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# TITLE

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

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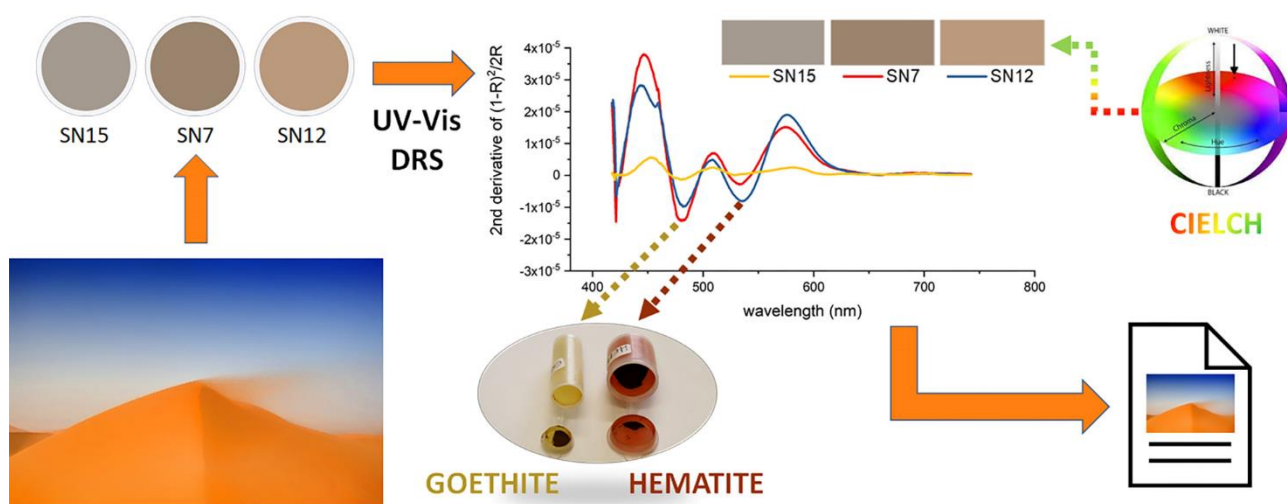
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## ABSTRACT

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

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

In this work, it is demonstrated how, as the concentration of iron mineral oxides and especially of hematite increases, the extent of redness color in PM filters grows up. Therefore, this technique can be extremely useful for a rapid, cheap and unambiguous identification of Saharan dust events in PM filters. The diagnosis of Saharan dust events was performed on PM<sub>10</sub> filters with a strong mineral dust component and demonstrated with the residence time analysis of back-trajectory ensembles, proving the reliability of this non-destructive methodology.



## KEYWORDS

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

## HIGHLIGHTS

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

## 1. INTRODUCTION

Mineral dust uplifted into the atmosphere by the wind in arid and semi-arid regions of North Africa are often transported across thousands of kilometers including both the northernmost European territory and/or the American continent (e.g., Middleton & Goudie, 2002). Overall the Mediterranean basin owing to proximity and average circulation patterns, is directly and widely affected by these phenomena throughout the year with events whose intensity and frequency are object of extensive research (Brattich et al., 2015a; Brattich et al., 2015b; Cabello et al., 2016; Cuevas et al., 2017; Cusack et al., 2012; Israelevich et al., 2012; Riccio et al., 2009; Tositti et al., 2014). The frequency and intensity of Saharan Dust outbreaks are presently reported as increasing due to the effect of global warming (Middleton & Goudie, 2002; Soleimani et al., 2020). These events can contribute to an increase of PM<sub>10</sub> levels above the limits allowed by the air quality regulations drawing attention on health threatens as well as on their correct management (Diapouli et al., 2017; Krasnov et al., 2014; Matassoni et al., 2011; Nava et al., 2012; Querol et al., 2019). In particular, the European Air Quality Directive 2008/50/EC establishes that PM<sub>10</sub> (i.e. PM with an aerodynamic diameter less than 10 µm) daily mean value may not exceed 50 µg/m<sup>3</sup> more than 35 times in a year and that the PM<sub>10</sub> annual mean value may not exceed 40 µg/m<sup>3</sup> (EU, 2008). similarly, the World Health Organization (WHO) recommends a PM<sub>10</sub> daily mean value less than 50 µg/m<sup>3</sup> but with a considerably lower PM<sub>10</sub> annual mean value of 20 µg/m<sup>3</sup> (WHO, 2006). The European legislation allows subtracting the contribution 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 PM<sub>10</sub> 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 PM<sub>10</sub> 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 (Ca<sup>2+</sup>) (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 PM<sub>10</sub> 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);



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

## **2.1 Particulate matter samples**

PM<sub>10</sub> samples analyzed in this work were collected at Sierra Nevada a high altitude site (37.096 N, -3.387 W, 2550 m a.s.l.) in Southern Spain, within the framework of the Spanish national project FRESA (Impact of dust-laden African air masses and of stratospheric air masses in the Iberian Peninsula. Role of the Atlas Mountains, Ref: CGL2015-70741-R). The sampling station is located in an area scarcely influenced by traffic and other anthropogenic sources, but strongly impacted by Saharan dust incursion events due to its proximity to North Africa (Figure 1).

PM sampling was carried out using a high volume PM<sub>10</sub> sampler (CAV-A/mb, MCV S.A.) on a weekly basis (168 h at 30 m<sup>3</sup>/h) using quartz filters (Ø 15 cm, Whatman QM-A quartz filters) from 8 June to 11 October 2016. Nineteen weekly PM<sub>10</sub> samples (labeled as SN1-SN19) were overall obtained and processed along with three field blanks (labeled as B1-B3).

