

Multiscale physical-chemical analysis of the impact of fracture networks on weathering: Application to Nickel redistribution in the formation of Ni-laterite ores, New Caledonia

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1	Multiscale physical-chemical analysis of the impact of fracture
2	networks on weathering: Application to Nickel redistribution in the
3	formation of Ni-laterite ores, New Caledonia
4	
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15 Abstract

16 Supergene processes related to downward water migration and dissolution of primary minerals in the 17 New Caledonian peridotites result in Ni enrichment at the bottom of the lateritic profile near bedrock. 18 However, the combined effects of relief, lateral fluid flow, and complex inherited discontinuity 19 networks filled by serpentine generate a more complex Ni distribution than a simple flat horizon. In 20 surficial environments, fluid flow and mass transfer are not limited to the vertical scale; they can also 21 be related to lateral fluid circulation and local drainage along damage zones in the vicinity of faults. In 22 this study, we present new structural, petrophysical and geochemical data for core samples to shed 23 light on the impact of a fracture network on the alteration and progression of the Ni-enrichment front 24 during weathering. The analysis of element redistribution by XRF mapping coupled with fracture 25 extraction from computed tomography (CT) establishes that the serpentine fracture network 26 comprises sets of sub-orthogonal fractures whose orientations are potentially determined by pre-27 existing magmatic foliation. The progression of the olivine dissolution front leads to the redistribution 28 and concentration of Ni into the serpentine fracture network as secondary Mg-Ni talc-like phases. The 29 increase in porosity from <1% in the bedrock to >10% in the saprock is mainly driven by the formation 30 of nanopores. Nanoporosity may control the development of the weathering front and the 31 redistribution of Ni by small-scale diffusion. Structural and geochemical characteristics identified at 32 the sample scale are analogous to those observed on the larger scale of the outcrop and deposit. Scale 33 invariance in the structural organization and geochemical evolution indicates the potential upscaling 34 of the processes involved in the redistribution of Ni.

35

36 Keywords

37 Fractures, Multiscale, Weathering, Nickel

38 I. Introduction

39 Ni-laterite deposits in New Caledonia represent 7 % of the global mine production in 2021 (USGS, 40 2022). Laterite is a surficial formation developed after the deep weathering of rocks under a hot, wet 41 tropical climate. The rainwater leaches most of the soluble elements, which leads to the residual 42 concentration of more insoluble elements such as Al, Fe or Ni (Best, 2015). In the New Caledonian 43 mining district, laterites originated from the weathering of ultramafic rocks consisting of relics of the 44 obducted Peridotite Nappe during the Eocene (Chevillotte, 2005; Gautier et al., 2016; Sevin et al., 2014; 45 Trescases, 1975). The downward migration of meteoric waters leached most elements from the 46 peridotites (Mg, Ni, Si) except iron. Waters are saturated at depth with respect to newly-formed Si-47 Mg-Ni minerals in the saprolite (Butt and Cluzel, 2013; Freyssinet et al., 2005; Golightly, 2010; Haldar, 48 2017). During this process, Ni is trapped in Fe-(oxy)-hydroxides in the oxide zone and phyllosilicates in 49 the saprolite ore (Dublet et al., 2012; Fritsch et al., 2016; Santoro et al., 2022; Villanova-de-Benavent 50 et al., 2014). Ni is redistributed towards the bottom of the laterite profile during laterite formation due 51 to the goethite dissolution-recrystallisation process, so-called the goethite ageing (Dublet et al., 2015). 52 The tectonic uplift enhanced the downward redistribution process from the oxide zone toward the 53 saprolite ore. Indeed, rejuvenating the topography and lowering the water tables maintained the 54 drainage conditions required to redistribute elements and differentiate the laterite profile (Maurizot 55 et al., 2020). Therefore, the competition between tectonic uplift and weathering rate controls the dominant ore type in Ni lateritic deposits. Oxide ore formation is favored in poorly uplifted areas 56 57 dominated by weathering, whereas saprolite ore formation occurs in more uplifted areas (Butt and Cluzel, 2013; Maurizot et al., 2020). However, a pure "per descensum" model fails to fully explain the 58 59 Ni heterogeneities observed in the saprolite (Quesnel et al., 2017), where several fissure infillings with 60 distinct habitus and mineral-chemistry are found (Cathelineau et al., 2016, 2017). The Ni concentration 61 in the saprolite ore is generally around 2 wt% and locally up to 5 wt.% (Butt and Cluzel, 2013; Tauler et al., 2017). It results from the contribution of several Mg-Ni phyllosilicates (predominantly talc-like) 62 63 forming the so-called "garnierite ore", whose Ni contents vary from a few % up to 30 wt.% Ni

64 (Cathelineau et al., 2017). This complexity results from a multiphase redistribution model based on the 65 pre-concentration of Ni in fractures. The garnierite ore is subsequently affected by the downward migration of a dissolution front guided by fracture networks at the outcrop scale (Cathelineau et al., 66 2017). Fu et al. (2018) suggested that the origin of the "garnierite ore" is closely linked to the 67 68 preferential flow regime in conduits and is often associated with dissolution pipes. Iseppi et al. (2018) 69 pointed out the structural control at the deposit- to massif- scale on the Ni remobilization. Inherited 70 fractures control erosion at a massif scale and act as drains and permeability walls due to the 71 impermeable serpentine coating at the deposit scale. Myagkiy et al. (2019) highlighted the effect of 72 topographic slope and occurrence of fractures on Ni redistribution through the 2D modelling of 73 reactive transport.

