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► **To cite this version:**

Emma Caminiti, Cateline Lantz, S Besse, Rosario Brunetto, C Carli, et al.. Effects of ion irradiation on Mercury terrestrial analogues in the visible to mid-infrared. *Icarus*, 2024, 420, pp.116191. 10.1016/j.icarus.2024.116191 . hal-04682668

HAL Id: hal-04682668

<https://hal.science/hal-04682668v1>

Submitted on 30 Aug 2024

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Effects of ion irradiation on Mercury terrestrial analogues in the visible to mid-infrared

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Keywords: Mercury, Spectroscopy, Experimental techniques, Surface processes, Solar wind

Highlights:

- Ion irradiation is at the origin of an exponential darkening, a reddening and a flattening of VNIR spectra of Mercury volcanic terrestrial analogues.
- MIR features like the Christiansen feature and Reststrahlen bands are shifted due to ion irradiation.
- The alteration of spectroscopic features is closely influenced by the composition and will likely participate in the origin of spectral heterogeneities on Mercury.
- After a certain irradiation dose/time exposure spectral differences among units on Mercury may be limited by the saturation of the surface.

The surface of Mercury is subject to space weathering that complicates remote sensing data analysis. We present an experimental study performed on Mercury volcanic surface analogues to provide a better constraint on spectral alterations induced by solar wind. We used 20 keV He⁺ with fluences up to 5×10^{17} ions/cm² to simulate ion irradiation reaching the surface. Terrestrial ultramafic lava already identified as good analogues for Mercury were used: a boninite, a basaltic komatiite and a komatiite. Spectra were acquired in the visible to mid-infrared (VMIR) wavelength range, between 0.4 and 16 μm . Spectral alterations induced by irradiation are observed. In the visible to near-infrared (VNIR) samples show an exponential darkening, a reddening and a flattening of spectra. Above a certain irradiation dose (1×10^{17} ions/cm² in our conditions), the darkening reaches a plateau while the reddening and flattening do not show any definable trend. In the mid-infrared (MIR) we observe a shift of Reststrahlen bands towards longer wavelengths ($\leq 0.42 \mu\text{m}$). The Christiansen feature is shifted towards longer or shorter wavelengths according to the irradiation dose ($\leq 0.2 \mu\text{m}$). The spectral alteration is closely influenced by the composition. As Mercury's surface is compositionally heterogeneous, the degree of spectral alteration varies on the planet and putatively participates in the heterogeneous spectral properties of the surface. This work provides ground-truth data for future ESA-JAXA-

46 BepiColombo observations. The alteration of VMIR spectral features induced by ion
47 irradiation simulated in the laboratory will be used for future SIMBIO-SYS (Spectrometer and
48 Imaging for MPO BepiColombo Integrated Observatory SYStem) and MERTIS (Mercury
49 Radiometer and Thermal Infrared Spectrometer) data analysis.
50

51 1. Introduction

52 Mercury's surface, as well as other atmosphere-free bodies, is subject to space
53 weathering (SpWe). SpWe includes processes occurring in the space environment that alter
54 the observable mineralogical, physical and chemical properties of a surface over time. Solar
55 wind electrons and ions (Meyer-Vernet, 2007), solar photons, cosmic radiation and impacts
56 are part of this phenomenon. Mercury's global environment and SpWe were described by
57 Domingue et al. (2014) and compared with other airless bodies in Pieters and Noble (2016).
58 Contrary to the Moon and asteroids, Mercury's internal magnetic field influences solar wind-
59 surface interactions (Ogilvie et al., 1974; Lavorenti et al., 2023). However, this magnetic field
60 being weak solar wind access to the surface may be only slightly lower than at the Moon
61 (Domingue et al., 2014). Moreover, because of a faster SpWe than regolith overturn, we
62 expect more SpWe-derived products on Mercury than on the Moon (McKay et al., 1991;
63 Killen et al., 2007; Lucey and Riner, 2011).

64 SpWe, including ion irradiation, can introduce a bias in the analysis of remote sensing
65 data (Nittler et al., 2004). In the visible to the near-infrared (VNIR) wavelength range, SpWe
66 modifies spectral features by changing the spectral albedo (decreasing the reflectance:
67 darkening or increasing the reflectance: brightening), the slope of the reflectance spectrum
68 (increasing slope towards longer wavelengths: reddening, increasing slope towards shorter
69 wavelengths: blueing) and by reducing spectral contrasts (Brunetto and Strazulla, 2005;
70 Brunetto et al., 2006; Loeffler et al., 2009; Lantz et al., 2015, 2017; Pieters and Noble, 2016;
71 Pentillä et al., 2020; Rubino et al., 2024). In the mid-infrared (MIR) wavelength range, bands
72 are very sensitive to SpWe (Lantz et al., 2017; Brunetto et al., 2020). Irradiation induces an
73 alteration in band profiles and a shift of the Christiansen Feature (CF) as well as the
74 Reststrahlen Bands (RBs) and the Transparency Feature (TF) (Vernazza et al., 2013; Brunetto
75 et al., 2014, 2020; Jäggi et al., 2021; Lantz et al., 2015,2017). Understanding spectral
76 alterations induced by SpWe is necessary to analyse remote sensing data and decouple the
77 intrinsic properties of the surface from the effect of the space environment.

78 On Mercury, specific spectral features like hollows (Barraud et al., 2020, 2023) and
79 spectral differences between color units (Caminiti et al., 2023) can be explained by the
80 difference in compositions. Nevertheless, on a global scale spectral colour units (Robinson et
81 al., 2008; Denevi et al., 2009; Ernst et al., 2010; D'Incecco et al., 2015; Murchie et al., 2015)
82 and compositional units (Weider et al., 2012, 2014, 2015; Peplowski et al., 2015; Peplowski
83 and Stockstill-Cahill, 2019; Namur & Charlier, 2017; Vander Kaaden et al., 2017; Wilson et al.,
84 2019; Nittler et al., 2020) are not necessarily correlated highlighting that the composition is
85 not the only parameter at the origin of spectral heterogeneities (Weider et al., 2014; Riner
86 and Lucey, 2012). SpWe is not expected to produce the same effects on surfaces of different
87 compositions and has to be considered while studying the diversity of spectral and
88 compositional units and their origins (Fu et al., 2012; Riner and Lucey, 2012; Chrbolková et
89 al., 2021). The elemental composition of the surface will directly be affected by SpWe as
90 solar wind interaction processes lead to material removal such as light elements (Nittler et
91 al., 2001). Constraining the effect of SpWe is necessary to reconstruct the geological history
92 of the planet using compositional and spectral data.

93 Laboratory studies have been done to characterise good analogues representing the
94 diversity of geochemical units on Mercury (Carli et al., 2013; Morlok et al., 2017, 2019, 2021,
95 2023; Weber et al., 2019; Reitze et al., 2021; Mari et al., 2023). Additional works have shown
96 the importance of the physical properties of the surface in the understanding of spectral
97 properties (Jäggi et al., 2021; Bruschini et al., 2022; Pisello et al., 2023). The effects of
98 extreme temperatures on Mercury have also been investigated on varied analogues
99 (Benthley, 2005; Helebert et al., 2013; Maturilli et al., 2014; Bott et al., 2023a). However,
100 even if some studies analysed the effect of micrometeorite bombardment (Benthley, 2005;
101 Sasaki and Kurahashi, 2004; Weber et al., 2019; Thompson et al., 2021; Bott et al., 2023b)
102 less has been done concerning spectral alterations induced by the solar wind under
103 Mercury's conditions (Jäggi et al., 2021). Jäggi et al. (2021) have noted significant spectral
104 alterations induced by ion irradiation in the MIR highlighting the necessity to deeper
105 investigate SpWe under Mercury's conditions using a higher energy.

106 Ion irradiation on airless bodies has already been simulated in the laboratory
107 especially concerning asteroids and minerals (Brunetto and Strazzulla, 2005; Loeffler et al.,
108 2009; Fu et al., 2012; Vernazza et al., 2013; Brunetto et al., 2014, 2020; Lantz et al., 2015;
109 2017; Chrbolková et al., 2021; Jäggi et al., 2021; Rubino et al., 2024). Results from Mariner

110 10 (Dunne and Burgess, 1978) and the MErcury Surface, Space ENvironment, GEOchemistry,
111 and Ranging (MESSENGER) (McNutt et al., 2006; Solomon et al., 2007) missions have shown
112 that the surface of Mercury has been molded by volcanism (Denevi et al., 2013; Byrne et al.,
113 2016; Wright et al., 2021). From a morphological point of view, notably through the study of
114 Borealis Planitia, komatiite has quickly been proposed as a good lava analogue (Head et al.,
115 2011). The surface elemental composition of Mercury revealed by MESSENGER is
116 heterogeneous leading to the definition of several geochemical terranes (Weider et al.,
117 2012, 2014, 2015; Peplowski et al., 2015; Peplowski and Stockstill-Cahill, 2019; Wilson et al.,
118 2019; Vander Kaaden et al., 2017; Nittler et al., 2020). In the absence of direct
119 measurements of mineralogical composition, geochemical models have estimated the
120 mineralogy of the surface. They concluded that Mercury's surface can be compared to
121 terrestrial boninite and komatiite (Nittler et al., 2011; Stockstill-Cahill et al., 2012; Weider et
122 al., 2012, 2015; Charlier et al., 2013; Vander Kaaden and McCubbin, 2016; Vander Kaaden et
123 al., 2017; Peplowski and Stockstill-Cahill, 2019). In the absence of Mercury's samples, these
124 terrestrial lavas represent good analogues for laboratory measurements.

125 The upcoming observations of the ESA-JAXA-BepiColombo mission (Benkhoff et al.,
126 2021) will provide new datasets allowing the study of the surface of Mercury from the UV to
127 the IR. The Spectrometer and Imaging for MPO BepiColombo Integrated Observatory SYStem
128 (SIMBIO-SYS) (Cremonese et al., 2020) and the Mercury Radiometer and Thermal Infrared
129 Spectrometer (MERTIS) (Hiesinger et al., 2020) will map the surface of Mercury in the VMIR
130 and the effect of SpWe including ion irradiation is a crucial factor for the interpretation of
131 future data. Together these instruments will enable us to gain a deeper understanding of the
132 spatial definition and origin of spectral units on Mercury as well as the mineralogy of the
133 surface. VIHI, the hyperspectral imager of the SIMBIO-SIS payload and MERTIS will map the
134 surface between 0.4-2 μm and 7-14 μm respectively. This study aims to simulate, for the first
135 time, solar wind ion irradiation on Mercury's terrestrial rock analogues (boninite, basaltic
136 komatiite and komatiite) in the VMIR using strong energy and fluences. The main objectives
137 are to characterise the spectral alteration induced by ion irradiation and provide a
138 benchmark understanding of Mercury's surface weathering in preparation for BepiColombo.

139 2. Terrestrial analogues and Mercury's environment

140 2.1. Sample characteristics

141 As no direct information from returned samples is available, we used three terrestrial
142 ultramafic rocks that represent good analogues for Mercury's volcanic surface: a boninite, a
143 basaltic komatiite and a komatiite. Smooth plains on Mercury are often compared to flood
144 basalts and komatiite (Head et al., 2011) that formed the Large Igneous Provinces (LIPs) on
145 Earth. Both the komatiite and the basaltic komatiite come from the Caribbean plateau at
146 Gorgona Island known for its upper cretaceous ultramafic rocks sequence including
147 komatiite (Aitken and Echeverria, 1984; Serrano, 2009; Serrano et al., 2011; Carli et al.,
148 2013). The Gorgona Island is one of the least deformed and most recently accreted pieces of
149 the Caribbean Large Igneous Province (CLIP). The boninite comes from Troodos ophiolitic
150 Massif located on Cyprus island (Mari et al., 2023). The conditions on Mercury vary
151 significantly from what we observe on Earth. Our samples underwent weathering and
152 alteration processes similar to those on Earth, resulting in some spectral characteristics that
153 are unexpected on Mercury (Mari et al., 2013).