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

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

UV-Vis Diffuse Reflectance Spectroscopy is a widely used, basic spectrophotometric technique for the analysis of powders and surfaces, requiring a negligible sample preparation (Torrent & Barrón, 2008). It is based on the surface dispersion of a fraction of the UV-Vis incident radiation on it. A UV-Vis collimated light beam is directed with a certain angle onto the sample and, as a result, an ensemble 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  $\lambda$  of the UV-Vis incident radiation (i.e.,  
215 reflectance spectra were obtained), based on the following instrumental parameters:  $\lambda$  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

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

### 2.2.3 Semi-quantification of iron oxide minerals

Diffuse reflectance measurements are extremely useful for the individuation and quantification of the most important sample pigments. Indeed, they exhibit attenuation in reflectance spectra due to their light-absorption in specific UV-Vis wavelength ranges. Iron oxide minerals absorption is associated with their electronic transitions within the  $3d^5$  shell of  $Fe^{3+}$  ion triggered by UV-Vis radiation (Scheinost et al., 1998) and, consequently, reflectance spectra can be useful for the assessment of 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  $\text{PM}_{10}$   
277 filters using a Tandetron 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/inf/)  
279 [partners/inf/](https://www.ionbeamcenters.eu/RADIATE-project-partners/inf/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,  $\text{PM}_{10}$  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  $\text{PM}_{10}$  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 PM<sub>10</sub> 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,

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

324

### 3. RESULTS AND DISCUSSION

Results and discussion section is organized as follows:

- (a) Subsection 3.1 reports the colorimetric parameters and the related digitized colours for the analyzed PM filters, highlighting and discussing their differences.
- (b) Subsection 3.2 details the semi-quantitative results of iron oxide minerals, and their processing from raw reflectance spectra.
- (c) Subsection 3.3 shows the correlation between the semi-quantitative data of the iron oxide minerals, the elemental iron obtained by Proton Induced X-ray Emission (PIXE) analysis, and the colorimetric parameters.
- (d) Subsection 3.4 reveals the PM filters that have been subjected to a Saharan Dust transport event and the validation of this diagnosis by residence time analysis.

#### 3.1 The color samples

For each portion of the PM filter analyzed, the color samples were obtained by the procedure described in subsection 2.2.2. The colorimetric parameters related to the CIELAB ( $L^*$ ,  $a^*$ ,  $b^*$ ) and CIELCH ( $L^*$ ,  $C_{ab}^*$ ,  $h_{ab}^\circ$ ) models and the subsequent color obtained for the blank filter (B) and for the 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 ( $h_{ab}^\circ$ ) 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  $h_{ab}^\circ$  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, ferroxhyte,  
 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 ( $h_{480\text{nm}}$  and  $h_{530\text{ nm}}$ ) 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.

### 3.3 Validation of UV-Vis DRS results

The similarity between the sample series of iron oxide minerals semi-quantitative data ( $h_{480\text{nm}}$  and  $h_{535\text{nm}}$ ) and elemental iron concentration (Fe) is reported in Figure 6. Figure 6 shows a comparable trend between the semi-quantitative data of iron oxide minerals and elemental iron concentration, validating the results and confirming the usefulness of the diffuse reflectance measurements in the analysis of iron mineral oxides. Table 3 reports the Spearman correlation coefficients between CIELCH parameters, iron oxide minerals semi-quantitative data, elemental iron, and  $\text{PM}_{10}$ . As can be observed, iron (Fe) is significantly linearly correlated with  $\text{PM}_{10}$  (+0.96), confirming that mineral dust is one of the main components in the analyzed samples. A high positive correlation between the height of the peaks at 480 nm and 535 nm and the concentration of iron is observed (respectively, +0.84 and +0.86) confirming the similar behavior shown in Figure 6 and proving the reliability of the semi-quantitative data on iron mineral oxides obtained from diffuse reflectance measurements. Among the two semi-quantitative data of iron oxide minerals,  $h_{535\text{ nm}}$  appears as the most significantly related to filter colors (+0.85 with chroma and -0.83 with hue). This result is extremely interesting because, as previously described in subsection 3.2, this peak is specific to hematite while the other peak ( $h_{480\text{nm}}$ ) is associated with several other iron oxides, and especially goethite. An increase in chroma and a decrease in hue leads to a higher degree in the redness of the PM filters (see subsection 3.1), which is exactly the characteristic color of hematite (Rossman, 1996). As such, the semi-quantitative data on iron oxide minerals (especially hematite) are strongly related to PM filter colors (especially the red color), and both parameters are indicative of Saharan dust transport events.

### 3.4 Diagnosis and validation of Saharan mineral dust events

Figure 7 reports the dendrogram obtained from Ward's clustering analysis applied to the UV-Vis DRS results (CIELCH parameters and iron oxide minerals semi-quantitative data), and where two main clusters are identified. In particular, 15 PM samples belong to cluster 1, while the other 4 PM samples belong to cluster 2. These clusters are compared with PM<sub>10</sub> data (normalized in air sampled volume,  $\mu\text{g}/\text{m}^3$ ) in Figure 8 (a). Indeed, the association of mineral dust transport events has already been clearly related to significant increases in PM due to the considerable mass load of this component in particulate material owing to the large fraction of the coarse particles (Krasnov et al., 2014; Matassoni et al., 2011). In order to facilitate the Saharan Dust detection, the UV-Vis DRS results reported in the previous subsections are reported again in Figure 8(b) and Figure 8(c). The residence time spent by the air parcels over different areas before reaching the sampling site is also reported in Figure 9 for each PM sample.

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

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

After the classification of the Saharan dust events in the analyzed filters, some particular considerations can be drawn for some of the other samples, i.e. SN12 and SN19. SN12 is one of the PM filters more impacted by a Saharan dust events, as from its high  $PM_{10}$  concentration and its pronounced reddish color, as previously described in subsection 3.1. The latter observation may be explained by the higher value of residence time over Mauritania ( $> 4\%$ ), as reported in Figure 9 (b), whose area is known to be an important source of reddish hematite (Journet et al., 2014; Schlueter, 2006; Waele et al., 2019). As previously assessed, sample SN19 is clearly a PM sample not affected by Saharan dust transport. This filter presents the “whitest” color amongst the samples analysed, as shown by the highest luminescence value ( $L^* = 73.23$ , look at Table 1) in the CIELAB/CIELCH colorimetric models. This result can be justified by a significantly long residence time over the Atlantic Ocean at low altitude ( $> 5\%$ ), as reported in Figure 9 (c), which presumably involves a strong influence of colorless sea salt component (mainly defined by sodium chloride and magnesium chloride) in the examined filter.