74 More generally, whatever the lithology, fractures may be organized as networks following fractal 75 organization, meaning that fracture networks show the same spatial distribution at different scales 76 (Basirat et al., 2019; Roy et al., 2007). Consequently, the structural organization observed at outcrop 77 and larger scales should also be found at a smaller scale. In New Caledonia, fracture networks play an 78 essential role in the Ni redistribution at the saprolitic level at the outcrop scale. There, the potential 79 fractal organization of the fracture network would imply that Ni redistribution processes may present 80 similarities at different scales. Furthermore, scaling laws have been widely applied to describe fracture 81 network statistical properties at different scales (Afshari Moein et al., 2019; Bonnet et al., 2001; Bour, 82 2002; Masihi and King, 2008). The impact of scaling laws on fluid flow or seismic hazard has been widely explored (Allègre et al., 1982; de Dreuzy et al., 2012; Hatton et al., 1993), but the applicability to ore 83 84 deposits formation remains to be addressed. The upscaling of Ni mineralization processes from the 85 core to the field scale would be of prime interest for mine exploration. Although structural data are 86 generally available at the regional and deposit scale (Jeanpert et al., 2019; Quesnel et al., 2017), 87 microstructural, petrophysical or geochemical characterization at the sample scale is often lacking.

This paper investigates the impact of fracture networks at the sample scale on Ni redistribution when parent rocks are affected by the progression of the silicate dissolution front. Representative core 90 samples of the dissolution front were collected from the Cagou pit (Koniambo Massif). Structural, 91 petrophysical and geochemical features identified at the sample scale are compared to those 92 documented at the scale of outcrops and mine workings, allowing discussion on the potential upscaling 93 of the genetic processes for a correct understanding of the factors controlling the ore geometry.

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95 Geological settings

96 The "Grande Terre" island of the New Caledonian archipelago, located in SW Pacific (Figure 1.a), 97 consists of ca. 30% of ultramafic rocks, which are relics of the obducted Peridotite Nappe on the 98 continental Norfolk Ridge during the Eocene period (Cluzel et al. 2001). The Peridotite Nappe is 99 nowadays observed in the "Massif du Sud" and in klippes mainly along the West Coast, including the 100 Koniambo Massif. Main fracture directions in the ultramafic rocks strike N130°-N150°, parallel to the 101 main elongation direction of the island, which is commonly thought to be the obduction azimuth. 102 However, several other sets of fractures striking N0°, N45°, and N90° at the island scale are related to 103 later deformation stages, including the Miocene extensional tectonics (Lagabrielle et al., 2005).

104 After the obduction (late Oligocene), the peridotite is affected by deep weathering in a hot and humid 105 climate, ending with a thick lateritic profile (Sevin et al., 2012). In the laterite profile, several domains 106 can be outlined, which from base to top are (i) a saprolite horizon resulting from the progressive 107 dissolution of the variably serpentinized peridotite bedrock mainly consisting of silicates (olivine), 108 allowing the precipitation of phyllosilicates and goethite (detailed explanation of the chemical 109 reactions are given in Butt and Cluzel (2013) and Myagkiy et al. (2017)), and of (ii) an oxide ore which is the final weathering stage, resulting from the dissolution and leaching of all minerals apart from 110 111 immobile goethite (Figure 1.c).

The Koniambo Massif (Figure 1.b) is defined by a highly marked relief and poly-directional fracture
network mainly oriented WNW-ESE, NS, NW-SE and NE-SW (Jeanpert et al., 2019; Quesnel et al., 2016).
The regolith observed in the massif consists of hectometer-sized patches of laterite profiles of varying

thicknesses ranging from a few meters to > 60m (Quesnel et al., 2017). Ni-rich saprolites are mainly
located on slopes where the oxide ore cover is thin or absent. In contrast, Ni-poor saprolites tend to
be situated in topographic highs under thick oxide ore cover (Quesnel et al., 2017). The Cagou open
pit, previously called Pit 207 in previous related studies describing the rock mineralogy (Cathelineau et
al., 2017, 2016; Muñoz et al., 2019), presents locally preserved oxide ore cover ranging from a few
meters to 20 m in thickness, and an intense fracture network in the rocky saprolite - bedrock contact
zone (Quesnel et al., 2016).