154 Boninite, basalt and komatiite are three high-Mg volcanic rocks. The main differences
155 are based on compositions and textures (spinel texture for the komatiite: preferential
156 orientation of dendritic olivine or pyroxene crystals). Boninites are defined by $\text{MgO} > 8 \%$,
157 $\text{TiO}_2 < 0.5 \%$ and $\text{SiO}_2 > 52 \%$ in comparison with komatiites defined by $\text{MgO} > 18 \%$, $\text{TiO}_2 <$
158 1% and $30 < \text{SiO}_2 < 52 \%$. Komatiites have a $\text{Na}_2\text{O} + \text{K}_2\text{O} < 2 \%$ whereas basalts have a
159 $\text{Na}_2\text{O} + \text{K}_2\text{O} > 2 \%$ (Le bas, 2000). A basaltic komatiite is compositionally close to a komatiite
160 with a lower MgO and higher SiO_2 . Regarding the elemental composition (Table 1) the
161 boninite used in the present work belongs to the low-Si boninite group and the two other
162 samples are well characterised by a basaltic komatiite and a komatiite (Le bas et al., 2000;
163 Pearce and Reagan, 2019).

164

Elemental composition	Terrestrial analogues (Serrano, 2009; Mari et al., 2023)			Geochemical units from Vander kaaden et al. (2017)						
	Boninite	Basaltic komatiite	Komatiite	CB	NP-LMg	NP-HMg	HMR	HMR-CaS	HAI	IT
wt%										
SiO_2	43.09	49.329	43.350	58.00 (4.75)	58.87 (7.43)	53.44 (7.49)	50.52 (7.04)	49.27 (5.95)	53.94 (4.23)	54.38 (8.05)
TiO_2	0.42	1.370	0.425	0.45	1.38	1.25	1.18	1.15	1.26	1.27

				(0.16)						
Al ₂ O ₃	13.78	13.759	10.846	16.35 (0.97)	11.79 (2.40)	12.24 (1.51)	9.80 (1.36)	9.04 (1.82)	16.34 (0.57)	13.68 (1.80)
FeO	18.83	10.094	11.330	0.97 (0.21)	1.57 (0.47)	1.97 (0.31)	1.94 (0.51)	2.14 (0.31)	0.94 (0.31)	1.79 (0.34)
MgO	8.82	8.487	19.758	12.86 (2.28)	12.13 (2.84)	19.75 (3.90)	23.54 (2.86)	24.52 (2.32)	16.06 (1.47)	17.46 (4.62)
CaO	11.52	11.246	8.875	5.62 (0.63)	5.27 (1.23)	5.43 (1.20)	6.82 (1.70)	7.57 (1.35)	5.58 (1.59)	5.59 (0.93)
Na ₂ O		2.746	0.695	3.51 (0.01)	6.47 (0.04)	3.51 (0.01)	3.51 (0.01)	3.51 (0.01)	3.51 (0.01)	3.51 (0.01)
K ₂ O	1.26	0.196	0.033	0.09 (0.02)	0.18 (0.04)	0.24 (0.04)	0.16 (0.03)	0.16 (0.03)	0.16 (0.03)	0.11 (0.02)
Cr ₂ O ₃	0.27									
MnO	1.59	0.171	0.188	0.70	0.71	0.65	0.61	0.59	0.65	0.66
NiO	0.16									
P ₂ O ₅		0.115	0.037							
CrO				0.71	0.72	0.65	0.62	0.60	0.66	0.66
S				1.50 (0.50)	1.82 (0.49)	1.75 (0.57)	2.60 (0.60)	2.89 (0.29)	1.83 (0.07)	1.76 (0.37)

165

166 *Table 1: Elemental composition of the terrestrial analogues in comparison with geochemical*
167 *units on Mercury defined by Vander Kaaden et al. (2017). CB: the interior smooth plains of*
168 *the Caloris basin, NP-HMg: a subset of the Northern volcanic Plains (NP) with relatively high*
169 *Mg content, NP-LMg: a subset of the NP with relatively low Mg content, HMR: the high-Mg*
170 *region, HMR-CaS: a sub-region of the HMR with the planet's highest Ca and S contents. HAl:*
171 *the high-Al regions. IT: intermediate terranes.*

172

173 The boninite is composed of orthopyroxene (66.8 wt% enstatite) and olivine (33.1 wt%,
174 forsterite) (Mari et al., 2023). The composition of the basaltic komatiite and the komatiite is
175 slightly more diverse (Serrano, 2009). The basaltic komatiite contains clinopyroxene (23-29
176 wt%, mainly Mg-rich clinopyroxene), olivine (15-25 wt%), plagioclase (19-27 wt%), volcanic
177 glass (23-26 wt%) and accessory minerals. The komatiite is composed of clinopyroxene (15
178 wt%, mainly Mg-rich clinopyroxene), olivine (35 wt%), plagioclase (21 wt%), volcanic glass
179 (12 wt%) and accessory minerals.

180 The MgO differences, and more generally the elemental compositional differences,
181 between the boninite, the basaltic komatiite and the komatiite can be used to investigate
182 the spectral differences between volcanic units on Mercury. We used geochemical units and
183 average compositions defined by Vander Kaaden et al. (2017) to compare our terrestrial
184 analogues with units on Mercury. The SiO₂ values are lower than Mercury's composition,
185 except for the basaltic komatiite, as well as the MgO of the boninite and basaltic komatiite.
186 The lower SiO₂ content indicates that our samples come from a more primitive magma. The
187 boninite and basaltic komatiite reflect the composition of Caloris interior volcanic smooth
188 plains (CB), high-Al regions (HAI) and a part of the Northern Plains lower in MgO (NP-LMg)
189 (Table 1). In comparison, the MgO composition of komatiite is closer to the High Magnesium
190 Region (HMR) and High MgO part of the Northern Plains (NP-HMg). The basaltic komatiite
191 almost coincides with the HMR-CaS region, especially regarding the SiO₂ composition
192 suggesting a reliable analogy even if the MgO content is lower. The IT is the most common
193 geochemical unit on Mercury. The MgO concentration of the IT is closer to the komatiite but
194 the best analogue seems to be the basaltic komatiite according to the SiO₂, and Al₂O₃
195 contents. The basaltic komatiite is the sample that fits the most units on Mercury in
196 comparison with the boninite and the komatiite.

197 One of the most distinctive features of Mercury's surface is its low iron content.
198 Earth-based observations (Vilas, 1985; Blewett et al., 2002; Sprague et al., 2002; Warell,
199 2002), MESSENGER flyby measurements (McClintock et al., 2008) and MESSENGER orbital
200 data (Evans et al., 2012; Izenberg et al., 2014; Weider et al., 2014; Nittler and Weider, 2019;
201 Nittler et al., 2020) have shown that Mercury's surface contains very little amount of iron in
202 contrast to its high bulk composition. Fe-O bonds in silicates produce a spectral reflectance
203 absorption band at ~ 1µm (Burns, 1993). On Mercury, the global lack of ~ 1µm absorption
204 band indicates that there are only a few percent of FeO in silicates. The exact amount
205 remains controversial and summarised in Domingue et al. (2014), noting for example a FeO
206 maximum abundance of < 1 wt% according to Klima et al. (2013), about 4 wt% by Nittler et
207 al. (2011) and Izenberg et al. (2014) or around 8 wt% according to Warell et al. (2010).
208 Nevertheless, the iron surface composition is heterogeneous and Warell et al. (2006) noticed
209 a shallow absorption band centred at 1.1 µm localised on Mercury (Boynton et al., 2007).
210 The comparison of MESSENGER observations with reflectance models and compositional
211 data suggests that surficial iron has mostly been converted to nanometer-scale metallic iron

212 particles (called: nanophase reduced iron or submicroscopic iron particle and noted
213 respectively: npFe⁰ or SMFe) due to SpWe (Noble et al., 2007; Lucey and Riner, 2011; Weider
214 et al., 2014; Trang et al., 2017). These particles bias the determination of available FeO in
215 the crust. With a better spatial resolution and Signal-to-Noise Ratio (SNR) BepiColombo is
216 eagerly awaited to precise the FeO content and spectral signature.

217 Even if our samples have undergone earthlike weathering and aqueous alteration and even
218 if the FeO compositions are above the estimations for Mercury's surface, they remain
219 reasonable and accessible candidates as natural terrestrial rock analogues. For a good
220 quality comparative analysis with Mercury data, it is important to avoid spectral features
221 that are not observable on the Mercury spectra at this time like the 1 μm absorption band.
222 In addition to the low iron content, SpWe could also explain the absence of features in
223 Mercury's VNIR spectra. We will therefore not make a detailed study of the band, but will
224 briefly discuss the effect of ion irradiation on the lack of features.

225

226 2.2. Hermean solar wind conditions

227 Mercury is surrounded by solar wind, solar wind ions being the major contributors to
228 SpWe on Mercury. Large portions of the surface are exposed to the solar wind because of
229 the lack of a dense atmosphere, the weak internal magnetic field and the strong solar wind
230 environment. Solar wind ions are H⁺ and He²⁺ with a weak contribution of heavy elements
231 (Bame et al., 1975). Typically, protons impact Mercury with an energy between ~ 500 eV to
232 ~ 1.5 keV, higher than ~ 1 keV on the Moon. Concerning electrons, they are mostly found in
233 a range of 0.1 to 10 keV (Lavorenti et al., 2023). The energy can reach 10 KeV, with protons
234 and electrons gaining energy at magnetic reconnection when they cross Mercury's cusp
235 (footprints of open magnetic field lines, magnetic lines that have one end connected to the
236 solar wind) regions (Masseti et al., 2007; Lavorenti et al., 2023). High-energy ions are also
237 emitted from the Sun through solar flares and active regions with significant effects on
238 surfaces (Johnson et al., 1990; Brunetto et al., 2014; Matsumoto et al., 2015). In general, the
239 plasma precipitation on Mercury is very heterogeneous (in latitude and longitude) and will
240 mostly depend on solar wind conditions as well as solar wind-magnetosphere interactions
241 (Lavorenti et al., 2023). Typically, a surface of $\sim 10^{12}$ - 10^{13} m² facing the Sun receives a flux of
242 $\sim 1.5 \times 10^{12} - 2 \times 10^{13}$ ions/m² s (Killen et al., 2007; Wurz et al., 2010; Massetti et al., 2007;

243 Lavoretti et al., 2023). The surface of Mercury is exposed to a fluence of $\sim 10^{14}$ protons/cm²
244 integrating over two Mercury orbits (176 Earth days) (Lavoretti et al., 2021).