## 4. CONCLUSIONS

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

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

The results obtained from this technique have been validated on the basis of the elemental iron concentration obtained by Proton Induced X-ray Emission (PIXE) analysis. Besides, it has been demonstrated the relation between the concentration of hematite increases and the higher reddish color of the filters with an increase in their PM<sub>10</sub> content. Therefore, the UV-Vis DRS has been proven to be extremely useful for a fast, cheap, and unambiguous identification of Saharan mineral dust events in PM filters.

The results obtained have been finally proven on the basis of residence time analysis of back-trajectory ensembles, whose outcomes are in excellent agreement with those obtained by UV-Vis DRS, except for one PM sample with a peculiar chemical-mineralogical composition likely associated 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 PM<sub>10</sub> 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

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507

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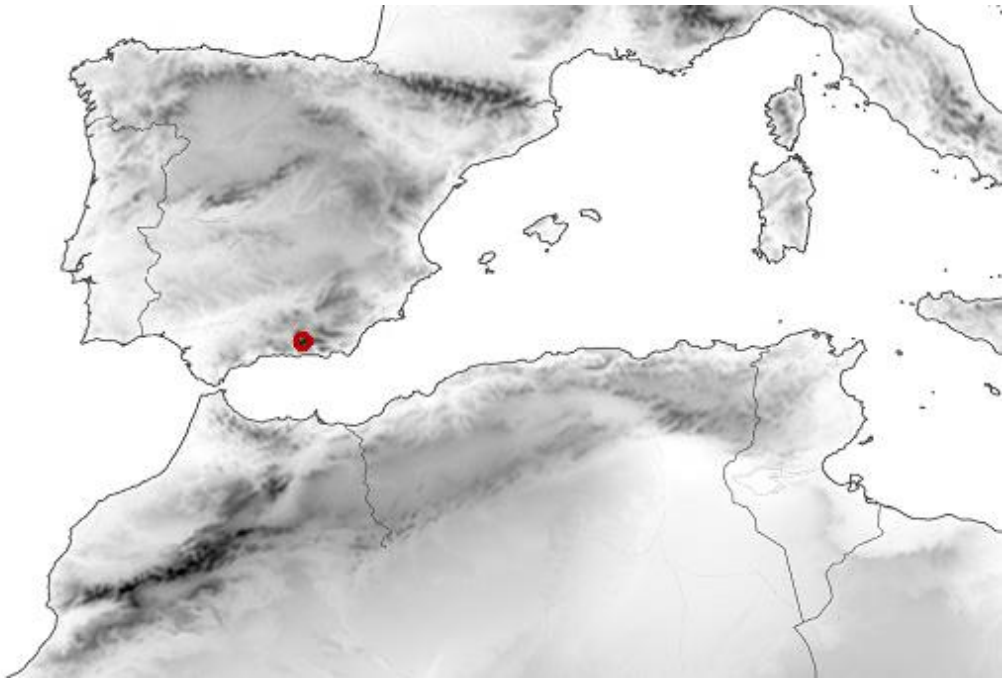
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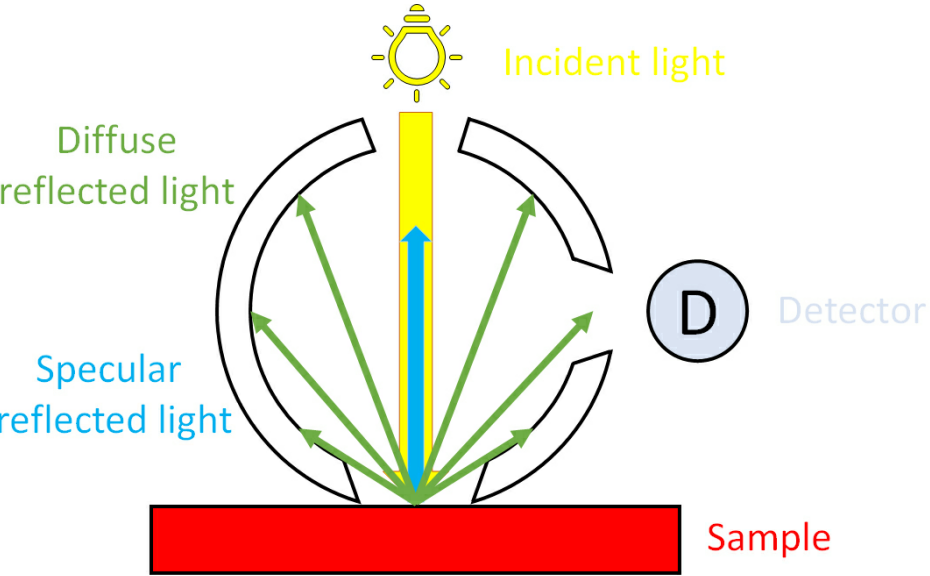
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818 **FIGURES:**



