
452 shown by the highest luminescence value ($L^* = 73.23$, look at Table 1) in the CIELAB/CIELCH
453 colorimetric models. This result can be justified by a significantly long residence time over the
454 Atlantic Ocean at low altitude (> 5%), as reported in Figure 9 (c), which presumably involves a strong
455 influence of colorless sea salt component (mainly defined by sodium chloride and magnesium
456 chloride) in the examined filter.

4. CONCLUSIONS

457

458 In this work, the feasibility of UV-Vis Diffuse Reflectance Spectroscopy for a rapid and non-
459 destructive diagnosis of Saharan dust events in particulate matter filters has been described, assessed,
460 and validated. This method has been applied to particulate matter filters sampled at high altitude
461 (2550 m a.s.l.) in an area heavily impacted by Saharan mineral dust incursion events due to its
462 proximity to North Africa (Sierra Nevada, Spain, 37.096 N, -3.387 W).

463 In particular, this analytical method allowed to identify unequivocally two absorption bands
464 corresponding to a well-defined set of iron mineral oxides contained in Saharan dust: the absorption
465 band at about 480 nm, representative of multiple iron oxide minerals (i.e., goethite, lepidocrocite,
466 maghemite, ferrihydrite, ferrosityte, akaganéite, and schwertmannite), and another at about 535 nm,
467 specific for hematite. Through appropriate processing of the obtained reflectance spectra, it has been
468 possible to obtain semi-quantitative data for these mineral oxides. Furthermore, starting from the
469 reflectance measurements, it has been possible to quantitatively parameterize the filter coloring as a
470 function of PM source.

471 The results obtained from this technique have been validated on the basis of the elemental iron
472 concentration obtained by Proton Induced X-ray Emission (PIXE) analysis. Besides, it has been
473 demonstrated the relation between the concentration of hematite increases and the higher reddish
474 color of the filters with an increase in their PM₁₀ content. Therefore, the UV-Vis DRS has been proven
475 to be extremely useful for a fast, cheap, and unambiguous identification of Saharan mineral dust
476 events in PM filters.

477 The results obtained have been finally proven on the basis of residence time analysis of back-
478 trajectory ensembles, whose outcomes are in excellent agreement with those obtained by UV-Vis
479 DRS, except for one PM sample with a peculiar chemical-mineralogical composition likely associated
480 with the dried Chott el-Jerid Lake (Tunisia). Furthermore, some other samples have been explored

481 by associating the color and the semi-quantitative data of iron oxide minerals with their particular PM
482 sources.

483 In conclusion, the UV-Vis DRS technique can be reliably adopted for Saharan dust events under
484 certain conditions:

485 - The prevailing emission source of PM filters must be mineral dust. Indeed, a complex mixture
486 of sources can alter the color and mineralogical composition of the sampled filters, making
487 the identification of Saharan dust events extremely complicated; however, it is important to
488 highlight that in this work weekly PM₁₀ samples have been used, certainly affected by a
489 complex mixture of emission sources and, despite this, reasonable outcomes have been
490 achieved.

491 - Iron oxide minerals prove as efficient but also practically utilizable markers of Saharan
492 mineral dust. This is valid in most cases even though, in this work, it has been highlighted
493 that this spectroscopic technique is not capable to identify whitish sand events, characterized
494 by low concentrations of iron oxide minerals.

495

496 **ACKNOWLEDGEMENTS**

497 This work has been carried out under the Spanish project FRESA (Impact of dust-laden African air
498 masses and of stratospheric air masses in the Iberian Peninsula. Role of the Atlas Mountains, Ref:
499 CGL2015-70741-R). Firstly, Lucas Alados-Arboledas (IISTA-CEAMA) is gratefully acknowledged
500 for making possible the field campaign at the sampling site. A Heartfelt thanks to Sara Arcozzi, for
501 her technical assistance on the Perkin Elmer Lambda 35 UV-Vis Spectrophotometer, and Christian
502 Cingolani, Josep Mestres Sanna, and Adrià Simon Ortiz for the assistance in the experimental activity
503 during their thesis work. Finally, the authors acknowledge the NOAA Air Resources Laboratory
504 (ARL) for the provision of the HYSPLIT transport and dispersion model and/or READY website

505 (<https://www.ready.noaa.gov>) used in this publication and the European Centre for Medium-range
506 Weather Forecast (ECMWF) for making available the ERA-Interim database.