245 3. Protocols and experimental setups

246 3.1. Pellets formation

247 Due to experimental constraints, we performed our analysis on pellets. Pellets
248 simulate an intermediate case between a fine regolith and a rock. A spectrum obtained from
249 a pellet will be bluer than one obtained from a powder of the same composition and grain
250 size (Carli et al., 2013; Vernazza et al., 2013). Mercury's regolith is assumed to be dominated
251 by fine-grained materials but we do not have precise knowledge of the regolith's grain size
252 (Langevin et al., 1997). Terrestrial samples were therefore crushed into powder of grains size
253 $\leq 36 \mu\text{m}$, close to an approximation of Mercury's regolith (Sprague et al., 2002; Warell et al.,
254 2010). We acknowledge that pressing the powders into a pellet reduced the surface
255 roughness, the grain size, and both the intra- and inter-granular porosity of the
256 uncompressed powder, however the extent of these possible alterations was not
257 determined. Pellets were prepared at the Institut d'Astrophysique Spatiale (IAS, Orsay,
258 France) by pressing ~ 270 mg of powder using a Specac manual hydraulic press. Each pellet
259 was pressed under 6 tons during 5 minutes. The resulting pellets are 13 mm in diameter,
260 about 1-2 mm thick and homogeneous. The produced pellets are not sufficiently thick and
261 are likely to move in the sample holder during the experiment. To avoid this, we prepared
262 KBr pellets of approximately the same size to place below the sample in the sample holder.
263 KBr pellets were created by pressing 400 mg of KBr at 5 tons for 4 minutes. From the studied
264 material, two pellets each of komatiite, basaltic komatiite and boninite were created for the
265 two experiments outlined in Section 3.2.

266

267 3.2. Ion irradiation

268 Ions of several species and energies are involved in the solar wind irradiation of
269 Mercury's surface. However, due to experimental limitations in time and resources, we
270 decided to focus on He⁺ at 20 keV. Irradiations performed in this work are thus not an
271 exhaustive reproduction of solar wind ions reaching Mercury. Hydrogen is the main
272 component ($\sim 96\%$) of solar wind while helium is the second most abundant species ($\sim 4\%$).

273 Unlike hydrogen, helium ions He^+ are non-reactive, allowing us to avoid complications with
274 implantation-induced chemistry (e.g. Brucato et al., 1997; Liu et al., 2012; Bandfield et al.,
275 2018) making them our first choice for this study. The solar wind is composed mainly of H^+
276 and He^{2+} , thus the use of He^+ instead of He^{2+} implies a much lower potential sputtering
277 effect (see, e.g. Hijazi et al., 2017; Szabo et al., 2020).

278 We used an energy of 20 keV (5 keV/amu) similar to those of suprathermal tail and
279 pickup ions, in between the fast solar wind and the low-energy range of solar energetic
280 particles (Mewaldt et al., 2007; Mason et al., 2008). The energy we use (5 keV/amu) is
281 approximately five times greater than the energy of the slow solar wind (~ 1 keV/amu). The
282 contribution of high-energy helium ions to SpWe is not negligible and can significantly
283 modify the surface of an airless body (Matsumoto et al., 2015). Moreover, the weathering
284 timescale is longer at high energy than at the average energy of 1-4 keV (Brunetto et al.,
285 2014). The energy of 20 keV is of particular interest as it has been highlighted by Jäggi et al.
286 (2021) that the effect of average solar wind is almost negligible on Mercury analogues.
287 Fluences can be adapted from previous studies based on meteorite and mineral
288 measurements. Experimental studies of SpWe on asteroids with stronger energy (40 keV)
289 and helium ions have shown that a fluence of $< 1 \times 10^{17}$ ions/cm² allowed an exponential
290 darkening and reddening of carbonaceous chondrites (Lantz et al., 2017). However, the
291 asymptote indicating a potential saturation was not reached. Furthermore, Chrbolková et al.
292 (2021) have shown only a weak spectral alteration of silicates in the visible at a fluence of $1 \times$
293 10^{17} ions/cm², and an energy of 20 keV, due to an insufficient fluence. To try to reach a
294 putative saturation of our samples discernible in spectra we decided to irradiate up to $5 \times$
295 10^{17} ions/cm².

296 Ion irradiations were performed with the SIDONIE electromagnetic isotope separator
297 (Chauvin et al., 2004) (CSNSM, Orsay, France) interfaced with the INGMAR (Irradiation de
298 Glaces et Météorites Analysées par Réflectance VIS-IR, IAS-CSNSM, Orsay, France) setup
299 through which in-situ reflectance spectroscopy measurements were done. We used He^+ at
300 20 keV under vacuum ($P \sim 10^{-7}$ mBar) with the following fluences 1×10^{16} , 3×10^{16} , 6×10^{16} ,
301 1×10^{17} ions/cm² including a measurement before irradiation. Additional measurements
302 were done in a second irradiation campaign reaching 5×10^{17} ions/cm² including several
303 intermediate doses according to the sample. The intermediate doses were not the same for
304 all the samples. We performed measurements before irradiation for each sample. The

305 irradiated komatiite was measured after fluences of 2.89×10^{17} ions/cm² and 5×10^{17}
306 ions/cm²; the basaltic komatiite was measured after 3×10^{17} ions/cm², 4×10^{17} ions/cm², and
307 5×10^{17} ions/cm² finally, concerning the boninite we performed measurements after
308 fluences of 4×10^{17} ions/cm² and 5×10^{17} ions/cm². The ion flux was $\sim 10^{13}$ ions/ (cm²s) on
309 average. For each sample, a part of the pellet (slightly less than half of it) was preserved
310 from irradiation by using a copper mask. This allowed us to keep a piece of pristine material
311 and perform additional measurements later on (especially ex situ in the MIR) to compare
312 before (fresh) and after irradiation spectra.

313 Unlike asteroids, Mercury is partially and unequally protected by its magnetosphere
314 making it difficult to relate ion irradiation simulations and alteration time scale. For a 20 keV
315 solar wind flux on Mercury, and considering only helium precipitation, the fluences would
316 correspond to an exposure time of ~ 16 thousand to 8 million years (see Lavorenti et al.,
317 2023 and Raines et al., 2022 for fluxes and energies). The shorter time corresponds to the
318 areas of the surface with the highest precipitation fluxes (and our minimum fluence), while
319 the longer time corresponds to the areas of the surface with the lowest (and our maximal
320 fluence). We would like to point out that these values correspond to a surface irradiated
321 only under our conditions. As Mercury's surface is irradiated by other ions at other energies,
322 the time scale will be shorter. Moreover, this is without taking into account the additional
323 components of SpWe.

324

325 3.3. VNIR measurements

326 Using the INGMAR setup, bidirectional reflectance spectroscopy was performed from
327 the visible (0.4-1.05 μm) to near-infrared (0.9-2.5 μm). Measurements were acquired after
328 each step of irradiation directly in the vacuum chamber at room temperature. Both VIS and
329 NIR measurements were performed on the same spot (Lantz et al., 2017). The illumination
330 spot sizes are 20 mm in the VIS and 12 mm in diameter in the NIR with a collecting spot size
331 of 3-4 mm. In the VIS we used a grating spectrometer (Maya2000Pro, Ocean Optics) with a
332 spectral resolution of 0.5 nm. In the NIR spectra were acquired using a Fourier Transform
333 spectrometer (Tensor37 Bruker) with a spectral resolution of 32 cm⁻¹. Spectra were collected
334 with a fixed emission angle $e=15^\circ$ and $i=15^\circ$ ($\alpha=20^\circ$) in the VIS in comparison with $i=20^\circ$
335 ($\alpha=15^\circ$) in the NIR. To calibrate the absolute reflectance, we used a Spectralon standard
336 (with 99% Lambertian reflectance; Labsphere). There is an offset between the VIS and NIR

337 spectra mainly due to a geometry effect as well as a slight non-linearity of the detectors. The
338 final spectra presented in this work were corrected by the Spectralon hydration and merged
339 in VNIR final spectra using a region of common wavelengths.

340

341 3.4. MIR measurements

342 MIR measurements, between 2 and 16 μm , were performed ex-situ on the samples
343 after the final irradiation dose. Those analyses were conducted at the Synchrotron SOLEIL
344 (France) using the SMIS beamline (Spectroscopy and Microscopy in the Infrared using
345 Synchrotron) (Dumas et al., 2006). We used a microspectrometer (model Cary 670/620)
346 having its internal Global light source and a MCT detector cooled with liquid nitrogen.
347 Reference spectra were collected on a gold sample. Measurements were done at ambient
348 temperature using a 15x objective; the spot size was between 160 and 400 μm^2 and the
349 spectral resolution was 4 cm^{-1} . Measurements on the non-irradiated part of each sample
350 were made accurately by removing the copper cover (see 3.2) at the very last moment.
351 Furthermore, the irradiated and non-irradiated sides were identifiable by slight differences
352 in colour. Spectra presented in this work are an average of three to five measurements
353 performed on several areas both for the non-irradiated and irradiated part of each sample.
354 Average spectra allow us to take into account the heterogeneities of the sample in the
355 interpretation. Despite this, certain differences in reflectance intensity may be linked to the
356 choice of the sampled area. To avoid any interpretation bias, we normalised the spectra to
357 the largest peak.

358 4. Results

359 4.1. Visible to Near-infrared

360 We obtained one VNIR spectrum for each sample at each fluence up to 1×10^{17}
361 ions/ cm^2 (Figure 1 A, C, E). The boninite exhibits a global reflectance that is lower than the
362 basaltic komatiite, which in turn is lower than that of the komatiite. All samples exhibit a
363 broad $\sim 1 \mu\text{m}$ band consistent with mafic mineralogy. Moreover, all samples show hydration
364 bands associated with aqueous alteration at $\sim 1.4 \mu\text{m}$ and $\sim 1.9 \mu\text{m}$ observable for the
365 boninite. Alteration can also explain the minor features at $\sim 0.65 - 0.75 \mu\text{m}$ and $\sim 2.2 - 2.4$
366 μm . Globally, the spectral alteration due to ion irradiation is weaker in the NIR. In the VIS we

367 observe three major alterations of spectroscopic features upon irradiation (Figure 1 B, D, F):
 368 a decrease in reflectance (darkening), an increase of the spectral slope with wavelengths
 369 (reddening) and a less important curvature in the near-UV to visible (flattening). The
 370 absorption band at $\sim 1 \mu\text{m}$ underwent only minor alterations after irradiation. We do not
 371 observe any significant shift or distortion of the band. After an irradiation of 1×10^{17}
 372 ions/cm², the minimum of the band is shifted of ~ 0.3 , 0.02 , and $0.005 \mu\text{m}$ respectively for
 373 the boninite, the basaltic komatiite and the komatiite. However, as the main absorption
 374 band at $\sim 1 \mu\text{m}$ was not observed yet on Mercury, spectral parameters used in this study are
 375 defined outside this absorption band. The irradiation conditions and analogues used in this
 376 study do not explain the absence of an absorption band in the VNIR spectra on Mercury. Ion
 377 irradiation may not be the main process of reducing spectral band signatures on Mercury
 378 (Lavorenti et al., 2023).