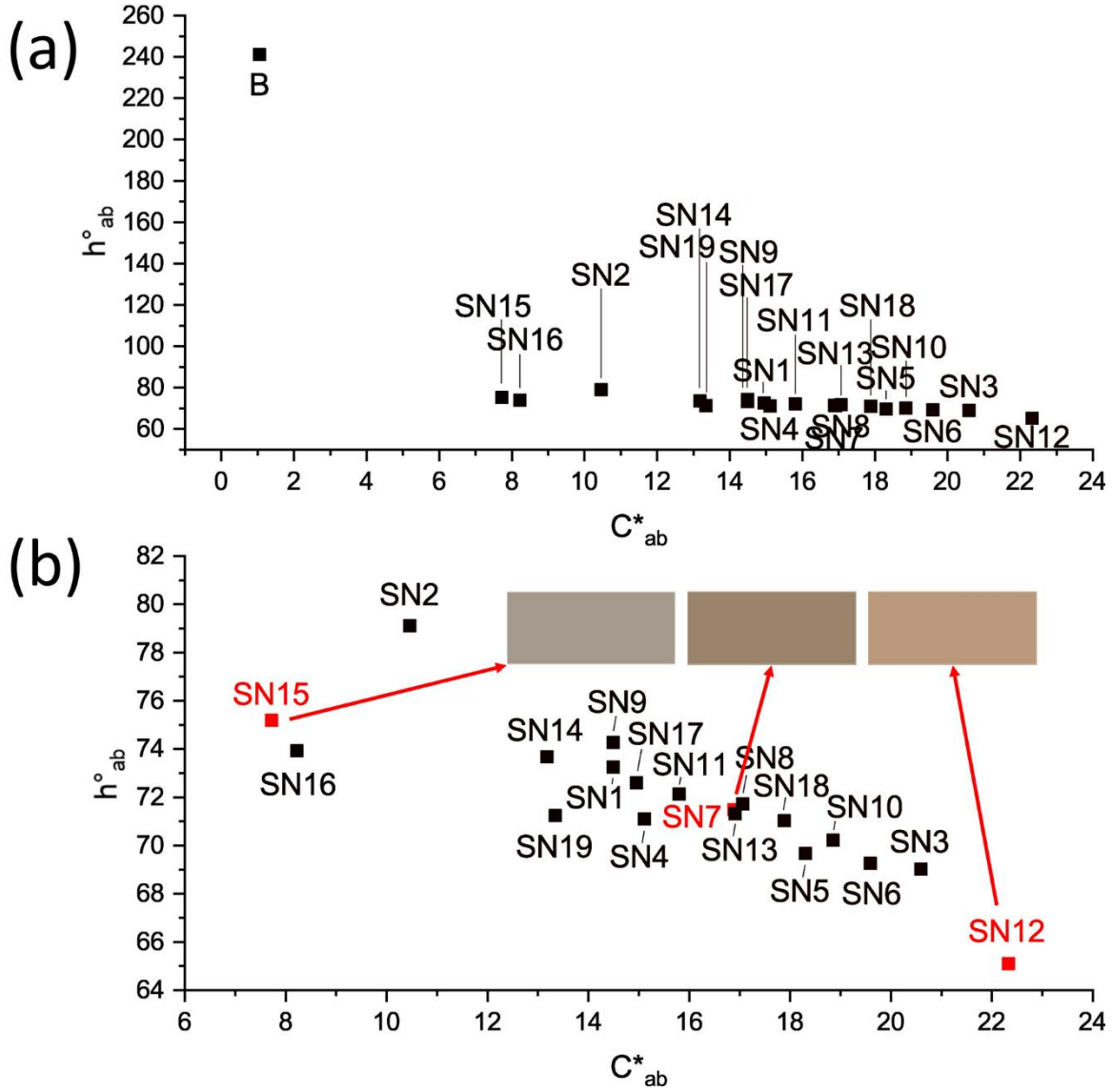
819

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



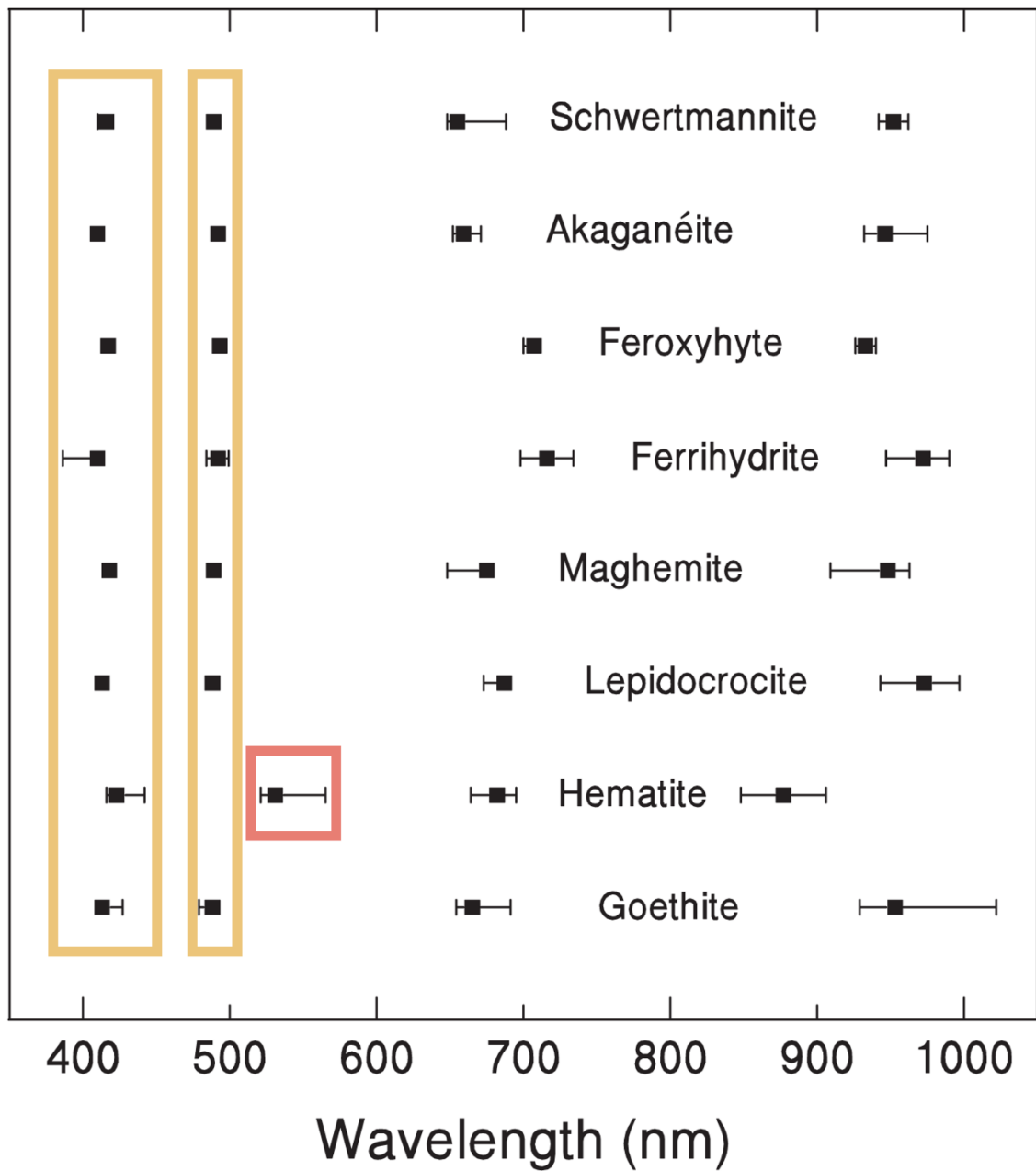
821

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

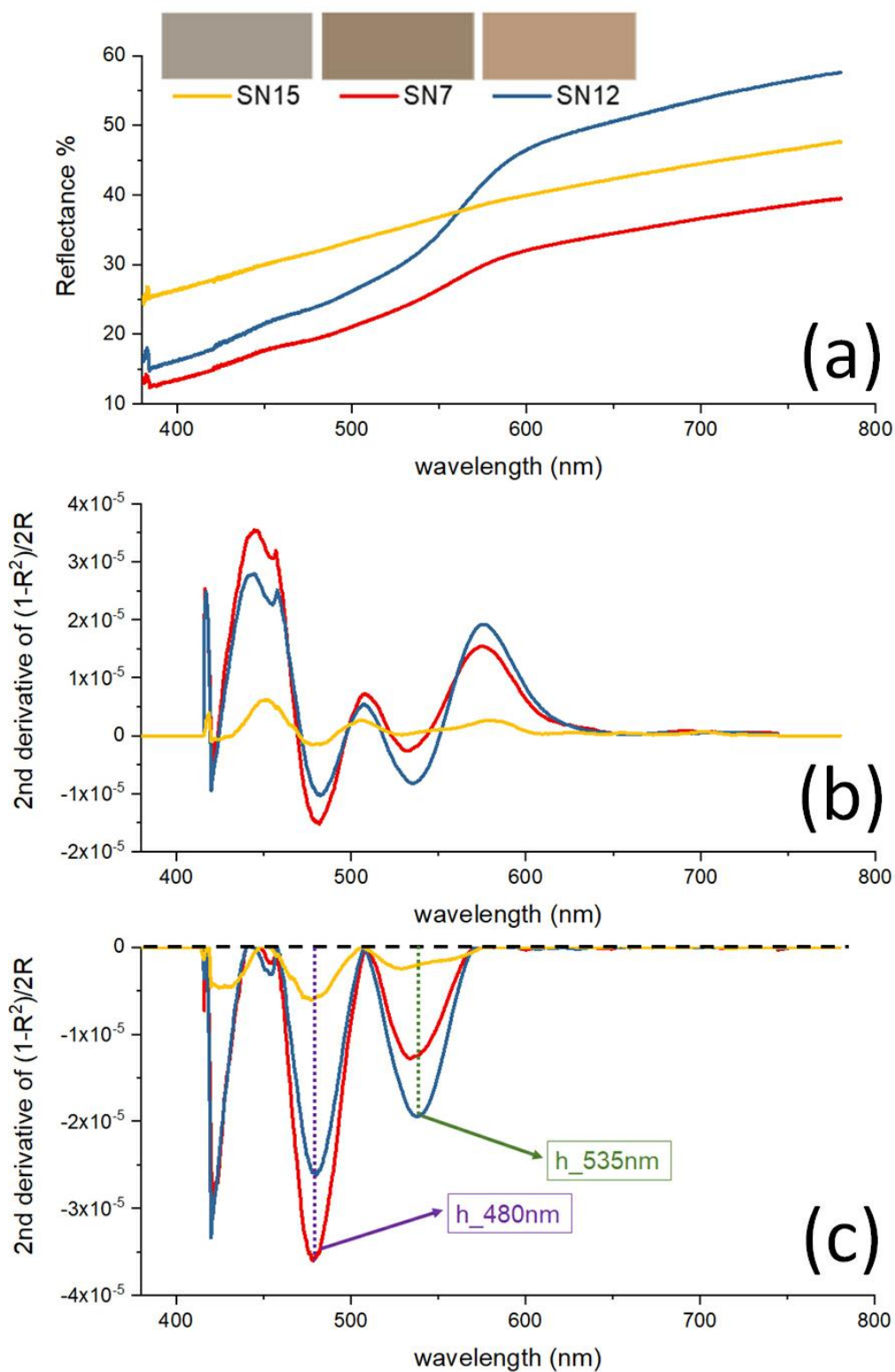
827 Figure 3. (a) Graphic representation of blank filter (B) and sampled filters (SN1, ...SN19) based on  
 828 chroma ( $C^*_{ab}$ ) and hue ( $h^{\circ}_{ab}$ ). (b) Zoomed plot on sampled filters. The colors of three specific samples  