507

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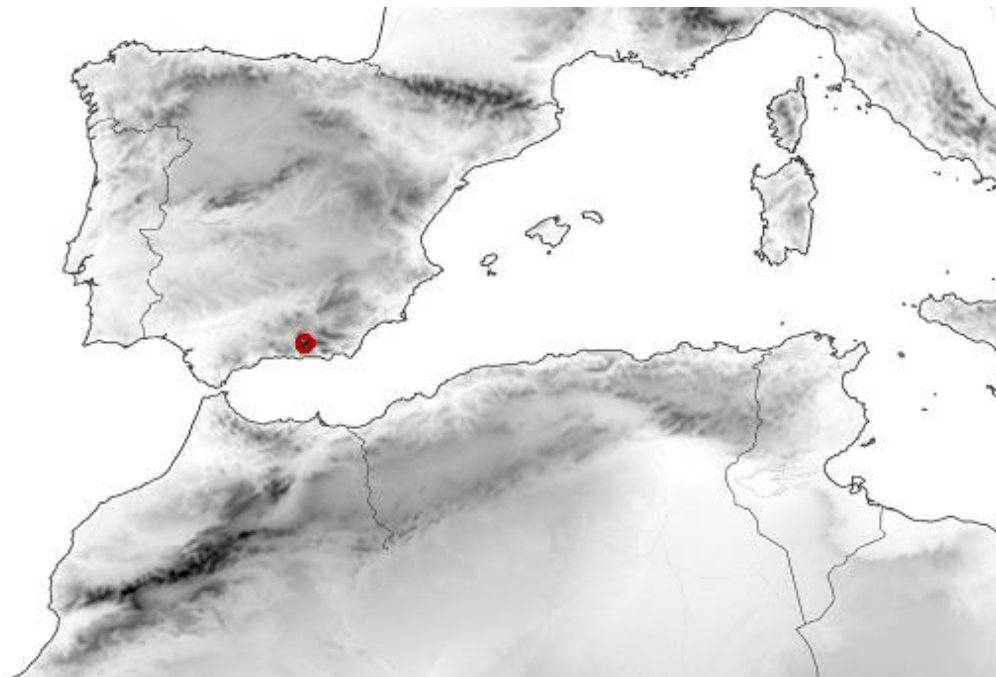
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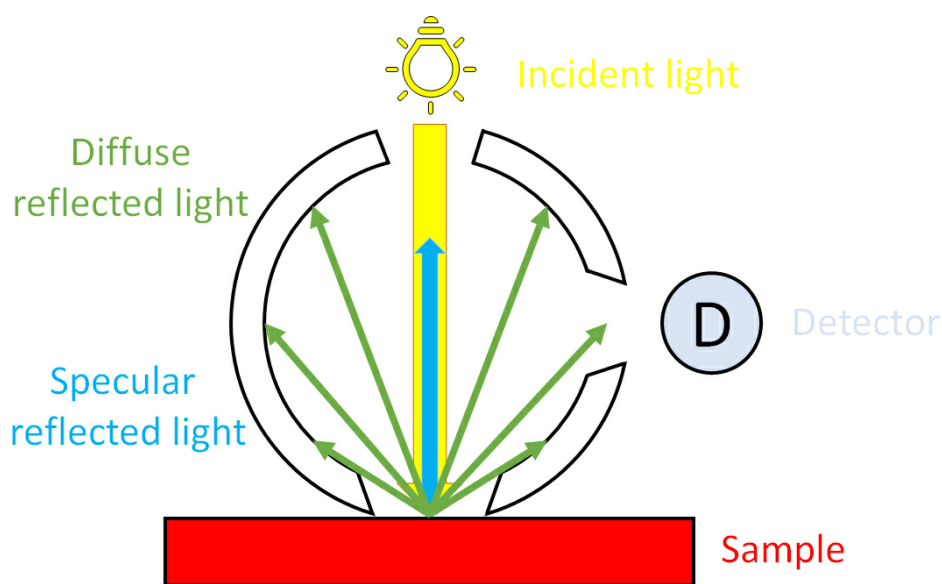
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818 **FIGURES:**