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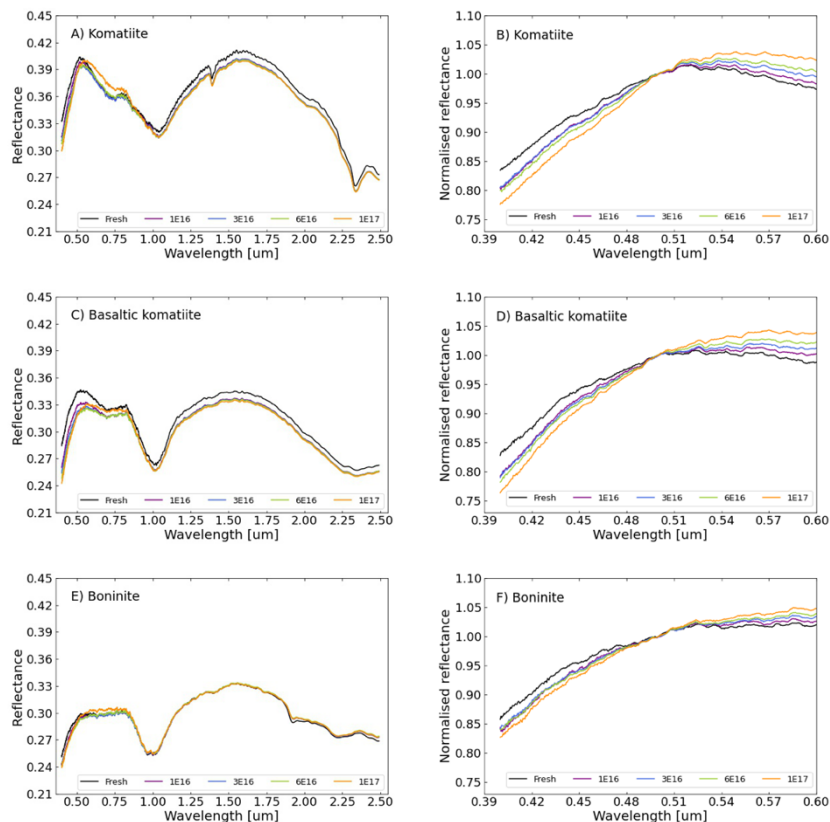
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395 *Figure 1:* A) C) E) VNIR spectra for the komatiite, basaltic komatiite and boninite respectively
 396 according to the fluence up to 1.10^{17} ions/cm². B) D) F) Normalised spectra at $0.5 \mu\text{m}$. The
 397 normalization highlights the slopes and shape of spectra according to the fluence.

398

399 Spectral parameters have been defined to constrain the alterations induced by irradiation
 400 (Table 2). We used the absolute reflectance at 0.45 μm (R450) as well as the slope
 401 (calculated by linear fit) between 0.41 and 0.5 μm (S410-500). An additional parameter
 402 (curvature) adapted from Barraud et al. (2020) was used to highlight the curvature alteration
 403 on the spectra between 0.41 and 0.5 μm . These spectral parameters have been chosen to
 404 avoid the 1 μm absorption band while being close to parameters used in remote sensing
 405 data analysis on Mercury (Barraud et al., 2020; Caminiti et al., 2023).

406

Spectral parameter	Spectral range [μm]	Mathematical expression	Description
R450	0.45	-	Reflectance at 0.45 μm
S410-500	0.41-0.5	Reflectance (λ)=S410* λ +b	Obtained by linear fit
Curvature	0.41-0.5	Reflectance (λ)= curvature* λ^2 +b λ +c	Obtained by polynomial fit (degree 2)

407 *Table 2: Parameters used to describe the alteration of spectroscopic features induced by ion*
 408 *irradiation.*

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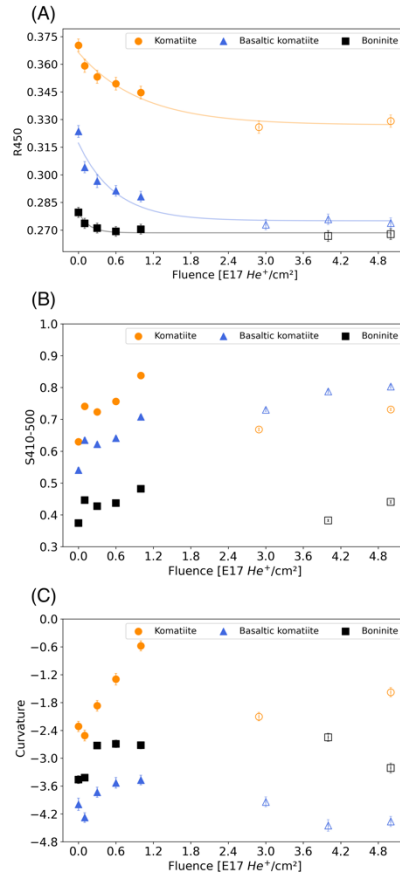
410 After the first irradiation experiment, giving results until 1×10^{17} ions/cm², and in order to
 411 clarify the trends, additional measurements were performed in a second experiment
 412 reaching fluences up to 5×10^{17} ions/cm² (see 3.2). It is worth noting that pellets made from
 413 the same fresh material have different initial (before irradiation) reflectance, slope and
 414 curvature between the first and second experiments. Any differences in composition
 415 between pellets of the same material would not affect the data presented here, due to the
 416 large collection area (3-4 mm diameter). The observed difference could stem from variations
 417 in crystal orientation and surface morphology of the pellets before ion bombardment.
 418 However, these discrepancies do not impact our study as we are focused on analysing
 419 relative changes. To facilitate data comparison, we adjusted the results from the second
 420 experiment to align with the first one by correcting the offset of fresh samples and applying
 421 the offset correction to all the dots obtained during the second experiment.

422 R450 decreases from the first irradiation dose until 5×10^{17} ions/cm², showing
 423 irradiation-induced exponential darkening (Figure 2A). S410-500 increases with irradiation
 424 showing a reddening until 1×10^{17} ions/cm² (Figure 2B). Unlike the exponential darkening,

425 the reddening shows no identifiable trend except a global increase with irradiation doses. It
426 is noticeable that the first irradiation dose at 1×10^{16} ions/cm² is slightly out of phase with
427 the following measurements. Several trends in the reddening can be observed. There is a
428 quick increase in reddening at the lowest doses until 3×10^{16} ions/cm², followed by a
429 transition with a slowdown in reddening until 6×10^{16} ions/cm². Finally, we notice a new
430 steep reddening increase going up to 1×10^{17} ions/cm². Unlike darkening, reddening does
431 not appear to reach a steady state at 1×10^{17} ions/cm². After an increase until 1×10^{17}
432 ions/cm², S410-500 seems to stabilise, showing no further identifiable trend with irradiation
433 up to 5×10^{17} ions/cm² (Figure 2B). The three samples reached a putative plateau and strong
434 irradiation doses even show a S410-500 close to the initial value of the fresh sample. The
435 irradiation modifies the concavity of the spectra between 0.41 and 0.5 μ m. This study shows
436 that the curvature (Figure 2C) increases with irradiation leading to a flattening of the
437 spectra. As for S410-500, the first irradiation dose is a bit out of phase. The curvature of the
438 komatiite increases from the fresh sample to 1×10^{17} ions/cm². However, for the basaltic
439 komatiite and the boninite, we notice an increase of the curvature until 6×10^{16} ions/cm²
440 and the beginning of stabilisation at higher doses (Figure 2C). Going up to 5×10^{17} ions/cm²,
441 the same type of putative plateau behaviour as for the reddening is observed for the
442 curvature even for the komatiite that had a different behaviour than the two other samples
443 (Figure 2C).

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473 Figure 2: Evolution of R_{450} , $S_{410-500}$, and the curvature upon ion irradiation up to 5×10^{17}
 474 ions/cm². Measurements were done on different pellets from the same material between the
 475 first experiment (up to 1×10^{17} ions/cm²) and the second one (up to 5×10^{17} ions/cm²).
 476 Pellets from the same material show different fresh values. This effect does not impact our
 477 study as we are focused on relative alteration more than absolute values. To facilitate data
 478 comparison, we adjusted the results from the second experiment to align with the first one
 479 by correcting the offset of fresh sample and applying the offset correction to all the dots. The
 480 error estimated on the reflectance is 1% in relative value for each sample. The error on the
 481 spectral slope includes the one on reflectances and the one related to the linear fit. If the
 482 error bar is not visible it indicates that it is smaller than the symbol. A) An exponential fit was
 483 used to fit the trend of the darkening ($y = a + b \cdot \exp(x \cdot c)$) with $a=0.32701211$,
 484 $b=0.03938215$, $c=-1.0026452$ for the komatiite; $a= 0.27503765$, $b=0.04220309$, $c=-$
 485 1.71623442 for the basaltic komatiite and $a=0.26849724$, $b=0.0107228$, $c=-5.77594749$ for
 486 the boninite. B) Evolution of the spectral slope. C) Evolution of the curvature.

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488 We propose to focus on the well-defined trends until 1×10^{17} ions/cm². If we normalise the
489 spectra of each dose by the fresh spectra and calculate R450 on the final spectra (Figure 3A),
490 we see that the intensity of the darkening depends on the sample. The reflectance of the
491 basaltic komatiite is more affected by ion irradiation than the komatiite, itself more altered
492 than the boninite. The decrease in reflectance is 3%, 7% and 11% respectively for boninite,
493 komatiite and basaltic komatiite at 1×10^{17} ions/cm² where a steady state starts to be
494 reached for the boninite. As for the reflectance, the reddening is more pronounced on the
495 basaltic komatiite than on the komatiite and on the boninite (Figure 3B). The flattening
496 follows a different trend and intensity according to the sample and is more pronounced on
497 the komatiite than on the boninite and on the basaltic komatiite (Figure 3C). It is noticeable
498 that the basaltic komatiite and the boninite have a similar evolution while the komatiite
499 shows its own trend with a stronger flattening (Figure 2C and 3C). As shown in Figure 1, it's
500 worth noting that the absorption band centred at approximately 1 μ m begins at a shorter
501 wavelength for komatiite compared to both basaltic komatiite and boninite. This
502 discrepancy can have an impact on the spectral characteristics and could explain the
503 komatiite trend isolated from the two others.

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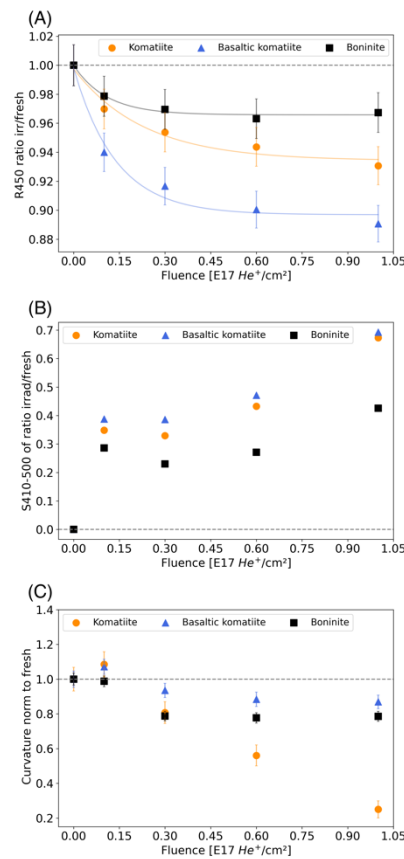
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546 *Figure 3: Evolution of R450, S410-500, and the curvature upon ion irradiation up to 5×10^{17}*
547 *ions/cm². R450 and S410-500 are calculated on the ratio of the irradiated and fresh spectra*
548 *while the curvature is here the ratio of curvature of the fresh spectrum and the irradiated*
549 *one. The error estimated on the reflectance is 1% in relative value for each sample. The error*
550 *on the spectral slope includes the one on reflectances and the one related to the linear fit. If*
551 *the error bar is not visible it indicates that it is smaller than the symbol. A) An exponential fit*
552 *was used to fit the trend of the darkening ($y = a + b \cdot \exp(x \cdot c)$) with $a=0.93416457$,*
553 *$b=0.06327671$, $c=-4.32889632$ for the komatiite; $a=0.89691885$, $b=0.10111709$, $c=-$*
554 *7.23221054 for the basaltic komatiite and $a=0.96578986$, $b=0.03405441$, $c=-9.28290193$ for*
555 *the boninite. The three fresh measurements are overplotted showing only the one of the*
556 *boninite in the three panels.*

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4.2. Effects of composition in the VNIR

559 The intensity and trends of spectral alterations after irradiation in the VNIR are
560 different according to the sample. The grain size being the same for all powders used in this