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124 Figure 1 – a) Geological map of New Caledonia. b) Geological setting of Koniambo Massif. C) Typical peridotite-derived

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lateritic profile (Quesnel et al. (2017)

126 II. Materials and Methods

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128 To study the impact of fractures on the weathering front progress at different scales, structural, 129 petrophysical and geochemical data were collected on a 30 cm-long core sample (Figure 2.a). The 30 130 cm-long core sample was collected at a depth between 6 m and 6.3 m of a vertical 41 m deep drill hole 131 in the Cagou Pit (latitude: 20°58'25.163" S, longitude: 164°47'29.683" E, altitude: 755m) (Figure 1.b). 132 The core sample shows a visible fracture network and an increase in weathering intensity, from the 133 fresh peridotite at the bottom to the altered peridotite at the top, with a typical brownish colouring 134 due to goethite. A 32 mm x 73 mm focus on the upper part of the core sample (named S_0) was used to describe the analyzed processes at the cm-scale. Various 20 mm x20 mm subsamples from the core, 135 136 respectively called S_1 to S_7 (Figure 2.a), were also studied to provide data at the mm scale. Since the 137 drill core is not oriented, all measured orientations are not in situ but relative ones. Yet, the subsamples 138 are still oriented consistently with the core sample.



148 density linked to differential absorption and were then compared to backscattered-electron (BSE)149 images obtained on S_2.

150 The CT scans were treated as image stack to extract fracture geometry using the FIJI software 151 (Schindelin et al., 2012). One of the analytical challenges was the low-density contrast between 152 fractures and the matrix in some areas. Indeed, the mineralogical phases in fractures and matrix mainly

consist of light elements such as Si and Mg, with a minor Ni amount in fractures. Depending on the 153 154 degree of mineral filling, the fractures appeared slightly darker than the matrix for poorly filled 155 fractures or slightly brighter for mineralized fractures. The challenge was thus to find a method able to 156 extract both poorly filled fractures and mineralized fractures from the matrix. Furthermore, the density 157 of the matrix also changes as a function of the weathering degree, making the extraction of the fracture 158 network even more challenging and hence the segmentation more complex. The point cloud of the 159 stack was then extracted and imported into CloudCompare software (GPL software, 2021) to extract 160 the fracture sets based on their dip/direction value. Finally, planes were adjusted from points using 161 the Ransac Shape Detection Plugin (Schnabel et al., 2007) and exported to analyze their spatial and 162 orientation distribution.

163 The orientation analysis is performed on the Stereonet software (Allmendinger et al., 2011; Cardozo 164 and Allmendinger, 2013). The spacing was analyzed by measuring the mean distance between plane 165 traces belonging to the same set on an orthogonal plan.

166 Chromite distribution provides a guide for identifying the traces of the magmatic beddings (Leblanc, 167 1980). Indeed, since chromite minerals are much denser than the matrix mainly consisting of Mg-rich 168 silicates, they are easily visible by tomography. Thus, point clouds of chromite minerals are extracted 169 from CT scans by using FIJI. Then the normal for each point is calculated before adjusting a plane for 170 each chromite alignment. Orientation of the magmatic foliation is then derived from the plane 171 orientation.

172 Fractal dimension analysis

Fractal dimension is an excellent tool to assess whether structures are statistically similar at different scales and characterize natural fracture networks (Basirat et al., 2019). Indeed, similar discontinuities have the same fractal dimension, whatever the scale. The fractal dimension of the fracture network is calculated on both the core sample and S_2 to evaluate the self-similar character of fracture patterns at the cm-scale and mm-scale. Since Ni is denser than Mg and Si, Ni is an excellent signal to discriminate mineralized fractures from the matrix. Thus, the micro-XRF Ni distribution map of the core sample was used to calculate the fractal dimension of the fracture network at the cm scale. Regarding the mmscale, the S_2 CT scan was selected because of the excellent density contrast observed between the fracture network and the matrix, which eases the extraction of the fracture network. Additional information on CT acquisition, structural features (fractures and chromite beddings) extraction and the fractal dimension analysis are given in Appendix A.

- 184
- 185 II.II. Petrophysical characterization.

186 Poro-permeability profile acquisition

187 Under weathering conditions, the hydrolysis of peridotites leads to the leaching of Mg and Si, causing 188 an increase in the porosity (Butt and Cluzel, 2013). Furthermore, previous hydrological studies at the 189 massif scale report changes in porosity and permeability with weathering stages in laterite profiles 190 (Jeanpert, 2017; Join et al., 2005). Thus, a porosity-permeability profile is acquired along the core 191 sample to describe better and characterize the weathering front in the sample. From the primary core 192 sample, gas permeability was estimated on the five sub-samples, S_3, S_4, S_5, S_6 and S_7, the others 193 being impregnated with resin for the micro-XRF mapping. For each sample, 2 to 3 different 194 permeability measures were carried out with a confining pressure of 120 bar and injection pressure 195 varying from 50 bar to 80 bar in the triaxial cell. Injection pressure and injection volumes were 196 measured over time to estimate the mass flux and thus the permeability k with Darcy's law.

197 The final permeability value corresponds to the mean value of the different measures for each sub-198 sample. Total water porosity was also estimated on the same sub-samples by measuring the weight 199 and volume differences of dry and wet samples.