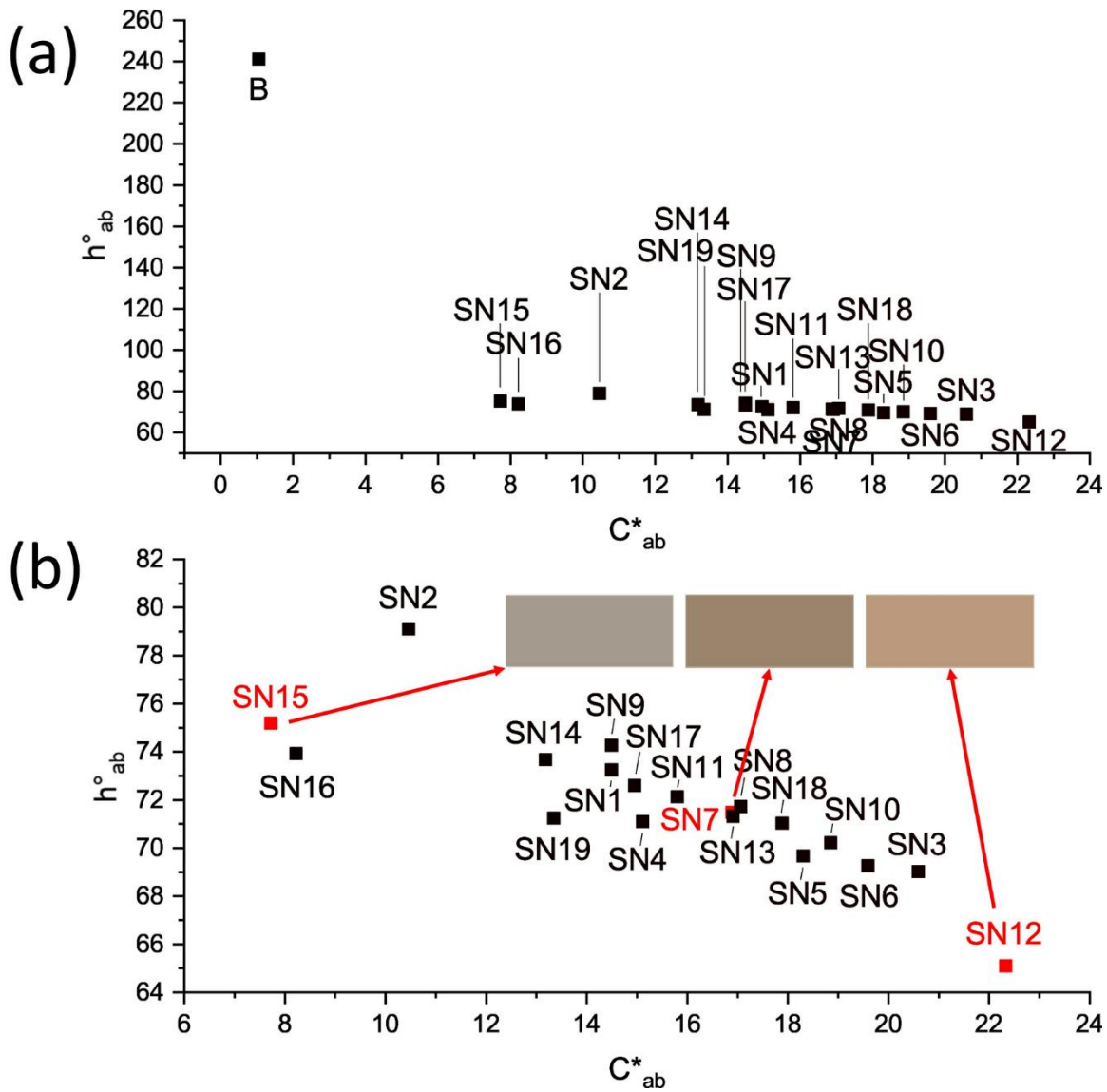
819

820 Figure 1. Map and location of Sierra Nevada sampling station.



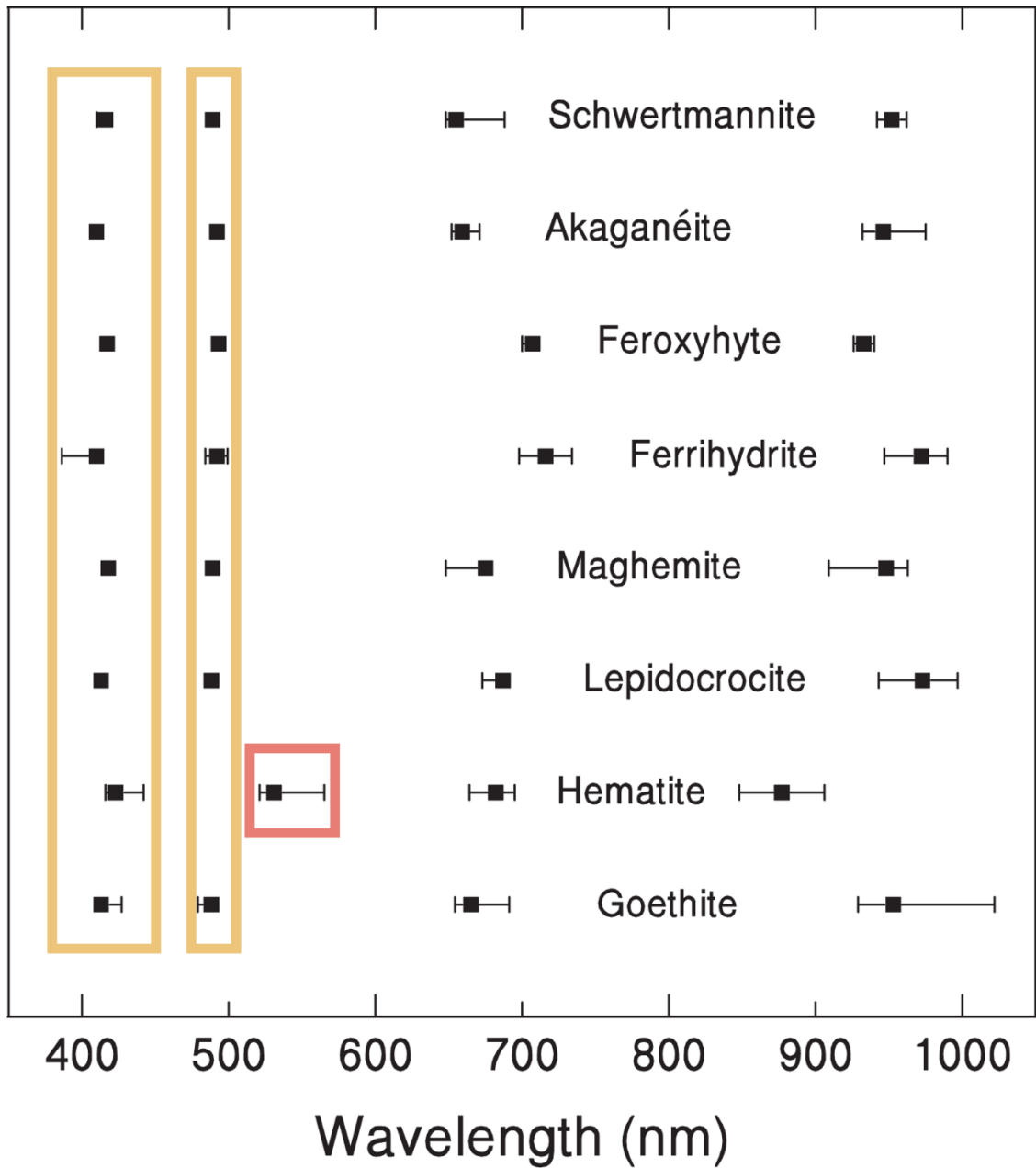
821

822 Figure 2. Scheme of the integrating sphere, 0°/d geometry. The UV-Vis incident light hits the sample
823 perpendicularly. Any specular component of reflection from the sample is excluded from
824 measurement since this component is directed out of the sphere through the transmittance sample
825 entrance port and, therefore, the detector only measures the diffuse reflectance component.