 829 (SN15, SN7, SN12) are shown.



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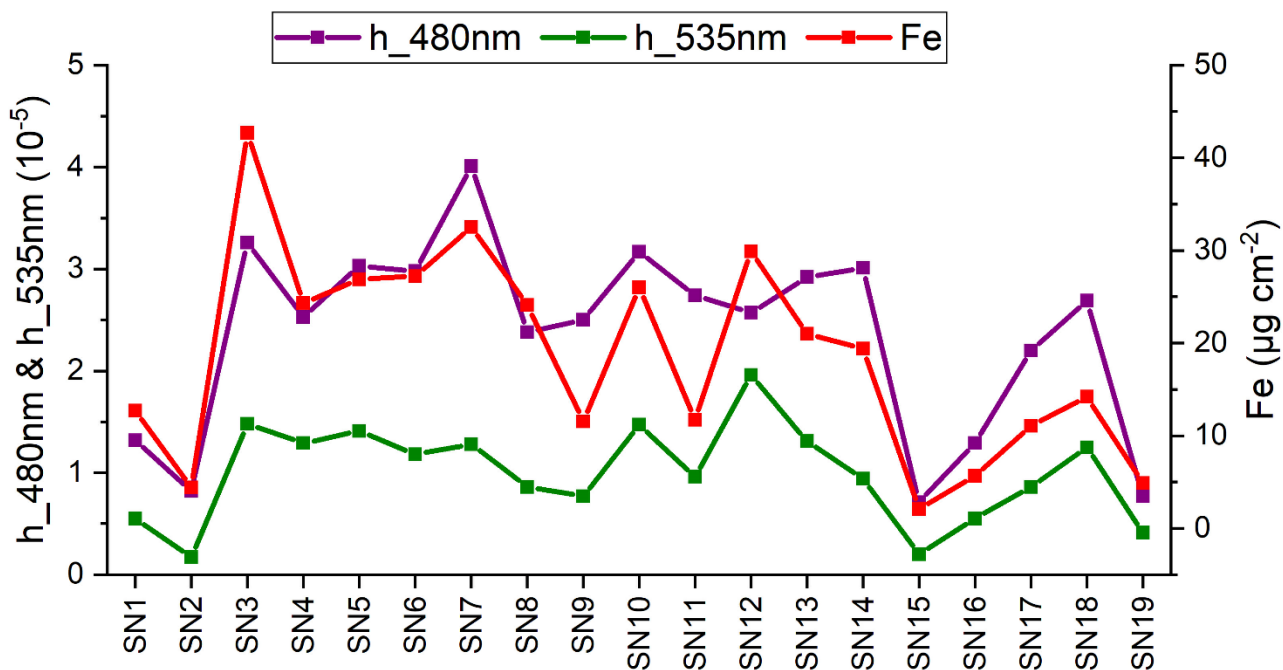
831 Figure 4. Median and range of the UV-Vis absorption bands of some iron oxide minerals in the second  
 832 derivative of K-M function spectra, adapted from Torrent & Barrón, 2008. It can be seen that below  
 833 600 nm, there are three absorption bands at around 420, 480, and 535 nm.