561 study and the collecting spot size being larger than the pellet heterogeneity, we attribute
562 the differences to the composition. There are correlations between R450 and S410-500
563 (Figure 4) and the SiO₂, TiO₂, FeO and MnO content. Concerning the curvature only one
564 correlation is observable with the MgO content. The darkening and reddening are stronger
565 for the basaltic komatiite than for the komatiite and less pronounced for the boninite. This
566 trend is correlated with a decrease of SiO₂ and TiO₂ (Figure 4A, B) and with an increase of
567 FeO and MnO (Figure 4C, D) from the basaltic komatiite to the boninite. The less SiO₂ and
568 TiO₂ in the sample, the less darkening and reddening. However, the less FeO and MnO in the
569 sample the greater the darkening and reddening. A similar trend is observed for the
570 evolution of R450 and S410-500 according to the SiO₂ and TiO₂ content. A different trend is
571 noticeable for the evolution of R450 and S410-500 according to the FeO and MnO content
572 (Figure 4). FeO and MnO seem to evolve progressively with irradiation. The evolution is
573 coherent and possibly linear. On the other hand, the link between SiO₂ and TiO₂ content is
574 less regular. In fact, we can see that two very close contents will have a very different effect
575 on irradiation, whereas two more distant contents will have a very similar reaction to
576 irradiation. This is even more marked for S410-500 than for reflectance. Concerning the
577 curvature, before 6×10^{16} ions/cm² there is no correlation between flattening and the
578 composition (Figure 5A). After a certain dose of irradiation, the flattening is more important
579 for the komatiite than the basaltic komatiite than the boninite. This can be correlated with
580 the MgO content (Figure 5B). The less MgO in the sample, the less flattening. Moreover, the
581 MgO content in the basaltic komatiite and the boninite are very close and their trends are
582 very similar. The flattening becomes almost constant after 6×10^{16} ions/cm² in comparison
583 with the komatiite for which the flattening increases linearly.

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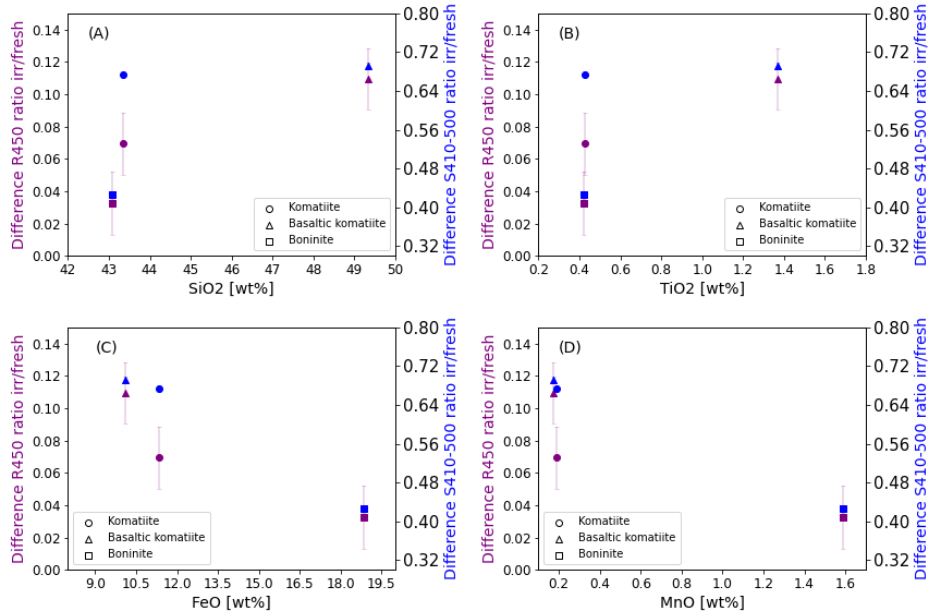
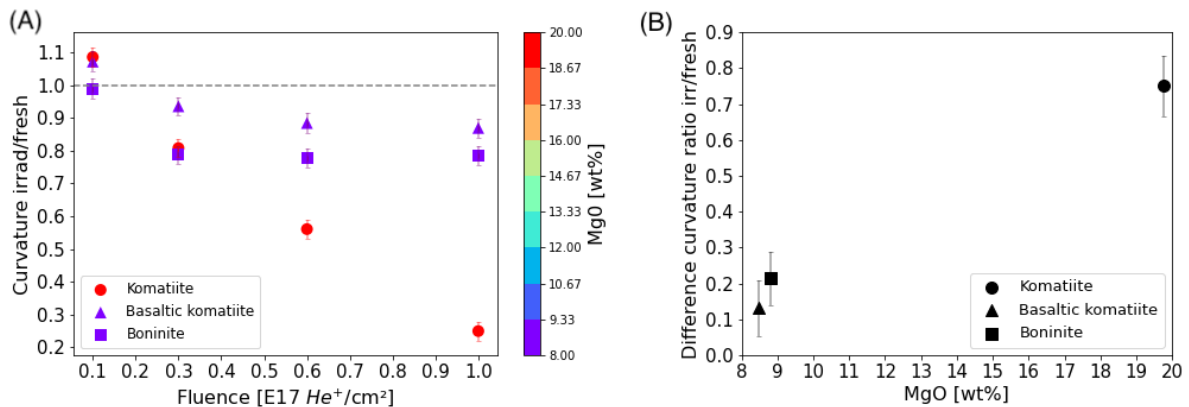


Figure 4: Evolution of R450 and S410-500 according to irradiation at 1×10^{17} ions/cm² and the composition. “Difference” refers to Figure 3A and B the difference between the fresh ratio and the 1×10^{17} ions/cm² ratio. Here are analysed only the differences between the fresh ratio and 1×10^{17} ions/cm² ratio. The maximal fluence at 5×10^{17} ions/cm² is not considered in order to study the evolution of spectral parameters before saturation. The intensity of the darkening and reddening is more pronounced for the basaltic komatiite followed by the komatiite and the boninite, correlating with the composition in SiO₂, TiO₂, FeO and MnO (Table 1).

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609 *Figure 5: Evolution of curvature according to the amount of MgO and irradiation A) Evolution*
610 *of the curvature upon ion irradiation until 1×10^{17} ions/cm² and the MgO content. B)*
611 *“Difference” refers to Figure 3C the difference between the fresh and irradiated 1×10^{17}*
612 *ions/cm² ratio measurement. The intensity of the flattening depends on the MgO content.*
613 *The flattening is increasingly marked between the basaltic komatiite and the komatiite. The*
614 *higher the MgO content, the greater the flattening. Here are analysed only the differences*
615 *between the fresh ratio and 1×10^{17} ions/cm² ratio. The maximal fluence at 5×10^{17} ions/cm²*
616 *is not considered in order to study the evolution of spectral parameters before saturation.*
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618 4.3. Mid-infrared

619 Although MIR range is dominated by silicate features, variations in composition
620 among the samples result in differences in their band shapes, widths and positions (Figure
621 6). We focus on the Christiansen feature (CF) around 8.5 μm and the Reststrahlen bands
622 (RBs) between 8.5 and 12 μm (Salisbury et al., 1991). The CF is an IR reflectance minimum
623 resulting from a rapid change in the refractive index shortward of the fundamental
624 molecular vibration bands. The RBs stem from fundamental molecular vibration bands due
625 to Si-O stretching and bending motions. We are interested in irradiation-induced alterations
626 of the main RB around 10 μm where there is a strong surface scattering, the CF and an
627 additional second RB between 10 and 12 μm . For the three samples, we observe a widening
628 of the RBs induced by irradiation as well as shifts of RBs and CF (Figure 6). We discuss first
629 the band identification before irradiation.

630 The boninite spectrum (Figure 6) is the less complex in shape because the composition is
631 only pyroxene and olivine. Additional minerals like plagioclase feldspar add features in the
632 spectra of the basaltic komatiite and the komatiite. The boninite shows a main RB at 9.6 μm
633 and a second one around 11.3 μm (Table 3). We attribute the main RB to the pyroxene
634 (enstatite), and the second RB to olivine (forsterite). Considering the basaltic komatiite
635 (Figure 6), the main RB around 10 μm shows the presence of pyroxene (here probably Mg-
636 rich pyroxene as diopside) and the second RB at 11.4 μm is related to olivine (Table 3).
637 Additional minor peaks are attributed to plagioclase. Finally, the komatiite has a very similar
638 composition to the basaltic komatiite but contains more olivine than pyroxene which could
639 explain the difference in intensity between the RB around 9.8 μm and the second RB just

640 before 11 μm compared to other samples (Figure 6). One has to note that the presence of
 641 volcanic glass can reduce the spectral contrast of bands. For example, absorption bands
 642 weaken and broaden as the glass content increases in feldspars (Nash and Salisbury, 1991).
 643 As we are working with scaled spectra, the presence of glass in basaltic komatiite and
 644 komatiite can only result in a broadening of the bands. In our data this is effectively what we
 645 observe with broader RBs for the basaltic komatiite (23-26 wt% of glass) than for the
 646 komatiite (12 wt% of glass), resulting in a less marked separation between the two RBs of
 647 the basaltic komatiite. Indeed, we evaluated the width of the main feature at 60% of its
 648 height and found values of $\sim 2.75 \mu\text{m}$ for the fresh basaltic komatiite (average of the two
 649 fresh samples) whereas it is $\sim 1.5 \mu\text{m}$ for the komatiite.
 650

Sample	Irradiation dose [ions/cm ²] and shift	Main RB $\sim 10 \mu\text{m}$ [μm]	CF [μm]	Second RB $\sim 11 \mu\text{m}$ [μm]
Boninite	Fresh	9.62 \pm 0.01	8.11 \pm 0.01	11.32 \pm 0.01
	1 x 10 ¹⁷	9.78 \pm 0.01	8.13 \pm 0.01	11.40 \pm 0.01
	Shift at 1 x 10 ¹⁷	0.16	0.012	0.07
	Fresh bis	9.66 \pm 0.01	8.20 \pm 0.01	11.40 \pm 0.01
	5 x 10 ¹⁷	9.77 \pm 0.01	8.06 \pm 0.01	11.42 \pm 0.01
	Shift at 5 x 10 ¹⁷	0.11	-0.14	0.025
Basaltic komatiite	Fresh	9.97 \pm 0.01	8.04 \pm 0.01	11.43 \pm 0.02
	1 x 10 ¹⁷	10.11 \pm 0.01	8.08 \pm 0.01	11.58 \pm 0.02
	Shift at 1 x 10 ¹⁷	0.14	0.040	0.15

	Fresh bis	9.97 ± 0.01	8.15 ± 0.01	11.42± 0.01
	5 x 10 ¹⁷	10.39 ± 0.01	8.11 ± 0.01	11.52± 0.01
	Shift at 5 x 10 ¹⁷	0.42	-0.040	0.10
Komatiite	Fresh	9.80± 0.01	8.20 ± 0.01	10.75± 0.02
	1 x 10 ¹⁷	9.99± 0.01	8.40 ± 0.01	10.78± 0.01
	Shift at 1 x 10 ¹⁷	0.19	0.20	0.03
	Fresh bis	9.78± 0.01	8.42 ± 0.01	10.74± 0.02
	5 x 10 ¹⁷	9.99± 0.01	8.24 ± 0.01	10.76± 0.01
	Shift at 5 x 10 ¹⁷	0.21	-0.18	0.02

651 *Table 3: Compilation of the main MIR features position before and after irradiation and the*
652 *associated shifts. Positions are evaluated as local maxima (RBs) or minima (CF). Fresh values*
653 *and fresh bis values correspond to the measurement before irradiation on the same powder*
654 *but two different pellets studied during irradiation experiments one and two respectively (see*
655 *3.2). Negative values indicate a shift towards shorter wavelengths (blue-shift) while positive*
656 *ones a shift towards longer wavelengths (red-shift).*

657
658 After irradiation, all samples show a shift of the main RB peak at ~ 10 µm towards higher
659 wavelengths (red-shift) with a maximum shift of 0.42 µm for the basaltic komatiite after an
660 irradiation of 1 x 10¹⁷ ions/cm² (see Figure 6 and Table 3 where all values discussed here are
661 compiled). Concerning the CF at ~ 8.5 µm, this work highlights two different behaviours
662 according to the irradiation dose (Figure 6 and Table 3). At 1 x 10¹⁷ ions/cm² the CF is red-
663 shifted while it is shifted toward shorter wavelengths (blue-shifted) at 5 x 10¹⁷ ions/cm². The
664 maximum shifts after irradiation happens for the komatiite with values of +0.20 µm at 1 x

665 10^{17} ions/cm and $-0.18 \mu\text{m}$ at 5×10^{17} ions/cm² both for the komatiite. We also investigate
666 the second RB between 10 and 12 μm (Figure 6 and Table 3). In general, the red-shift is less
667 pronounced than for the main Si-O peak, in agreement with what was observed by Brunetto
668 et al. 2014 (see this paper for a discussion on this phenomenon, beyond of the scope of the
669 current study), with a maximum shift of $0.15 \mu\text{m}$ for the basaltic komatiite after an
670 irradiation of 1×10^{17} ions/cm². We also emphasize here that the komatiite has a very
671 different spectral shape after both irradiation, with the secondary RB becoming the
672 maximum. We observe here a change in band intensity as already seen by Brunetto et al.
673 (2014 ; 2020) and Lantz et al. (2017, in press).