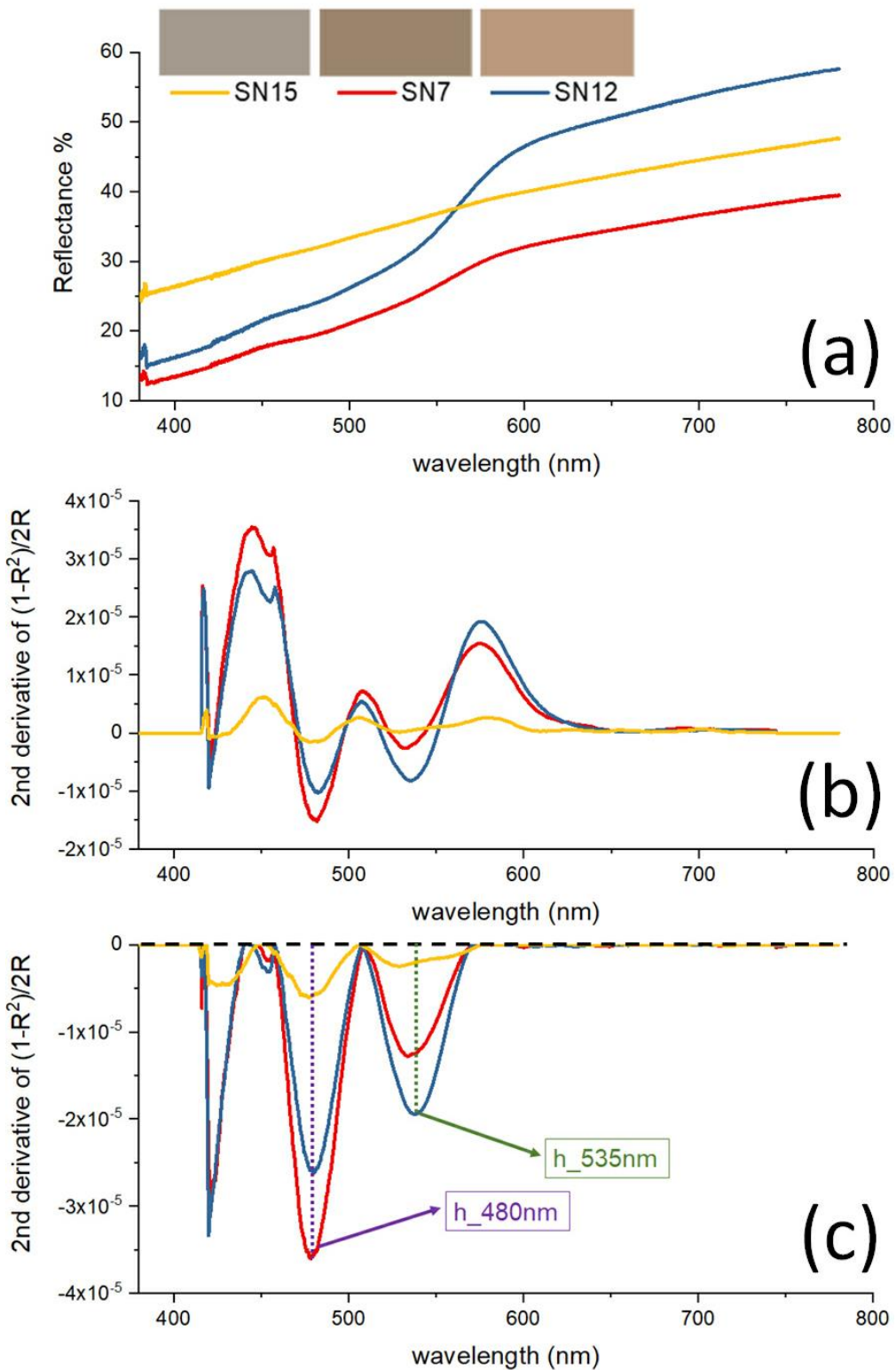
826

827 Figure 3. (a) Graphic representation of blank filter (B) and sampled filters (SN1, ...SN19) based on
 828 chroma (C_{ab}^*) and hue (h_{ab}°). (b) Zoomed plot on sampled filters. The colors of three specific samples
 829 (SN15, SN7, SN12) are shown.



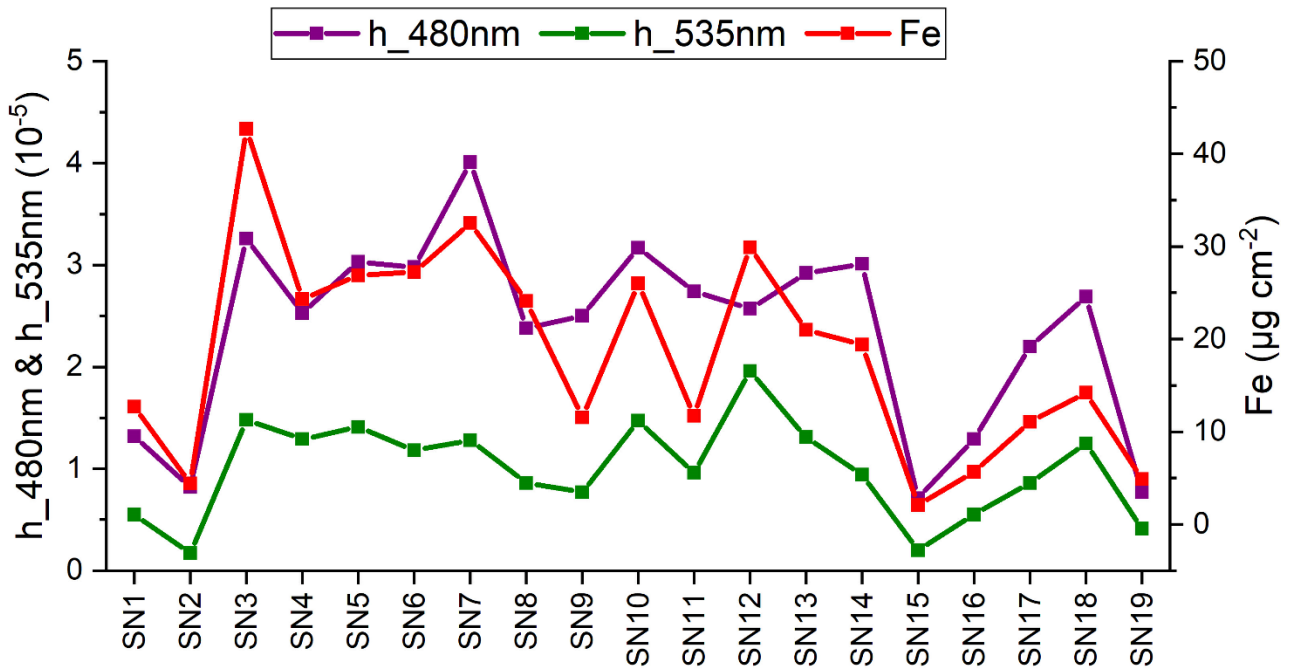
830

831 Figure 4. Median and range of the UV-Vis absorption bands of some iron oxide minerals in the second
 832 derivative of K-M function spectra, adapted from Torrent & Barrón, 2008. It can be seen that below
 833 600 nm, there are three absorption bands at around 420, 480, and 535 nm.