Based on the values obtained for these sub-samples and the mean X-ray attenuation profile from the tomographic acquisition of the main core sample, semi-quantitative water porosity and permeability profiles were obtained on the whole sample by interpolating the different values along the vertical.

This interpolation was based on the linear regression between the porosity and the mean attenuation displayed by the grey-scale pixel intensity values of the subsamples. A similar approach was then applied to the permeability. The linear regression equation should be taken carefully since only 5 points were used to estimate it, thus providing a qualitative estimation of porosity and permeability profiles.

208 Pore size distribution

209 Mercury injection porosimetry was performed in the pressure range from 8.4 kPa to 413 MPa, 210 corresponding to pore sizes from 88 µm to 1.8 nm. This technique analyses the macro and mesopores 211 and the pore size distribution. Sub-samples S_3, S_4, S_5, S_6 and S_7 were analyzed.

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213 II.III. Element distribution

214 μ-XRF element mapping

215 Chemical maps were acquired with an M4-Tornado Bruker micro-X-ray fluorescence spectrometer (µ-216 XRF). A voltage of 50 kV and a current of 400 μ A with an acquisition time of 15 ms/pixel and spacing of 217 100 µm for the core sample and a spacing of 20 µm for the S_0 sub-zone and the S_1 and S_2 sub-218 samples were chosen to capture the signal of both heavy elements (Co, Ni, Fe, Cr) and light ones (Mg, 219 Si). Semi-quantitative analysis was carried out to assess the concentration of the different elements 220 for each map using the built-in, standardless quantification functionality of the software, which is 221 based on Fundamental Parameter analysis. In addition, a semi-quantitative 300-point line was 222 acquired to study the evolution of Ni, Mg, Fe and Si concentrations along the core sample. The chemical 223 profile is compared with a geochemical log carried out along a drill core located 44 m away from the 224 studied core sample in the Cagou Pit (latitude: 20°58'23.795" S, longitude: 164°47'29.732" E, altitude: 225 730m).

226 III. Results

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227 III.I. Structural organization

228 III.I.I. Fracture network description

229 Poorly mineralized serpentine-bearing fractures occur as low-density objects, appearing thus as dark 230 grey structural elements on tomographic images. They are denser than the olivine- and 231 orthopyroxene-rich matrix when containing high Ni amounts (Figure 3). The S_2 CT image displays the 232 clear transition between the two fracture types: the lower part of the primary fracture crossing the 233 sample is light grey, thus denser than its upper part, which appears in dark grey (Figure 3.c). This 234 transition is also well visible in the BSE imaging of sample S_2 (Figure 3.d). In both cases, the contrast 235 between the matrix and the fractures enables the discrimination of two distinct fracture sets (F1 set in 236 red and F2 in yellow), whatever the scale (Figure 3). Chromites appear as intense white specks 237 corresponding as they have the highest density (Figure 3).



239 Figure 3 - CT scans exhibiting two fracture sets highlighted by red (F1) and yellow (F2) dotted lines. a) Sample A. b) Sample

S_0. c) S_2 d) BSE imaging of S_2

Data extraction as fractures from the CT has been applied on both core and S_2 samples (Figure 4). The samples are presented with the same orientation and display similar structural organization as shown by the dip direction stereonet (Figure 4.d, Figure 4.h). The main microfracture set, F1 (dark blue in Figure 4.c, Figure 4.g), is present in both samples. The second microfracture set, F2, is less well visible in yellow. The mean poles (trend, plunge) are N80E,42 for F1 and N259E,34 for F2 in the core sample. In S_2, the mean pole of F1 is N70E,41 and N249E49 for F2. However, the F2 set is composite and contains 2 sub poles families F2_a and F2_b.



Figure 4 - Fracture extraction of core sample and sub-sample S_2. a) & b) CT scans of core sample. c) Oriented point cloud
colorized by dip direction. d) Stereographic projections of fracture orientations in the core sample (poles). e) & f) CT scans of
sub-sample S_2. g) Oriented point cloud colorized by dip direction. h) Stereographic projections of fracture orientations
(poles) of sub-sample S_2 (poles).

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The microstructures in S_2 are very similar to those of the core sample, with F1 sub-orthogonal to F2.
Planes were adjusted on data from each structural set (Figure 5.a, Figure 5.b, Figure 5.e). The relative
angle between the mean F1 set (N82E, 46) and F2 set (N254E, 40) planes is estimated at 90±4° for the
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core sample, which confirms the orthogonality of the sets (Figure 5.d). On a smaller scale, in S_2, an
angle of 90±6° between F1 (N74E,60) and F2 (N238E,24) was found (Figure 5.f).

The mean spacing for each set has been estimated from the spatial distribution of the adjusted planes. F1 has a denser distribution than F2 in the core sample with a mean spacing of 4.8 ± 0.5 mm, while F2 is characterized by a mean spacing of 6.4 ± 0.8 mm with a 95% confidence interval. In S_2, F1 and F2 present a similar spatial distribution with a mean spacing of 1.2 ± 0.3 mm for F1 and 1.4 ± 0.2 mm for F2. Furthermore, scale invariance in the structural networks is confirmed by calculating the fractal dimension of networks: 1.79 for the core sample and 1.83 for sample S_2.