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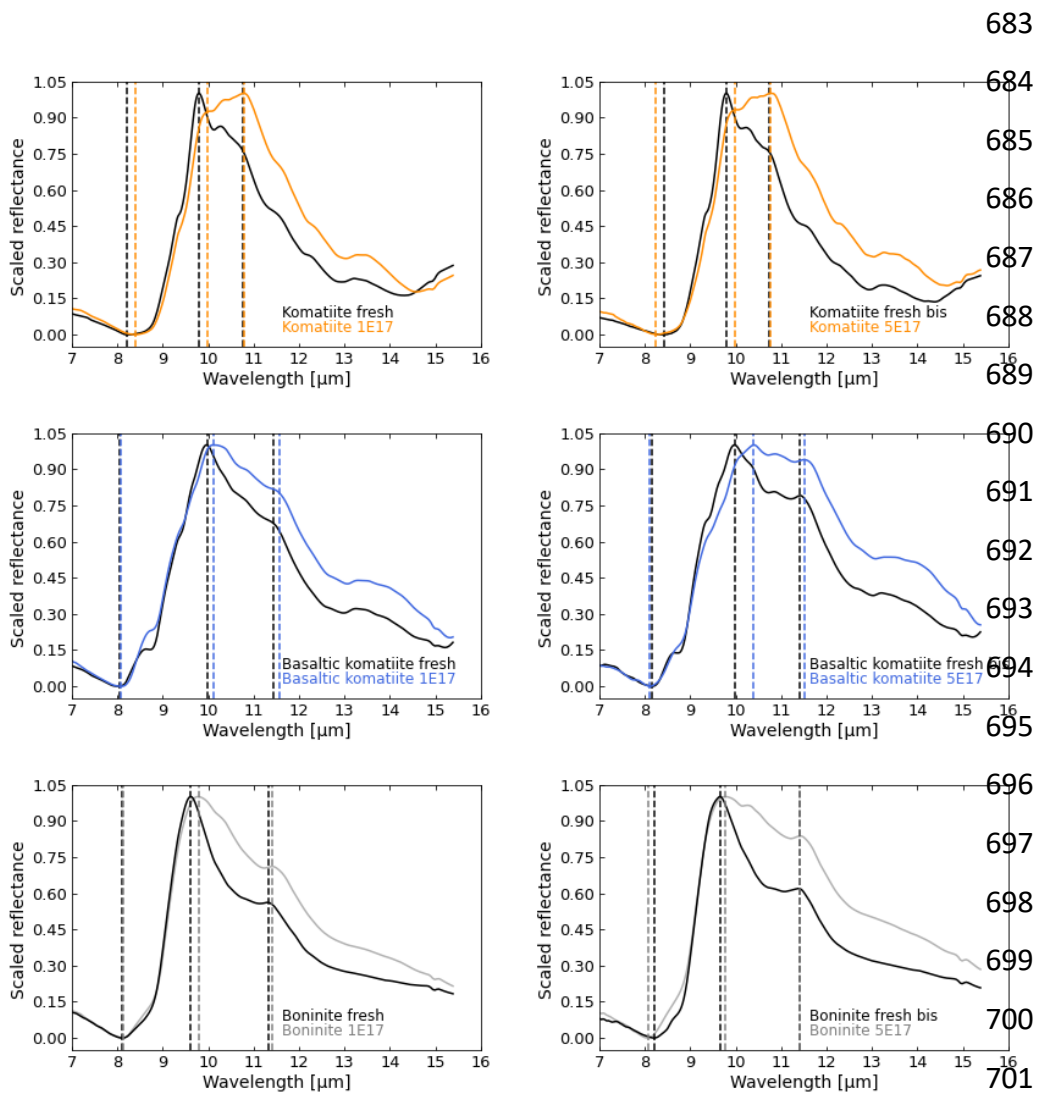
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703 *Figure 6: Mid-infrared reflectance spectra of the boninite, the basaltic komatiite and the*
 704 *komatiite. Spectra are scaled at the strongest peak. Irradiation doses are given in ions/cm².*
 705 *The left-hand column corresponds to the first experiment and the right-hand column*
 706 *corresponds to the second experiment (same notations as in Table 3).*

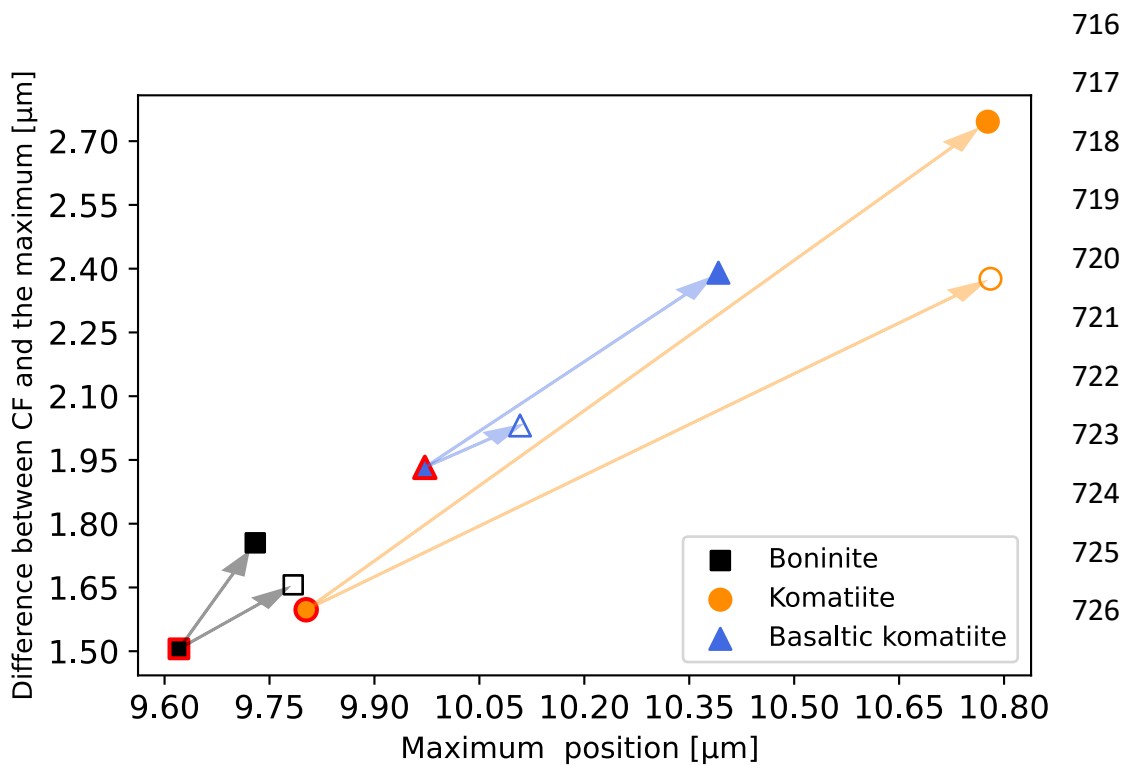
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708 We observed a broadening of the bands after irradiations for the three samples (Figure 6).
 709 For example, the width at 60% of the RB height for the komatiite is increased by $\sim 0.5 \mu\text{m}$
 710 between the fresh sample and 1×10^{17} ions/cm², and by $\sim 1 \mu\text{m}$ after 5×10^{17} ions/cm².

711

712 We decided to analyse our result in a two-component-size space (Figure 7)
 713 commonly used to sort measurements and have already proved their effectiveness in

714 classifying meteorites (Dionnet et al., 2022; Lantz et al., in press). The first thing to notice is
 715 that the fresh samples are well separated due to their different initial compositions.



727 *Figure 7: Evolution of the position of the maximum reflectance according to the difference*
 728 *between the CF and the position of the maximum reflectance. For each sample there are*
 729 *fresh samples in full symbols with red edges, linked to two other dots, the open being 1×10^{17}*
 730 *ions/cm² and the full one 5×10^{17} ions/cm². As measurements were done on the same*
 731 *powders but different pellets there is a difference between fresh measurements before*
 732 *1×10^{17} ions/cm² and 5×10^{17} ions/cm². This effect does not impact our study as we are focused*
 733 *on relative alterations more than absolute values. To facilitate data comparison, we adjusted*
 734 *the results from the second experiment (up to 5×10^{17} ions/cm²) to align with the first one (up*
 735 *to 1×10^{17} ions/cm²) by correcting the offset of fresh sample and applying this offset to the*
 736 *irradiated sample (same process as Figure 2).*

737
 738 All samples follow the same trend of maximum position redshift upon irradiation for both
 739 experiments. The shape of the komatiite spectrum is strongly modified upon irradiation
 740 (Figure 6), shifting the maximum of the spectrum towards the longest wavelengths at the
 741 level of the second RB. This intense modification of the spectrum is responsible for the
 742 longer vectors compared with the boninite and the basaltic komatiite. This difference

743 specific to komatiite could be explained by the composition or by the fact that one
744 component may have been more affected by irradiation, resulting in this change in shape.
745 The particular alteration of the komatiite could be due to olivine. The komatiite is the richest
746 sample in olivine, a mineral that is very sensitive to SpWe (Fu et al., 2012). While the
747 boninite and komatiite seem to have reached their maximum shift already at 1×10^{17}
748 ions/cm², the highest irradiation produces a bigger shift on the basaltic komatiite. It is likely
749 that these differences are linked to the composition of the samples and is in line with VNIR
750 results showing that the basaltic komatiite is more altered by the irradiation. Furthermore,
751 there is a bigger effect on the γ -parameter of this plot after the highest irradiation. Finally,
752 there seems to be a common trend among all samples after the highest irradiation defining a
753 "vector" of SpWe as all arrows become aligned. The steepness of that vector varies between
754 each rock: steeper for the komatiite than for the boninite and the basaltic komatiite. Thus,
755 the steepness of the vector follows the initial composition in MgO and these results could
756 help to constrain the MgO content of different rocks under the same ion irradiation. We
757 highlight here that the distance between the CF and the maximum is a good indicator of
758 SpWe (as proposed by Lantz et al., in press). Together with the maximum position they
759 define a SpWe vector whose inclination helps to relatively compare the irradiation level.
760 When MERTIS data will be available this kind of plot will help in understanding the effect of
761 the environment on remote sensing data, both for composition and SpWe effects.