834

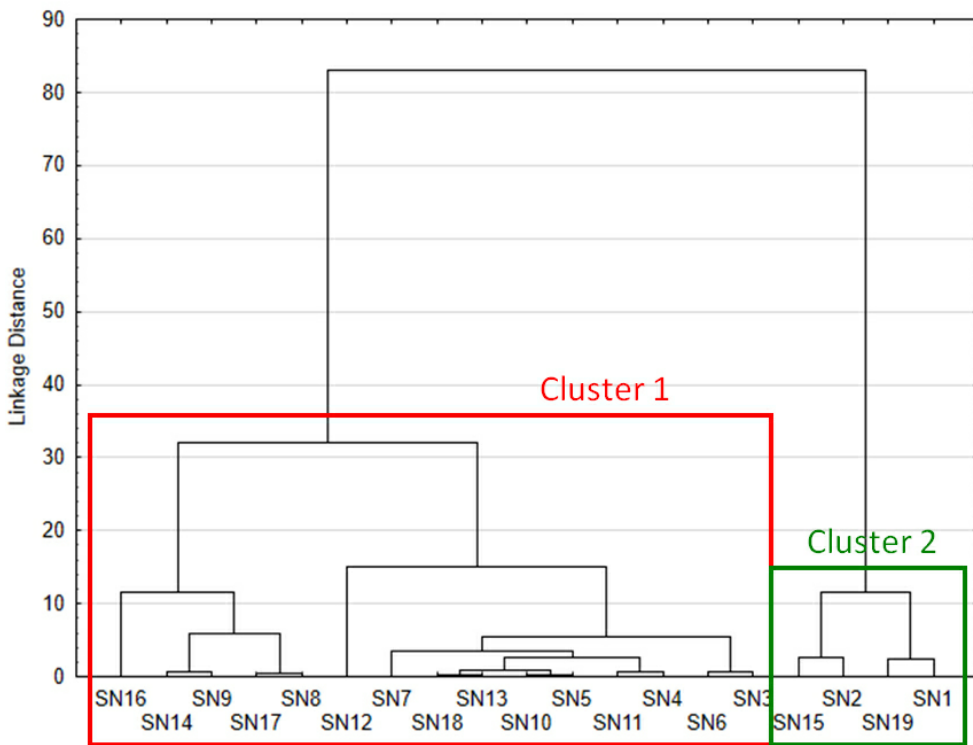
835 Figure 5. Reflectance spectra (a), Savitzky-Golay second derivative of Kubelka-Munk (K-M)
 836 function spectra (b), and baseline subtraction and quantification of relevant peak heights (c) for SN15
 837 (grayish filter), SN7 (brownish filter), and SN12 (reddish filter).



838

839 Figure 6. Sample series of h_480nm (semi-quantitative data of several iron oxide minerals, mainly

840 goethite), h_535nm (semi-quantitative data of hematite), and elemental iron concentration (Fe).