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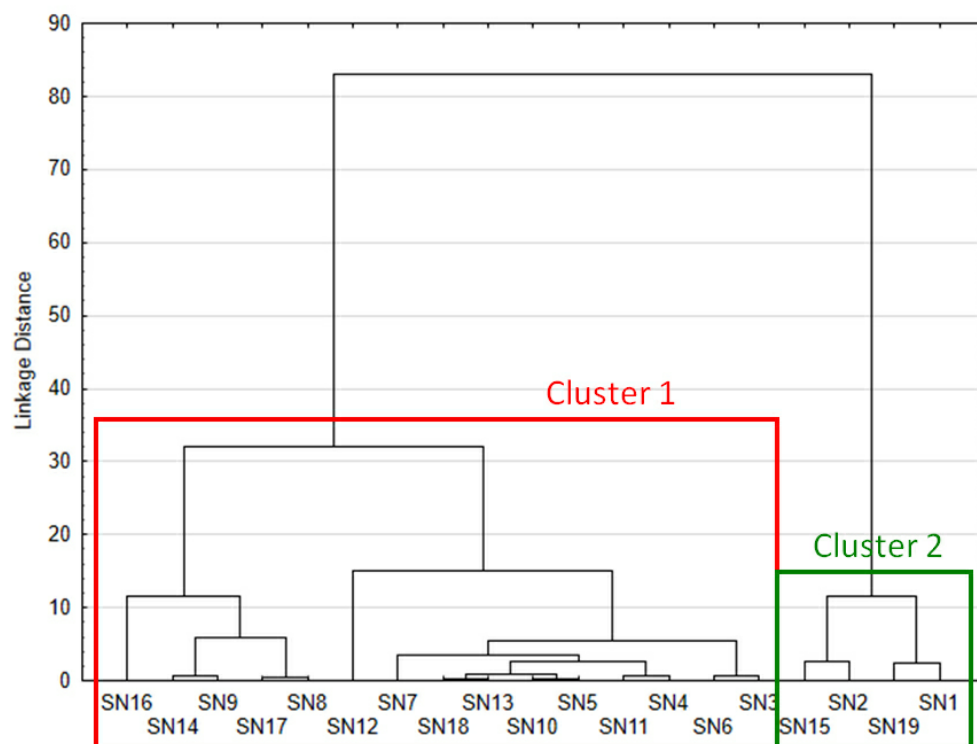
835 Figure 5. Reflectance spectra (a), Savitzky-Golay second derivative of Kubelka-Munk (K-M)  
 836 function spectra (b), and baseline subtraction and quantification of relevant peak heights (c) for SN15  
 837 (grayish filter), SN7 (brownish filter), and SN12 (reddish filter).



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).