762 5. Discussion

763 5.1. Global darkening and reddening

764 The reflectance and slope in the VIS constitute reliable spectral proxies of irradiation
765 on volcanic surfaces. Even if the carbon content and grain size have a significant effect on
766 spectral properties (Bruschini et al., 2022), SpWe is suggested to be the major contributor to
767 albedo variations on Mercury (Riner and Lucey, 2012). Riner and Lucey (2012) applied a
768 space weathering model to multispectral images of Mercury. They have shown that albedo
769 variations on Mercury cannot be explained by composition or grain size alone but can be
770 explained by the presence of SMFe (influenced by both exposure time to SpWe and
771 composition).

772 The present study confirms the well-known effects, darkening and reddening, of ion
773 irradiation on silicates (Brunetto and Strazulla, 2005; Fu et al., 2012; Loeffler et al., 2009;

774 Chrbolková et al., 2021). These spectral alterations on silicates have been linked to the
775 formation of SMFe (Hapke, 2001; Carrez et al., 2002; Noble et al., 2007; Loeffler et al., 2009;
776 Lucey and Riner, 2011; Pieters and Noble, 2016; Penttilä et al., 2020) due to solar wind
777 implantation. In comparison to the Moon and according to the low amount of available iron
778 on Mercury it is likely that, if there was more iron available in the past, a major part of it has
779 been processed into SMFe by SpWe. Even low concentrations of SMFe (0.02 wt% Noble et
780 al., 2007) can influence surface reflectance properties (Noble et al., 2001, 2007; Loeffler et
781 al., 2009). On Mercury, Lucey and Rinner (2011) have shown that reduced metallic iron could
782 be around 3.5 wt%, more than enough to influence the spectral properties of the surface.
783 This phenomenon is usually accompanied by an amorphization of the target depending on
784 the fluence (Demyk et al., 2001; Carrez et al., 2002; Brucato et al., 2004; Brunetto et
785 Strazulla, 2005). Ion irradiation induces gas implantation which leads to vesiculation (Rubino
786 et al., 2024). The darkening in the visible spectral range could then also be linked to the
787 increase in porosity of the sample surface due to ion implantation (Sultana et al., 2021).

788 Reddening and darkening process in the VNIR does not remarkably affect the mafic
789 silicate absorption feature (Brunetto et al., 2006, Loeffler et al., 2009). We confirm here that
790 there is no significant shift of band position (Brunetto et Strazulla, 2005; Marchi et al., 2005)
791 showing that variations due to chemical compositions dominate ion irradiation spectral
792 alterations (Fu et al., 2012). Mercury spectral units show an evolution from dark material
793 with a shallow slope to bright material with a steeper slope (Robinson et al., 2008; Denevi et
794 al., 2009; Ernst et al., 2010; Murchie et al., 2015; Barraud et al., 2020). Our results confirm
795 experimentally the darkening induced by SpWe through time. In addition, our study is in
796 agreement with observations of Mercury, where the reddening that accompanies darkening
797 is absent after a certain exposure time. We propose two hypotheses to explain this
798 darkening with no reddening:

799 1) The formation of Britt–Pieters [>50 nm SMFe (Lucey and Noble, 2008)] particles
800 causes darkening with no reddening of the surface. Spectral effects of SMFe are different
801 according to the concentration and particle sizes (Noble et al., 2007; Penttilä et al., 2020).
802 Different types of SMFe have been identified including “nanophase particles” that both
803 darken and redden the surface and larger “Britt–Pieters particles” that only darken (Noble et
804 al., 2007; Lucey and Riner, 2011; Trang et al., 2017; Penttilä et al., 2020). On Mercury, we

805 expect larger SMFe than on the Moon (Noble and Pieters, 2003). High-velocity impacts on
806 Mercury produce more melt and vapour, moreover, we expect more agglutinates leading to
807 a growth of metallic iron particles (Lucey and Riner, 2011). The high tempera- ture on
808 Mercury's surface will help the Ostwald ripening process (Noble and Pieters, 2003; Deutsch
809 et al., 2024), a mechanism that participates in the coalescence and growth of SMFe. On the
810 3.5 wt% of SMFe predicted by Lucey and Riner (2011), 3 wt% constitute Britt–Pieters
811 particles. They are at the origin of a darkening with no reddening of the surface.

812 2) It is possible that the reddening of a heavily space-weathered volcanic surface
813 becomes solar wind flux independent with time. We show that after a certain dose of
814 irradiation, the reddening is no longer observable with values that can be close to the value
815 of the fresh sample. On Mercury, after billions of years of irradiation, it is possible that the
816 slope becomes independent from irradiation.

817

818 5.2. Relation between near-UV features and SpWe

819 This work highlights the flattening of spectra in the near-UV due to ion irradiation.
820 We discussed the link between the Fe content and the spectral property in the visible (see
821 2.1 and 5.1) but Fe can also modify spectra in the near-UV. MESSENGER reflectance spectra
822 exhibit a downturn from the VIS to the near-UV often used to characterise the surface
823 through several spectral parameters (Goudge et al., 2014; Barraud et al., 2020). This feature
824 in the near-UV can be attributed to Oxygen-Metal Charge Transfer (OMCT) (Bruns et al.,
825 1993; McClintock et al., 2008) and is mainly associated with younger surfaces (Izenberg et
826 al., 2014). Indeed, matured material shows a globally lower albedo and a less curved
827 continuum (McClintock et al., 2008; Robinson et al., 2008; Blewett et al., 2009; Domingue et
828 al., 2014; Murchie et al., 2015; Pieters and Noble, 2016). This preponderance is attributed to
829 the transformation of FeO to SMFe that is more advanced in mature regions (Noble et al.,
830 2007; Lucey and Riner, 2011; Riner and Lucey, 2012; Pieters and Noble, 2016).

831 Barraud et al. (2020; 2023) support the idea that sulphides are at the origin of the
832 near-UV to VIS curvature observed in hollows. However, the samples used in this work do
833 not contain any sulphide showing that in addition to the composition, the curvature is also
834 influenced by SpWe. This observation emphasises that the shape of the spectra in the near-
835 UV to VIS can bring information on the maturity of surfaces. The curvature can be used with

836 confidence only for fresh surfaces as the tendency is unclear after 1×10^{17} ions/cm² with the
837 energy used in this study. Hollows, younger than ~100 thousand years (Wang et al., 2020),
838 are good candidates to explore the variability of the curvature on Mercury according to ion
839 irradiation. We experimentally validate that the near-UV features are, to a certain extent,
840 controlled by solar wind ions irradiation and reflect the soil maturity for the younger
841 surfaces having the same composition which makes it a reliable spectral proxy of irradiation.

842

843 5.3. Influence of the composition on spectral properties

844 We confirmed in this work that under the same conditions, different compositions in
845 silicates show a different sensitivity to SpWe even if they all show a global darkening and
846 reddening until 1×10^{17} ions/cm² (Fu et al., 2012; Chrbolková et al., 2021). The present results
847 imply a putative diversity of SpWe on Mercury according to the geochemical terranes.

848 By comparing olivine, plagioclase and basaltic glass Fu et al. (2012) have shown that
849 olivine is the most sensitive to darkening followed by plagioclase and then basaltic glass.
850 Moreover, olivine is more impacted by reddening than pyroxene (Marchi et al., 2005;
851 Strazzulla et al., 2005; Brunetto et al., 2006, Chrbolková et al., 2021). The curvature follows
852 this trend with a curvature alteration intensity decreasing from the komatiite to the basaltic
853 komatiite where the content in olivine decreases. The curvature is also led by Mg content
854 and a putative preferential sputtering (see 5.4). In fact, the boninite and basaltic komatiite
855 have a very similar content in Mg while the komatiite contains more than twice the amount
856 of Mg. The intensity of the alteration of the curvature after irradiation is very similar for the
857 basaltic komatiite in comparison with the komatiite. However, the darkening and reddening
858 do not follow this tendency concerning olivine. The boninite is dominated by pyroxene (66.8
859 wt%) (Mari et al., 2023) which could explain the weaker intensity of darkening and
860 reddening in comparison with the basaltic komatiite and the komatiite that are more
861 balanced (Chrbolková et al., 2021).

862 Variations in reflectance and spectral slope on Mercury do not appear to be directly
863 linked to variations in Fe content. Fe concentration can only play a minor role in surface
864 variation across Mercury (Weider et al., 2014). Nevertheless, by analysing individual
865 mechanisms (darkening mineral content, SpWe, effect of grain size, etc.) Fe content seems
866 to be a key parameter to be taken into account to clarify spectral variation on Mercury.
867 Bruschini et al. (2022) have shown that the amount of iron in silicates influences the

868 alterations of the spectral slope to graphite content. Silicates containing lower iron content
869 are characterised by a greater alteration in spectral slope versus graphite content. Our study
870 confirms the strong influence of iron content on spectral alterations. The less iron the
871 sample contains, the more sensitive it is to darkening and reddening induced by irradiation.
872 Moreover, Maturilli et al. (2014) have shown that a synthetic analogue with less iron was the
873 most strongly altered after thermal processing, highlighting the importance of iron in
874 spectral alteration induced by the extreme environment. These observations are crucial for
875 the surface of Mercury. As we expect a very low iron content, the effect of darkening and
876 reddening induced by solar wind may be stronger than what is expected (Bruschini et al.,
877 2022).

878 On Mercury, geochemical units will react differently to SpWe. The Caloris basin (CB
879 column in Table 1) has a high SiO₂ content and a low FeO content in comparison with other
880 units making it one of the most sensitive units to SpWe. The Northern Plains are
881 compositionally heterogeneous (Table 1) and will therefore have a non-uniform SpWe effect
882 that will intensify spectral differences. It is likely that the HMR will be more sensitive to
883 curvature alteration than other units making the flattening more important. Moreover, the
884 HRM, as the NP, shows different sub-area according to the composition that will increase
885 spectral heterogeneities in the region. Mn and Ti are minor elements and it is unlikely that
886 they have a major influence.

887

888 5.4. Mid-infrared

889 Based on laboratory evidence of MIR spectral alteration upon irradiation, we
890 explored the possibility of detecting the irradiation effect through the future remote sensing
891 observation of MERTIS, using band shifts as proxies. Indeed, shifts of the main RB and a part
892 of the shifts of the second RB and CF are higher than the MERTIS spectral resolution of 90
893 nm. As comparisons with data from Mercury's surface are currently limited in this
894 wavelength range, MERTIS data are expected to be used for comparison with this study.