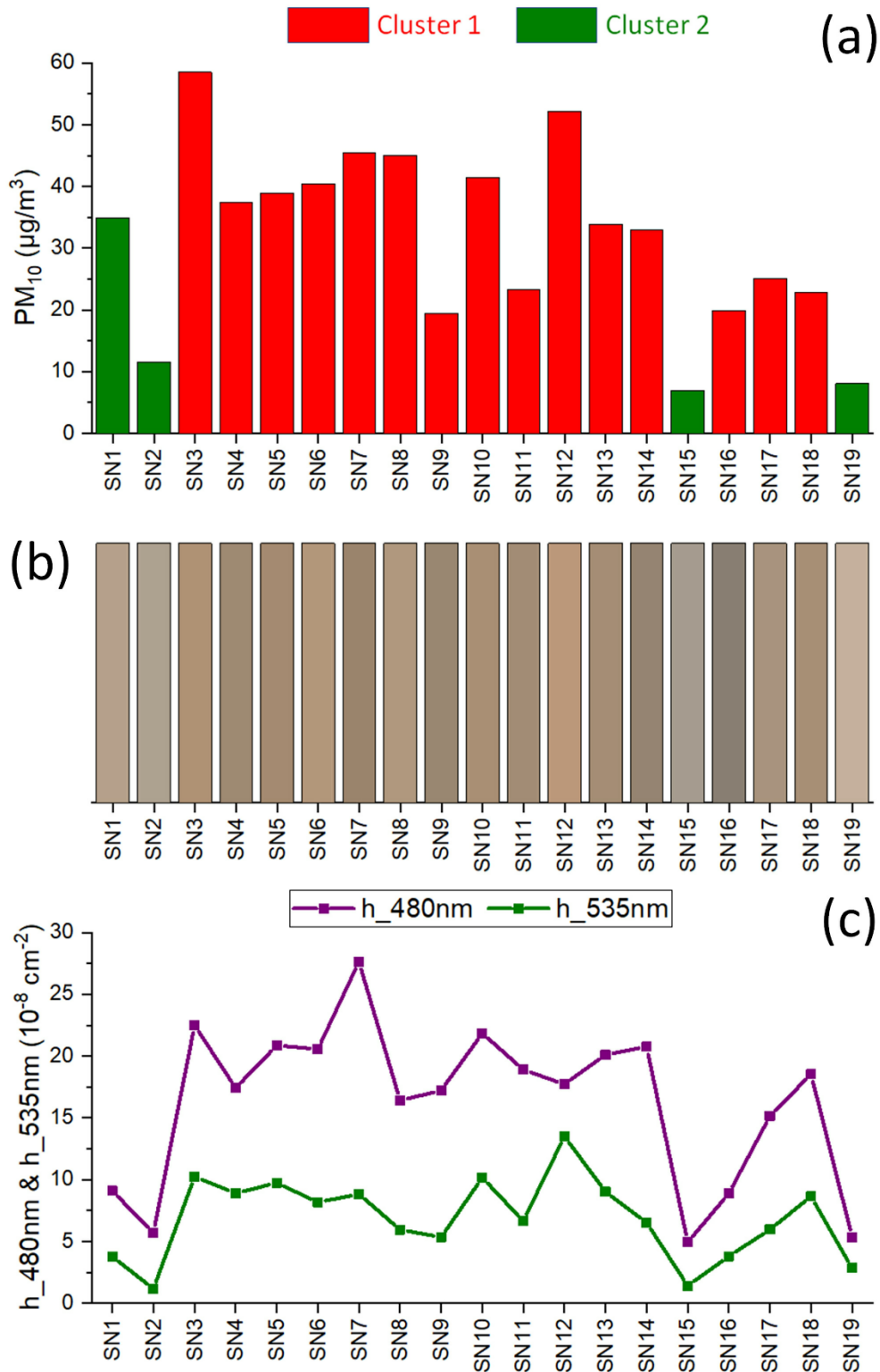


841

842 Figure 7. Dendrogram of Ward's hierarchical clustering method starting from CIELCH parameters

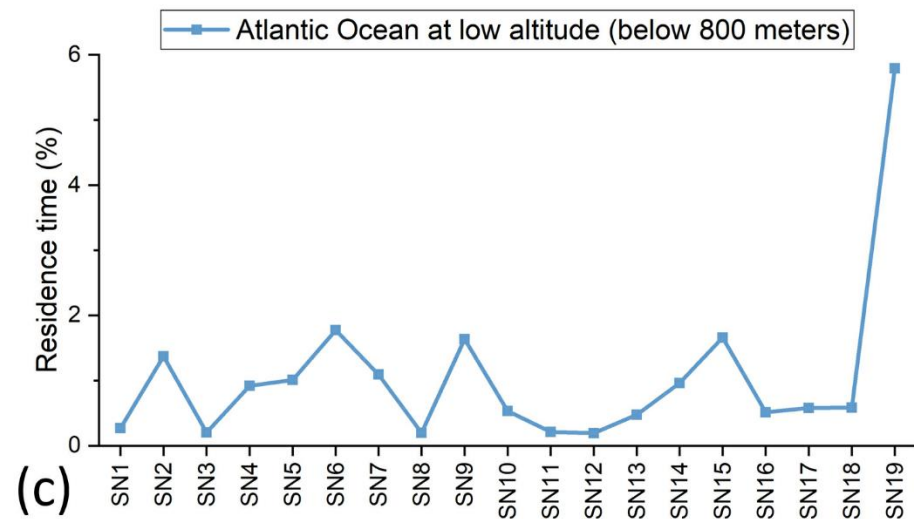
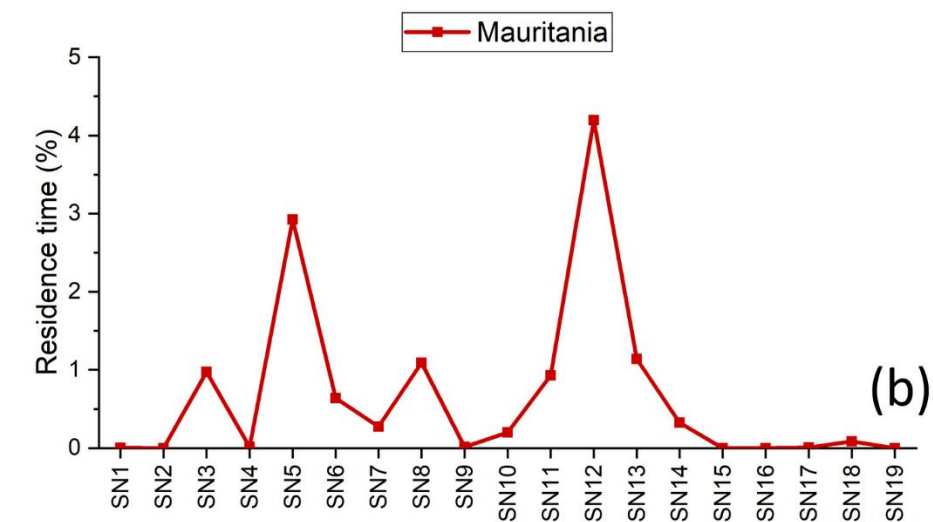
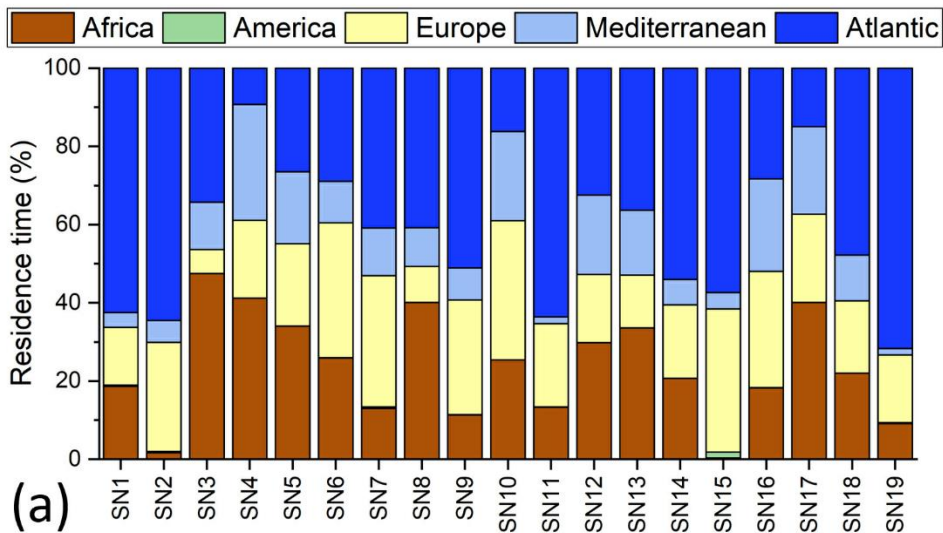
843 and semi-quantitative data of iron oxide minerals. The clustering solution with two clusters is

844 highlighted by squares.