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266 III.I.II Magmatic foliation analysis

267 The orientation of the magmatic foliation, restored through the analysis of the distribution of chromite 268 grains, was estimated and compared to fracture sets F1 and F2 (Figure 5). Despite a significant 269 scattering of the magmatic foliation orientations in the core sample, they are better aligned with F1 270 and subsequently orthogonal to F2 (Figure 5.c, Figure 5.d). Indeed, the Kamb contour points out the 271 significant clusters in the poles distribution of the chromite alignments (Kamb, 1959). Thus, the slight 272 angle deviation between the F1 mean pole and the center of the highest Kamb contour, estimated at 273 0±14°, supports the alignment of F1 with the magmatic foliation. This result is also found at a smaller 274 scale in sample S_2 (Figure 5.e, Figure 5.f).





Figure 5 – Comparison of structural sets with chromite grains alignment. a) 3D view of adjusted planes to F1 (blue) and F2
(yellow) of the core sample. b) North view of adjusted planes to F1 (blue) and F2 (yellow) of the core sample. c) Distribution
of the chromites (in red) in the core sample and localization of the sub-sample S2 (yellow). d) Pole distribution of F1 (blue),
F2 (yellow) with their mean vectors, and contour plot of chromites alignment (red) of the core sample. e) 3D view of
adjusted planes to F1 (blue), F2 (yellow) and chromites distribution (red) in S_2. f) Pole distribution of F1 (blue), F2 (yellow)
with their mean vectors, and contour plot of chromites alignment (red) of S_2.

282 III.II Poro-permeability characterization

The gas permeability and total water porosity along the longitudinal axis of the core increase upwards along with weathering intensity. Three different zones can be outlined (Figure 6.a,b): i) a first one defined by low porosity (< 2.5%) and low permeability (< 1E-20 m²), ii) a transition zone then occurs where porosity increases from 3% to 12%, while the permeability increases from 1E-20 m² to more than 1E-19 m², iii) in the most weathered portion of the core sample, the porosity is relatively constant at about 13%, while permeability is about 1.2E-19 m².

289 Mercury porosimetry confirms the observed trends (Figure 6.c, d). The bottom of the core sample is 290 characterized by low porosity (< 1%), while the top exhibits a higher porosity (> 7%). Furthermore, the 291 change in pore size distribution reveals that the increase in porosity results from the opening of 292 nanometer pores (< 50 nm). The connected porosity of the weathered part of the core sample (samples S_4 and S_7) is characterized at 90% by nanopores with sizes ranging from 1.8 nm to 30 nm and a 293 294 minor (10%) family of micropores with sizes ranging from 4 µm to 80 µm. Conversely, the fresh part of the core sample (S_3 and S_5) is characterized by micropores ranging from 4 µm to 80 µm, 295 296 accounting for 90% to 100% of the effective porosity. The minor family of nanopores, with sizes ranging 297 from 1.8 nm to 30 nm, accounts for 0% to 10% of the porosity for S_3 and S_4. The sub-sample S_6 298 presents intermediate features with 70% of the porosity due to nanopores and 30% to micropores 299 (Figure 6.d)

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Figure 6 - Total water porosity and permeability distribution in the core sample. a) Total water porosity. The plotted points
 represent the measured porosity, while the curve results from the linear regression between the measured porosity and the
 mean greyscale intensity value. b) Permeability. The plotted points represent the measured permeability, while the curve
 results from the linear regression between the measured permeability and the mean greyscale intensity value. c) Pore size
 distribution based on cumulated Hg porosity. d) Pore size distribution based on normalized cumulated Hg porosity

308 III.III. Element redistribution

III.III. I. Element mapping

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310 The Fe, Ni, and Mg XRF maps provide a good image of the microfracture network and their infillings at 311 different scales. Ni is mainly present in fractures at each scale, while Si, Mg, and Fe dominate within 312 the matrix (Figure 7). Two distinct areas can be distinguished (Figure 7.a, Figure 7.b). The saprock zone 313 exhibits an enrichment in Fe at the expense of Mg compared to the least weathered portion located 314 below the line. In addition, the apparent density of Ni-bearing fractures is higher in the saprock (Figure 315 7.a, Figure 7.b). In the map obtained on S 1, located in the most weathered part of the ore sample, Ni 316 occurs in both major and secondary fracture networks (Figure 7.c). On the contrary, the map obtained on S_2, located in the least weathered part of the core sample, shows that Ni is solely present within 317 318 the major fracture (Figure 7.d).



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320 Figure 7 – RGB composite map of Fe-Ni-Mg distributions in a) Core sample. b) Sample S_0. c) Sample S_1. d) Sample S_2

The semi-quantitative Ni and Fe chemical maps support previous observations: Ni is mainly located in fractures where grades vary from ca. 5 to > 9 wt% (Figure 8). In the weathered part, the matrix is richer in Ni (grades up to ca. 3 wt%). On the contrary, the Ni content drops from ca. 2 wt% Ni near the line to
1 wt% Ni in the freshest part of the sample (Figure 8.a.1, Figure 8.a.2). In sample S_2, the matrix is Ni
poor with values close to the fresh rock (Figure 8.a.4).