895 In the MIR, we observe a red-shift of the RBs positions induced by He⁺ irradiation.
896 Amorphization and Mg loss have been proposed to explain the red-shift of Si-O stretching
897 around 10 μm. The volatile Mg could be easily sputtered leading to a loss of magnesium
898 after irradiation (Brunetto et al., 2014; Lantz et al., 2015; 2017) possibly leading to
899 amorphization (Brucato et al., 2004; Demyk et al., 2001; Brunetto et al., 2014). Irradiation

900 experiments on olivine demonstrated chemical changes including a decrease in Mg/Si ratio
901 supporting this theory (Demyk et al., 2001; Carrez et al., 2002). Some similar shifts have
902 been found with no relation to the elemental composition. Dukes et al. (2015) did not
903 observe a reduction in Mg content after ion irradiation of the Tagish Lake meteorite.
904 Moreover, Rubino et al. (2024) have shown that Mg depletion is not systematic in irradiated
905 samples. So, changes in composition alone do not with certainty explain the observed MIR
906 shifts. The shifts may be caused by the amorphization of the silicates (Brucato et al., 2004;
907 Vernazza et al., 2013). Disorder of the mineral crystal lattice could lead to Si-O stretching
908 feature alteration. Ion implantation appears to preferentially sputter Mg during the
909 amorphization process. However, the sputtered Mg does not necessarily escape from the
910 sample but could remain trapped inside the bubbles produced by vesiculation during
911 amorphization of the sample (Rubino et al., 2024). Thus, the amorphization is not necessarily
912 accompanied by a change in chemical composition (Jäger et al., 2003). Taking into account
913 the composition of our samples and fluences used in this work, it is likely that ion irradiation
914 induced an amorphization and preferential sputtering of Mg, with or without chemical
915 alteration, at the origin of the MIR red-shifts (Demyk et al., 2001; Carrez et al., 2002; Brucato
916 et al., 2004; Brunetto et al., 2014, 2020). We notice that both the main and second RB shifts
917 between fresh and 1×10^{17} ions/cm² samples are correlated with the MgO content in the
918 fresh sample (Figure A1: A, F). However, for an irradiation of 5×10^{17} ions/cm² the RBs at ~ 10
919 μm shifts are correlated with SiO₂, TiO₂, FeO and MnO while the minor RBs still follow the
920 MgO content (Figure A1: B, C, D, E, G). The correlation between the SiO₂ content and the
921 strongest RB positions is discussed in the case of Mercury analogues in Morlok et al. (2017,
922 2019, 2021). Here we confirm a correlation between the RB at $\sim 10 \mu\text{m}$ position and SiO₂. In
923 our case, we observe that higher SiO₂ content is correlated with higher wavelengths of main
924 RB positions. SpWe does not modify this trend.

925 We observe that in general, the shift is stronger for the intense peak around $10 \mu\text{m}$ than for
926 the less pronounced feature between 10 and $12 \mu\text{m}$ showing that the SpWe effect is
927 wavelength dependent. The more intense the band is, the higher the red-shift (Brunetto et
928 al., 2014; Lantz et al., 2017). The strongest absorption bands probe a thinner depth in the
929 sample than low absorption bands and thus are more sensitive to the irradiated surface
930 layer. Photons emerging from the smaller bands come from a deeper layer not significantly
931 modified by irradiation leading to weaker alterations. These shifts do not conflict with the

932 putative SMFe formation (Hapke, 2001) and show that different bands provide different
933 proxies of irradiation processes by probing different surface layers.

934 The Christiansen feature depends on the mineralogical composition. Thus, the CF
935 position is expected to be linked to the RB displacement (Brunetto et al., 2020). Our study
936 confirms this trend until 1×10^{17} ions/cm². However, the CF alterations are not unique and
937 show a different behaviour with a blue-shift after 5×10^{17} ions/cm². An idea could be that
938 amorphization due to irradiation modifies the CF behaviour. Chrbolková et al. (2021) showed
939 that after irradiation, the olivine CF is red-shifted while the pyroxene CF is blue shifted. We
940 suggest that the olivine being more sensitive to space weathering it will lead spectral
941 alteration until 1×10^{17} ions/cm² showing a red-shift while after an intense alteration,
942 pyroxene will dominate showing a blue-shift at 5×10^{17} ions/cm². In fresh samples, until $5 \times$
943 10^{17} ions/cm² we do not see any correlation between CF and SiO₂ content (Morlok et al.,
944 2017; 2019; 2021). We notice a correlation between the CF shift and CaO, K₂O and Al₂O₃
945 content until 1×10^{17} ions/cm² (Figure A1: H, I, J) though the trend changes after 5×10^{17}
946 ions/cm² showing a correlation with MgO (Figure A1: K).

947 The widening of bands is possibly linked with amorphization although chemical
948 alterations are not negligible. The deformation of the bands after irradiation comes from the
949 variation of the relative intensity of the peaks according to the composition. In addition,
950 alterations of the area and wideness of the band can be related to the disordering of the
951 mineral crystal lattice and alterations in composition.

952

953 6. Conclusion

954 Mercury's surface has been space-weathered over time, complicating the
955 interpretation of data derived from remote sensing observations. We presented the first
956 VMIR reflectance spectra of three terrestrial analogues: a boninite, a basaltic komatiite and a
957 komatiite after simulating solar wind irradiation under Mercury's conditions. We used 20
958 KeV He⁺ with fluences up to 5×10^{17} ions/cm².

959

960 - Ion irradiation leads to an exponential darkening of the spectra in the VNIR. The
961 darkening is confirmed by looking at spectral units defined on Mercury with older
962 units being darker than the younger ones.

963

964 - Ion irradiation leads to a reddening and flattening of the spectra in the near-UV to
965 the visible. After an irradiation of 1×10^{17} ions/cm² under the conditions of this
966 study, the reddening and flattening stop increasing showing ambiguous trends.
967 Looking at spectral units defined on Mercury, the reddening does not necessarily
968 increase with time. Submicroscopic Britt–Pieters metallic-iron particles could explain
969 this darkening with no reddening. The differences in curvature between spectral
970 units on Mercury are reliable proxies for the alteration of the youngest units.

971

972 - The intensity of spectral alterations induced by ion irradiation depends on the
973 composition of the surface. SpWe as well as composition participate in the spectral
974 heterogeneities of the surface of Mercury. We suggest that the FeO, SiO₂, MgO and
975 of less importance TiO and MnO contents can influence the intensity of alterations
976 induced by solar wind in the VMIR. As well as Al₂O₃, CaO and of less importance K₂O
977 in the MIR. Mercury's low iron content certainly increases spectral alterations
978 induced by solar wind.

979

980 - In the MIR, we detected a consistent red-shift of the RBs induced by ion irradiation.
981 The higher the peak, the greater the red-shift after irradiation. However, shifts of the
982 CF depend on the irradiation dose. RBs and CF shifts represent reliable proxies to
983 detect irradiation effects through future MERTIS observations.

984

985 Even if ion irradiation may not be the main process of reducing spectral band signatures on
986 Mercury, the BepiColombo observations are eagerly awaited to provide a better
987 understanding of the absence/weakness of absorption bands in the VNIR. If the rocks on
988 Mercury's surface are comparable to our current terrestrial analogues, it is conceivable that
989 BepiColombo will detect spectral features comparable to those in this study. Waiting for
990 BepiColombo, our work in the VNIR can be used to better understand MESSENGER data. The
991 terrestrial alteration detected in these samples is not expected on Mercury's surface.
992 Moreover, it is important to note that this study simulates only partly the SpWe effects on
993 Mercury's surface and additional work should be done to understand the cumulative effects
994 of processes happening in this extreme environment.

995

996 Acknowledgments

997 The authors acknowledge the Centre National d'Etudes Spatiales (CNES) for
998 continuous and long-term support. We also acknowledge the Domaine d'Intérêt Majeur en
999 Astrophysique et Conditions d'Apparition de la Vie+ (DIM ACAV+), île de France, for their
1000 support. This research is part of a joint IAS-CSNSM project (INGMAR) and we warmly thank
1001 O.Mivumbi and engineers for their help on INGMAR. This work is part of a LESIA-IAS
1002 collaboration and the preparation of the ESA-JAXA BepiColombo mission. Emma Caminiti
1003 warmly thanks Aleksandra Stojic for reading this work and providing helpful advice. The
1004 authors acknowledge Noah Jäggi and the anonymous referee for their helpful comments at
1005 the review stage.

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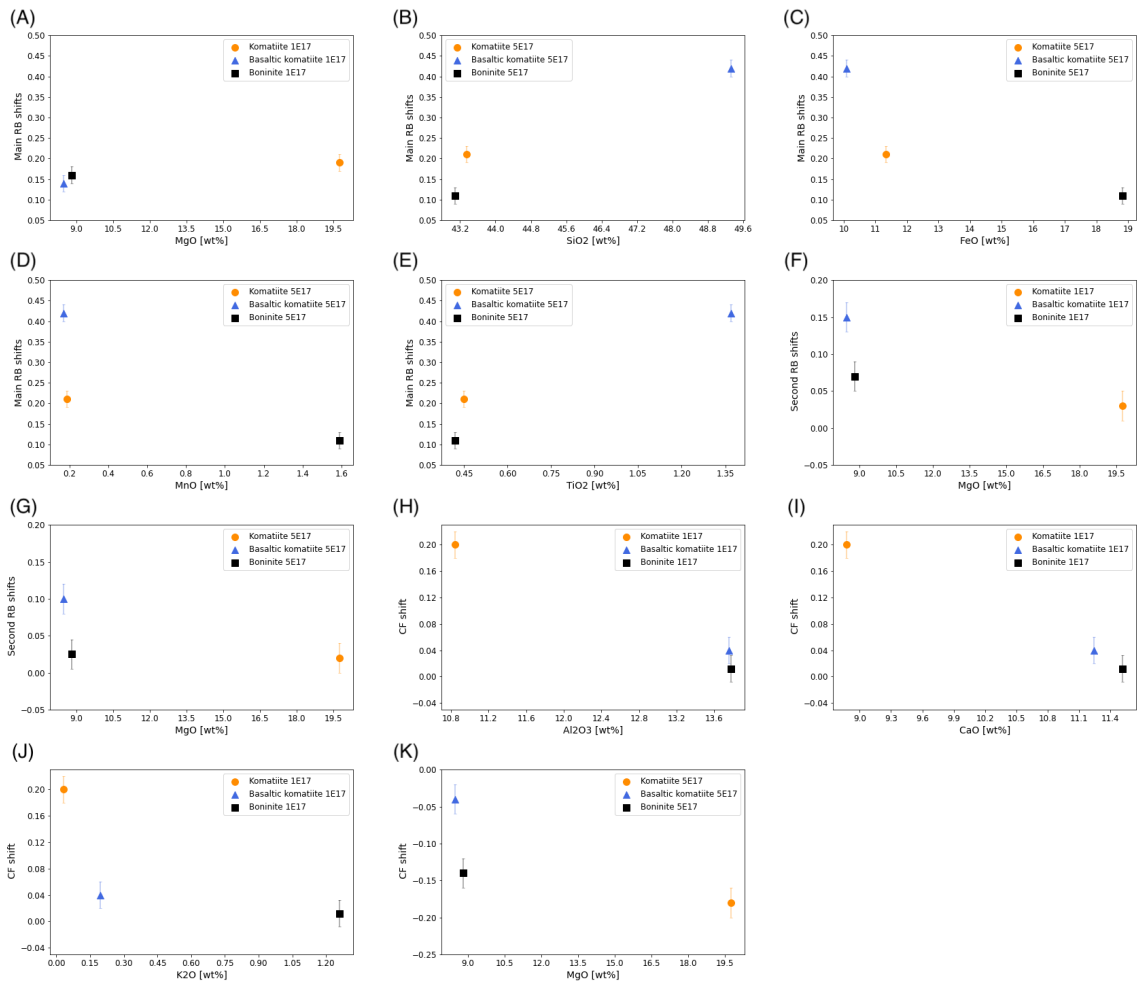
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1015 Appendix A: Supplementary figures to support MIR discussion

1016 We present here 11 panels showing the link between the composition and the alteration of
1017 MIR features. This figure is a visual representation of correlations between Table 1 and Table
1018 3. Only the correlations mentioned in the text are shown. MIR features showing no
1019 correlation with the composition are not represented.

1020



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Figure A1: Alteration of MIR features according to the composition.

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