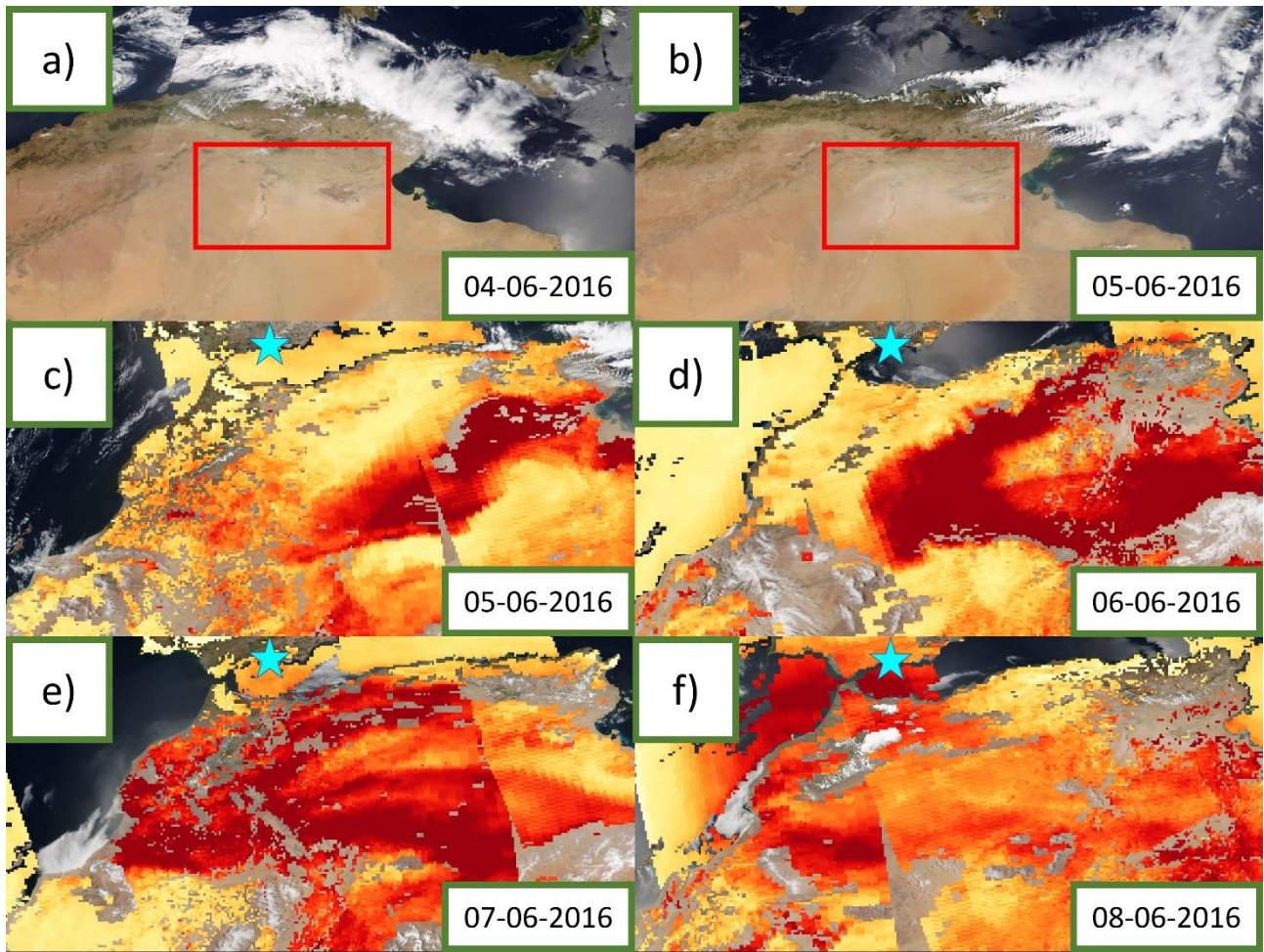
845

846 Figure 8. (a) Sample series of PM₁₀ data (μg/m³) with the association of PM filters to the respective
 847 clusters (cluster 1 and cluster 2). (b) Display of PM filters colors, look at subsection 3.1. (c) Sample
 848 series of h_{480nm} (semi-quantitative data of several iron oxide minerals) and h_{535nm} (hematite),
 849 look at subsection 3.2.



850

851 Figure 9. (a) Stacked bar chart of the percentage residence time over each examined region and PM
 852 filter; Sample series of residence time over Mauritania (b) and the Atlantic Ocean, below 800 meters
 853 (c).



854

855 Figure 10. Corrected reflectance (True Color) Terra/MODIS snapshots for 4 June 2016 (a) and 5 June
 856 2016 (b); Merged DT/DB Aerosol Optical Depth (Land and Ocean) Aqua/MODIS snapshots from 5
 857 June 2016 (c) to 8 June 2016 (f). Higher AOD values are indicated by a more reddish color and allow
 858 following the dust plume transport. The red square in a) and b) highlights the Chott el-Jerid Lake
 859 (Tunisia) while the light blue star in c), d), e), and f) indicates the PM sampling station in Sierra
 860 Nevada (37.096 N, -3.387 W, 2550 m a.s.l.). Images have been retrieved from the NASA EOSDIS,
 861 worldview tool at <https://worldview.earthdata.nasa.gov>.

862





















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866 **TABLES:**

867 Table 1. Average values and standard deviations (numbers in brackets) of the colorimetric
 868 parameters for the analyzed samples. The colors obtained from the conversion to RGB color model
 869 by the online tool nix Color Sensor (<https://www.nixsensor.com/free-color-converter/>) are displayed
 870 in the last column of this table.