Figure 8- Semi-quantitative concentration maps of a) Ni of 1. Core Sample. 2. Sample S_0. 3. Sample S_1. 4. Sample S_2.
and of b) Fe of 1. Core Sample. 2. Sample S_0. 3. Sample S_1. 4. Sample S_2.

The Fe-map also displays two chemical domains for the core sample and sample S_2 (Figure 8.b). The Fe-rich region occurs in the uppermost part of the core sample, closer to the top than the Ni-rich area (Figure 8.b.1, Figure 8.b.2). Fe content exceeds the 10 wt% in the weathered area and remains close to 8 wt% in fresher rock. Fractures are free of Fe, as underlined by the white arrows.

333 III.III. II. Chemical profiles

334 The Ni, Fe, Mg, and Si chemical transect across the core sample reveals the geochemical impact of 335 weathering at the cm-scale (Figure 9.a, Figure 9.b). To draw comparisons with systems at a larger scale, 336 we report a geochemical log carried out along a drill core located in Cagou Pit (Figure 9.c). From the 337 least to most weathered part of the core sample (i.e. from bottom to top), Fe and Ni concentrations 338 increase at the expense of Mg, while Si remains constant (Figure 9.a). Three different zones are thus 339 distinguished in the profile. The first one is characterized by low Ni (0.5 wt% - 1 wt%) and Fe (7 wt%) 340 concentrations and high Mg (27 wt%) and Si (20 wt%) concentrations. The second zone exhibits an 341 increase in Ni up to 2 wt%, Fe up to 9 – 10 wt%, and Mg decrease to 20 wt%. The third zone is associated with a sharp rise of Ni and Fe contents up to 3 wt% and 15 wt%, respectively, while the Mg content remains around 20 wt%. Whereas the broader trends represent the evolution of the matrix composition, Ni spikes result from microfractures. The analysis of the Ni signal in the transition zone using a Fast Fourier Transform reveals a periodicity of the spikes of 7 mm, similar to the estimated spacing of F1 (5 mm) and F2 (6 mm) in the core sample.

At a larger scale, the 30 m-long drill core crosses from bottom to top, i) peridotite rock (up to 26 m), ii) earthy saprolite interlayered with slightly weathered rock (from 26 m to 19 m), and (iii) earthy saprolite (up to 5 m). The drill core exhibits geochemical trends similar to those observed within the 30 cm-long core sample (Figure 9.c). Specifically, from bottom to top, Fe and Ni concentrations increase at the expense of Mg, while Si remains constant at about 20 wt%. Furthermore, the three geochemical zones identified in the core sample are expressed at a larger scale along with the entire drill core.



Figure 9 – a) Semi-quantitative Ni, Fe, Mg, Si profile in wt% along with the core sample. b) RGB composite map of Fe-Ni-Mg
in the core sample, defining three distinct geochemical zones – 1: Poorly weathered area – 2: Transition area – 3: Weathered
area. c) Geochemical log (Ni, Fe, Si, Mg in wt%) of a drill core. d) Ni-rich fracture network (green filling) at the outcrop scale.
Two sub orthogonal fracture sets are observed: F1 (black) and F2 (red).

358 IV. Discussion

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IV.I. Weathering rate along the penetrating alteration front

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362 The evolution of whole-rock density, porosity and permeability in the core sample corresponds to a 363 petrophysical and chemical gradient from the quasi-fresh facies at the bottom to an advanced 364 alteration state at the top. The main feature is a density decrease correlated to porosity increase. The measured petrophysical properties of the fresh rock (i.e., low porosity of ~1% and very low 365 366 permeability < 1E-20 m²) are similar to those obtained on fresh Oman harzburgites (Dewandel et al., 367 2005; Katayama et al., 2020). Both peridotites are comparable in terms of chemistry and mineralogy 368 (Al-Khirbash, 2015). Therefore, the properties of both Oman and New Caledonia ophiolite aquifers 369 would be similar (Dewandel et al., 2017; Jeanpert, 2017).

370 The chemical evolution at the core sample scale is similar to that at the outcrop scale 371 (Cathelineau et al., 2017; Muñoz et al., 2019; Teitler et al., 2019). As attested by high Mg and Si bulk 372 contents, the olivine remains preserved from dissolution in the fresh facies. Ni is locally present in the 373 prominent fractures as newly-formed phyllosilicates within an almost unaltered matrix. The newly-374 formed phyllosilicates correspond to Mg-Ni talc-like, as already observed in previous studies from the 375 same area (Cathelineau et al., 2017, 2016; Muñoz et al., 2019). In the weathering front, the porosity 376 and permeability increase with nanoporosity development. Mg and Si remain elevated, while Ni and 377 Fe start to increase. Ni enrichments correspond to the two sub- orthogonal microfracture sets, e.g. F1 378 and F2. The FFT analysis from the Ni profile (zone (2)) reveals a spatial periodicity of 7 mm, which is 379 close to the mean spacing of F1: 5 mm and F2: 6 mm. When weathering progresses, olivine dissolves 380 and the bulk Fe and Ni content increase significantly while Mg decreases. Fe precipitates as goethite 381 and serpentine (mostly lizardite) remains preserved (Myagkiy et al., 2017; Teitler et al., 2019). Ni enrichment mainly occurs in the fractures and within the micro-fractured matrix in the altered zone.
At this weathering stage, the porosity is relatively high (around 10%), and the nanopores account for
almost all the connected porosity.