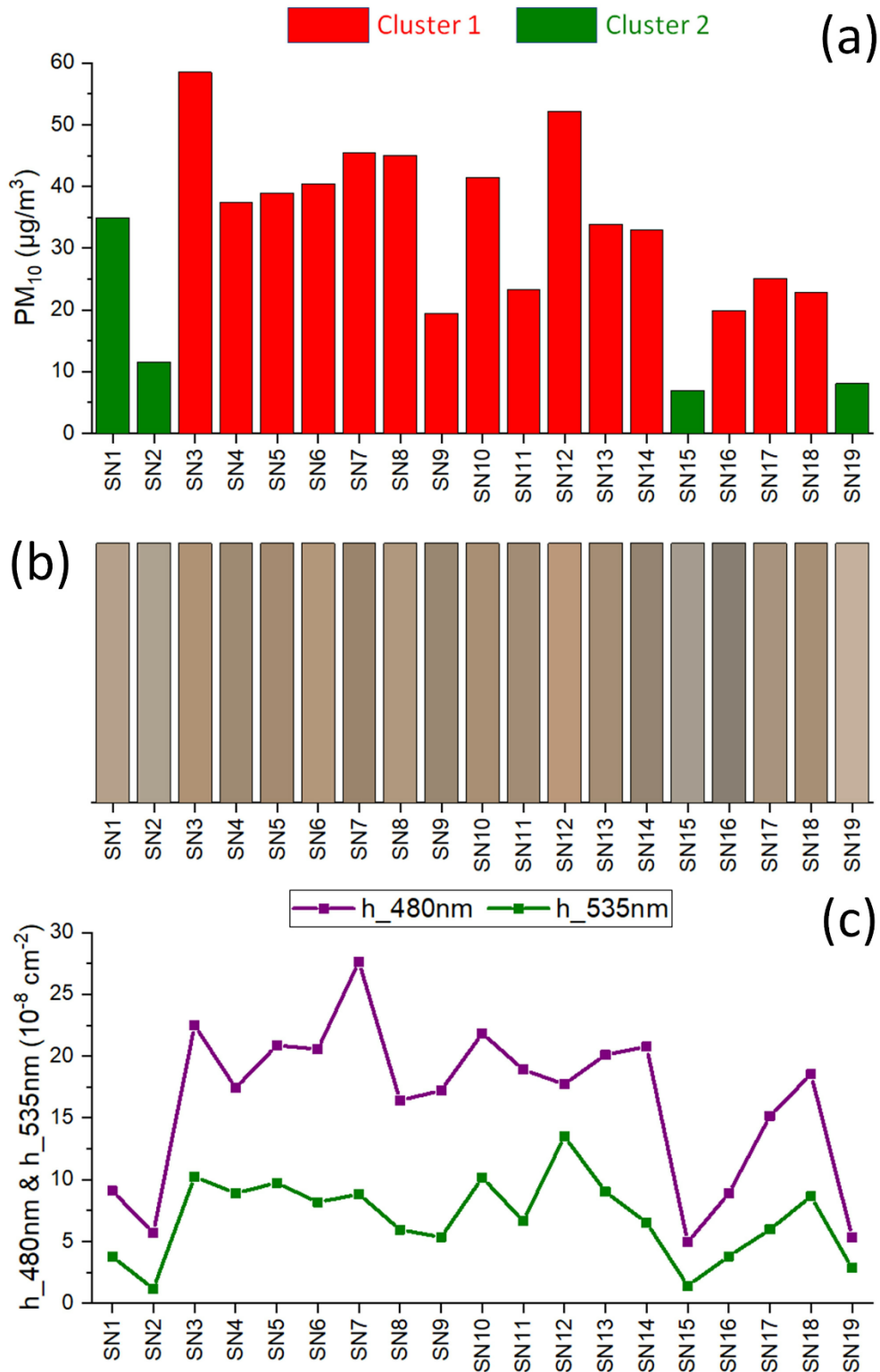


841

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

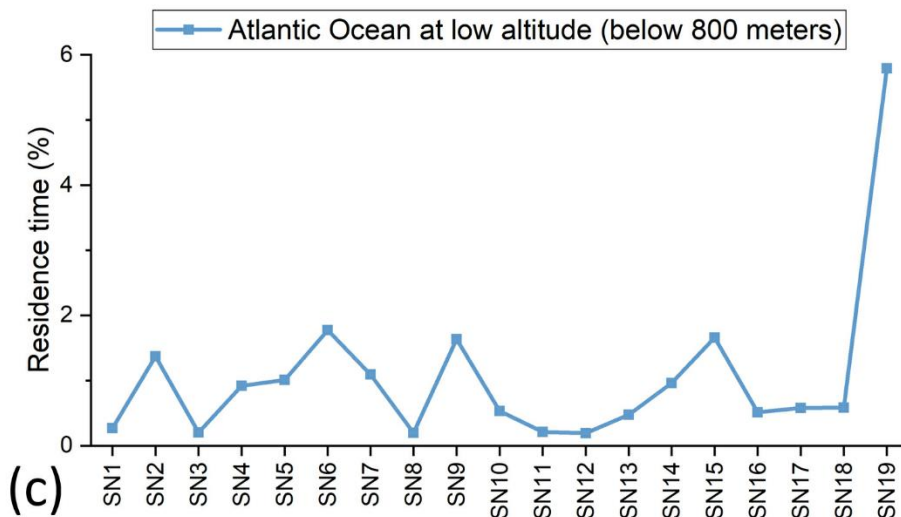
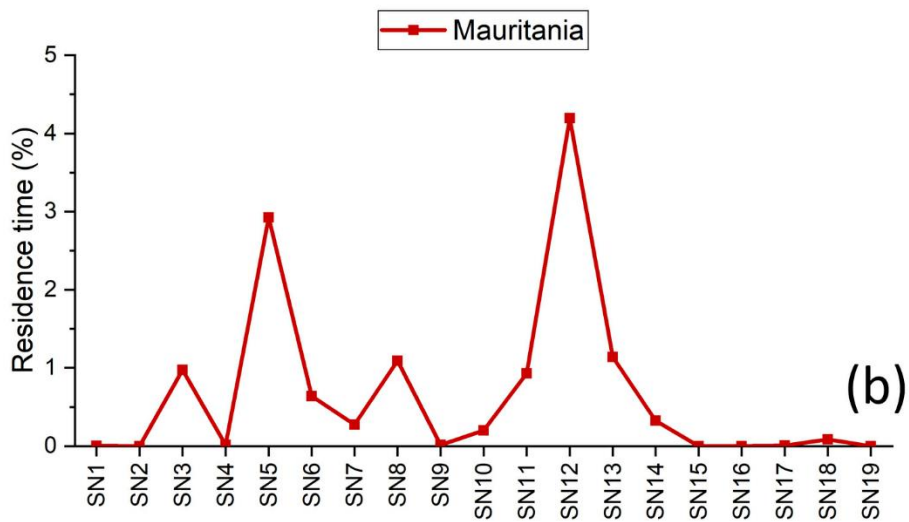
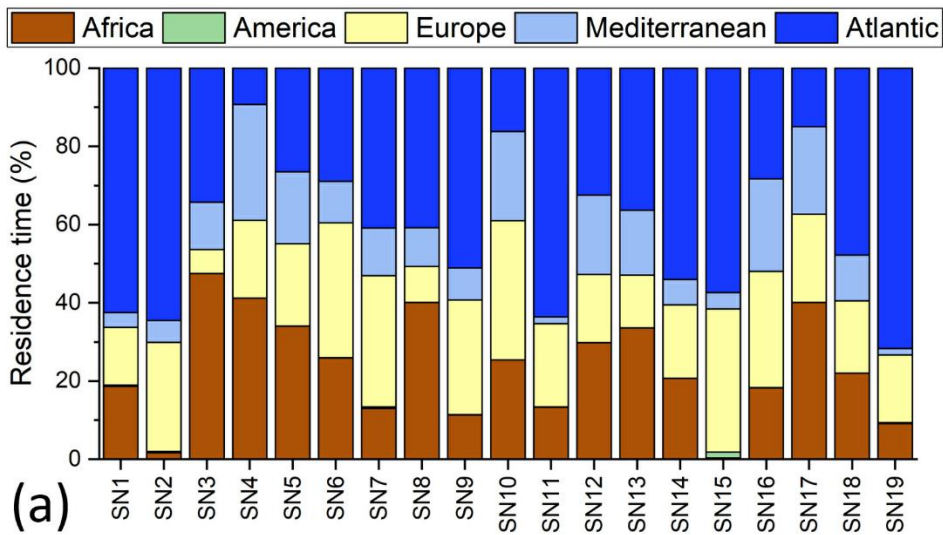
843 and semi-quantitative data of iron oxide minerals. The clustering solution with two clusters is

844 highlighted by squares.