Sample	L*	a*	b*	C _{ab} *	h _{ab} °	Color
B	95.46 (0.11)	-0.51 (0.05)	-0.92 (0.13)	1.05 (0.14)	241.15 (0.81)	
SN1	67.49 (2.68)	4.14 (1.19)	13.89 (4.42)	14.49 (4.57)	73.26 (0.64)	
SN2	66.85 (3.60)	1.99 (0.43)	10.27 (1.08)	10.46 (1.13)	79.11 (1.53)	
SN3	62.63 (3.07)	7.37 (0.56)	19.23 (1.36)	20.59 (1.47)	69.03 (0.11)	
SN4	58.53 (2.46)	4.86 (1.55)	14.31 (4.99)	15.11 (5.22)	71.10 (0.71)	
SN5	59.27 (2.79)	6.33 (0.09)	17.16 (1.64)	18.30 (1.56)	69.67 (1.66)	
SN6	64.43 (2.91)	6.84 (0.46)	18.35 (3.35)	19.59 (3.28)	69.27 (2.52)	
SN7	56.77 (2.12)	5.36 (0.39)	16.02 (1.22)	16.89 (1.28)	71.48 (0.14)	
SN8	64.35 (1.41)	5.37 (1.63)	16.20 (4.63)	17.06 (4.90)	71.72 (0.38)	
SN9	57.51 (3.59)	3.94 (0.51)	13.94 (1.26)	14.49 (1.35)	74.27 (0.70)	
SN10	61.20 (3.26)	6.41 (1.31)	17.72 (2.97)	18.85 (3.23)	70.22 (0.79)	
SN11	59.83 (3.63)	4.86 (0.81)	15.04 (2.11)	15.80 (2.25)	72.13 (0.89)	
SN12	65.59 (0.76)	9.41 (1.52)	20.24 (2.92)	22.33 (3.28)	65.11 (0.65)	
SN13	60.28 (3.57)	5.43 (0.80)	16.02 (1.76)	16.91 (1.93)	71.31 (0.78)	
SN14	56.44 (1.09)	3.72 (0.95)	12.65 (2.98)	13.18 (3.13)	73.68 (0.49)	
SN15	64.91 (2.89)	1.94 (0.32)	7.47 (1.86)	7.72 (1.88)	75.19 (1.56)	
SN16	53.75 (2.55)	2.25 (0.27)	7.90 (1.52)	8.22 (1.53)	73.93 (1.58)	
SN17	62.68 (2.65)	4.46 (0.53)	14.27 (2.03)	14.95 (2.09)	72.60 (0.90)	
SN18	61.01 (2.99)	5.80 (0.09)	16.91 (1.04)	17.88 (1.00)	71.04 (0.98)	
SN19	73.23 (2.41)	4.20 (0.56)	12.63 (2.16)	13.34 (1.97)	71.24 (4.53)	

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878 Table 2. Average values and standard deviations (numbers in brackets) of the semi-quantitative data
 879 of mixed iron oxide minerals (h_480nm) and hematite (h_535 nm).

Sample	h_480nm (10 ⁻⁵)	h_530nm (10 ⁻⁵)
B	0.025 (0.009)	0.011 (0.005)
SN1	1.32 (0.37)	0.55 (0.08)
SN2	0.83 (0.16)	0.17 (0.04)
SN3	3.27 (0.71)	1.49 (0.20)
SN4	2.53 (1.20)	1.29 (0.17)
SN5	3.03 (0.03)	1.42 (0.06)
SN6	2.99 (0.27)	1.19 (0.19)
SN7	4.01 (0.38)	1.28 (0.11)
SN8	2.38 (0.90)	0.87 (0.08)
SN9	2.50 (0.05)	0.78 (0.16)
SN10	3.17 (0.11)	1.48 (0.09)
SN11	2.74 (0.10)	0.97 (0.18)
SN12	2.57 (0.10)	1.96 (0.22)
SN13	2.92 (0.62)	1.31 (0.33)
SN14	3.02 (0.64)	0.95 (0.15)
SN15	0.72 (0.46)	0.21 (0.13)
SN16	1.29 (0.15)	0.55 (0.26)
SN17	2.20 (0.06)	0.87 (0.19)
SN18	2.69 (0.52)	1.26 (0.31)
SN19	0.78 (0.18)	0.41 (0.09)

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889 Table 3. Spearman correlation coefficients obtained for each pair of variables. L* = CIELAB
 890 lightness, C_{ab}* = CIELAB chroma, h_{ab}^o = CIELAB hue, Fe = elemental iron, PM₁₀ = particulate
 891 matter, h_480nm = semi-quantitative data of mixed iron oxide minerals, h_535nm= semi-
 892 quantitative data of hematite.

	L*	C _{ab} *	h _{ab} ^o	Fe	PM ₁₀	h_480nm	h_535nm
L*	1.00						
C _{ab} *	0.08	1.00					
h _{ab} ^o	-0.11	-0.90	1.00				
Fe	-0.20	0.85	-0.80	1.00			
PM ₁₀	-0.11	0.81	-0.74	0.96	1.00		
h_480nm	-0.47	0.69	-0.60	0.84	0.75	1.00	
h_535nm	-0.28	0.85	-0.83	0.86	0.81	0.81	1.00

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911 **CRedit author statement**

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913 **Pietro Morozzi:** Conceptualization, Methodology, Formal analysis, Investigation, Writing - Original
914 Draft, Visualization; **Barbara Ballarin:** Resources, Writing - Review & Editing; **Erika Brattich:**
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919 - Review & Editing, Supervision

920

921 **Declaration of interests**

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923 The authors declare that they have no known competing financial interests or personal
924 relationships that could have appeared to influence the work reported in this paper.

925

926 The authors declare the following financial interests/personal relationships which may be considered as
927 potential competing interests:

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