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IV.II. Element redistribution on fractures network

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387 The distribution of three different weathering rates in the core sample results from several processes 388 that are not all synchronous (Figure 9.b). When the olivine dissolution front progresses downwards, a 389 part of magnesium and nickel is trapped by the precipitation of phyllosilicates such as Mg-Ni talc-like 390 (Cathelineau et al., 2017, 2016; Myagkiy et al., 2017). These secondary minerals are richer in Si and Ni 391 and poorer in Mg than the original silicates, explaining thus the enrichments. The precipitation of Fe-392 oxy-hydroxides fixes iron (Fe^{3+}). Ni excess migrates following the fracture network towards deeper, 393 poorly weathered levels. This migration ahead of the dissolution front saturates fractures, creating the 394 transition area observed in the core sample. Ni enrichment occurs mainly in the vicinity of the main 395 microfractures, within the dense network of serpentine microcracks that forms the so-called "mesh" 396 structure (Cathelineau et al., 2017; Muñoz et al., 2019). In this area, silicates are mostly un-weathered. 397 The dissolution-precipitation process triggers the bulk permeability increase with the nanopore 398 formation. Thus, a transition occurs from diffusion- to advection-driven reaction front at the sub-mm 399 scale. The nanopores are probably controlled by the distribution of serpentine and its nano-defects. 400 Therefore, in increasingly serpentinized rocks, the nano-defects may affect the distribution of 401 nanopores (Tutolo et al., 2016). Dewandel et al. (2005) also reported a similar pore size distribution on 402 fresh peridotites from Oman and interpreted it related to the serpentine vein microstructures. Our 403 data indicate that the weathering processes can also contribute to the nanoporosity formation in 404 serpentinized peridotite. Inherited nano defects may probably explain the enhanced nanoporosity 405 development. In addition, the supergene weathering of the serpentine and the opening of inherited

serpentine microfracture and their infilling by newly formed phyllosilicates both impact the pore size
distribution. The dissolution front favors the onset of nanopores, which extends it to the whole sample.

408 This redistribution model highlights the impact of pre-existing fracture networks, which act as 409 preferential pathways for the Ni-Mg-Si transfers and their reprecipitation as newly formed minerals.

- 410

411 IV.III. Multiscale redistribution processes

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413 The structural organization of the fracture network and the chromite layering of both core sample and 414 sample S_2 exhibits a predominant fracture set (F1) parallel to the chromite alignments and a second 415 set (F2) sub-orthogonal to F1. Previous studies have highlighted the link between chromite layering 416 and magmatic foliation (Cassard et al., 1981; Cluzel et al., 2012; Prinzhofer et al., 1980) and have 417 suggested an N-S striking orientation for such lineation (Cluzel et al., 2012). Furthermore, this lineation 418 would be relicts of the oceanic lithosphere (Cluzel et al., 2012). Thus, a geometric link between the 419 fracture network and the magmatic bedding could be inferred, with F1 oriented parallel to the 420 magmatic lineation and F2 orthogonal to such orientation. Besides, recent studies point out that 421 fractures filled by primary serpentine (lizardite) are also inherited from relicts of the oceanic 422 lithosphere (Cathelineau et al., 2017; Iseppi et al., 2018; Quesnel et al., 2016). In addition, such an 423 inherited fracture network often displays sub-orthogonal sets of fractures striking N0-20°E, N50-70°E, 424 N90-100°E and N120-140°E with variable dips from 20 to 90° (Iseppi, 2018; Iseppi et al., 2018). Such a 425 common origin of the magmatic lineation and the inherited fracture network, combined with a similar 426 orientation between such structures, may allow suggesting an orientation of the fracture network in 427 the core sample close to what has been described by Iseppi et al. (2018). A similar structural 428 organization occurs on larger scales (Figure 10). Sub-orthogonal fracture sets from the Cagou and the 429 Test pit (Figure 1.b) are described from mm to 10 m scale. At the outcrop scale (1m to 10m), 430 Cathelineau et al. (2016) also describe sub-orthogonal structures for Ni talc-like crack-seal fractures 431 with two strike families of N150° and N50° at the Cagou pit. At the massif scale , such as the example of Koniambo, four dominant fracture sets strike N0°, N45°, N90° and N135° (Jeanpert et al., 2019; 432 433 Quesnel et al., 2016). The serpentine fracture network is characterized by an N130° dominant trend 434 associated with N90°, N45° and N0° directions (Iseppi et al., 2018). This general structural organization 435 at the island scale shows two different sets of two sub-orthogonal families (N0° associated with N90° 436 and N45° associated with N130°). Thus, it is proposed that a specific structural organization including 437 one to two sub orthogonal sets of fracture striking N0° – N90° and N45° – N135° would be developed 438 at different scales, which suggests that similar structural processes would have occurred to constrain 439 the system. A fractal dimension estimated on the core sample and the sub-sample S_2 can be proposed 440 at around 1.8 to quantify the self-similarity of the fracture network.