845

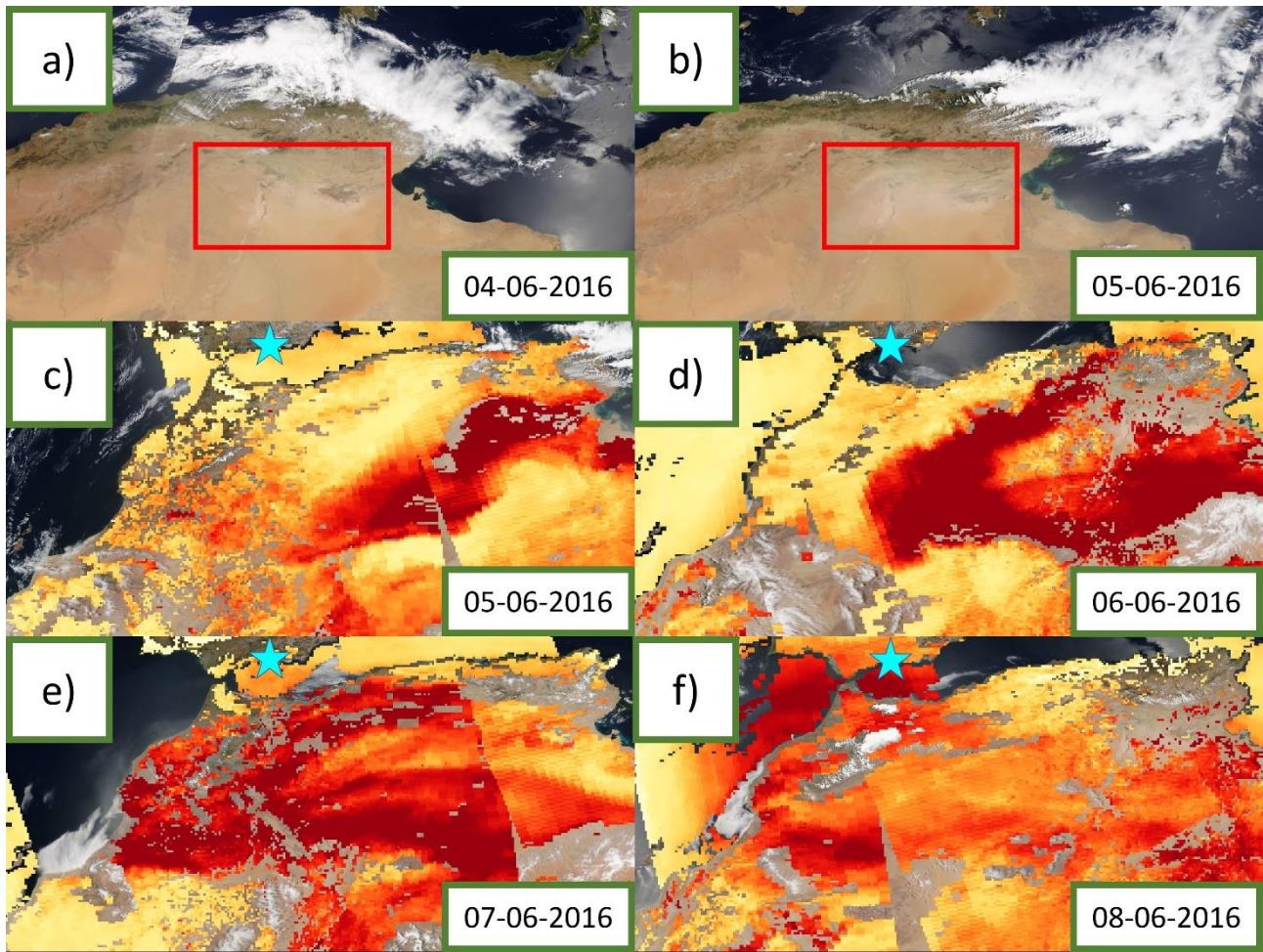
846 Figure 8. (a) Sample series of  $PM_{10}$  data ( $\mu g/m^3$ ) with the association of PM filters to the respective  
847 clusters (cluster 1 and cluster 2). (b) Display of PM filters colors, look at subsection 3.1. (c) Sample  
848 series of  $h_{480nm}$  (semi-quantitative data of several iron 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

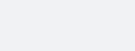



















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865

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 <sub>ab</sub> *	h <sub>ab</sub> °	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)	

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 <sup>-5</sup> )	h_530nm (10 <sup>-5</sup> )
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, Cab\* = CIELAB chroma, hab° = 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*	Cab*	hab°	Fe	PM10	h_480nm	h_535nm
L*	1.00						
Cab*	0.08	1.00					
hab°	-0.11	-0.90	1.00				
Fe	-0.20	0.85	-0.80	1.00			
PM10	-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:**  
915 Writing - Review & Editing; **Franco Lucarelli:** Validation, Resources, Writing - Review & Editing;  
916 **Silvia Nava:** Validation, Resources, Writing - Review & Editing; **Pedro J. Gómez-Cascales:**  
917 Investigation; **Jose A. G. Orza:** Software, Formal analysis, Resources, Writing - Review & Editing,  
918 Supervision, Project administration, Funding acquisition; **Laura Tositti:** Conceptualization, Writing  
919 - Review & Editing, Supervision

920

921 **Declaration of interests**

922

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:  
928

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