442 Figure 10 - Multiscale image panel of fracture networks. The images are coming from samples located in Cagou and the Test

443 444 pit (Fig1.b). On the interpreted images, two sub orthogonal sets are observed: F1 (black and yellow) and

445 (Jeanpert et al., 2019) have already pointed out the relationship between fracture distribution and 446 permeability at the outcrop scale. Our core-scale analysis supports this scaling effect on permeability. 447 At the decimeter scale, the permeability of a fresh peridotite is estimated around 1e-15 m². Such value 448 is far higher than the measured permeability in the core sample, i.e. between 1e-21 m² and 1e-19 m². 449 Instead, such permeability would result from fractures with a spacing of 0.35 m (Figure 10), yielding 450 an aperture of 1.6e-5 m using a cubic law (Jeanpert, 2017). Similarly, at the 10 m scale, the permeability 451 increases up to 1e-13m² - 1e-11m², which is interpreted as draining fractures with a spacing of 30 m 452 and an aperture of 1.5e-3 m, consistent with in situ observations (Figure 10).

453 This self-similarity in the structural organization at the different scales is also observed in the different 454 weathering stages. Specifically, at the sample scale, this study highlights that the progression of the 455 silicate dissolution front through a fracture network results in the redistribution of Ni and the 456 development of different weathering facies within a few tens of centimetres. Such a gradation of the 457 weathering products is also observed at the outcrop scale. The base of the lateritic profile usually 458 includes a transition domain, up to a few meters in thickness, where Ni-rich fractures crosscut the 459 unaltered bedrock. Upwards, this transition domain is delimited by the silicate dissolution front. In the 460 saprolite above the front, secondary phyllosilicates formed away from the primary mineralized fractures (Cathelineau et al., 2016). The chemical (this study) and mineralogical (previous studies at 461 462 the same location: Cathelineau et al. (2017, 2016); Muñoz et al. (2019); Teitler et al. (2019)) evolutions 463 observed in lateritic profiles mimic, at a larger scale, the cm-scale processes observed in the core 464 sample. In particular, the Ni distribution between the fractures and the saprolite matrix is similar at 465 both scales. Thus, an average Ni grade in the saprolite ore is about 2-5 wt% (Butt and Cluzel, 2013), while Mg-Ni talc-like veins grade over 10 wt% (Cathelineau et al., 2017). 466

The chemical profile acquired on the core sample can thus be compared with drill core assay data
(Figure 9). Indeed, when the log presents a continuous weathering process, the geochemical trends
observed at the scale of a few decimeters are similar to those found commonly in the saprolite zone

of ultramafic-derived lateritic profiles (Teitler et al., 2019) (Figure 9.b). Such structural, weathering and
geochemical similarities observed from 1e-3 m to 10 m scale highlight the impact of fractures network
on weathering and redistribution processes at each observation scale. Hence a better understanding
of the structural organization of the sample could provide light on the spatial geochemical distribution.

474 V. Conclusion

This study presents a multiscale approach combining structural, petrophysical, and geochemical
analysis. The goal was to study the impact of the structural organization on the weathering processes
and the Ni redistribution at different scales to discuss the similarities. Observation conducted at a small
scale gives insight into the general mechanisms typical of the whole lateritic profiles. The main findings
are:

Weathering and Ni-enrichment are fracture controlled. The magmatic foliation guided the
development of the serpentine networks, which constitute the active network favoring the
penetration of meteoric waters. The main tectonics lineations observed in the Cagou sample
and at the scale of Koniambo massif are NS, EW, NWSE, and SWNE. Microfracture sets are
organized similarly at the sample scale and the open pit scale or even larger scale. This scale
invariance suggests a common origin of the fracture networks, confirmed by their similar
serpentine infillings and fractal distribution.

The proposed multi-step Ni-redistribution at the core scale is consistent with existing chemical
 profiles in laterites. Fractures constitute sites of early fixation of Ni as newly-formed Mg-Ni phyllosilicates. The reactivated serpentine microfractures are the primary enrichment locus
 before a bulk nickel redistribution by the dissolution front.

The increase in porosity results from several superimposed processes: i) mesopore formation
 through olivine dissolution, ii) partial to full infilling of the pores by newly formed minerals
 such as neo-formed phyllosilicates and smectite described in previous works (Muñoz et al.,
 2019; Teitler et al., 2019). The newly formed phases display a remarkable high content of

495 nanopores. Further studies will be needed to determine the increase of the nanoporosity as it
496 may affect the redistribution processes at a microscopic scale by favoring diffusion over
497 advection.

The weathering fronts observed at a small scale on a core sample are excellent analogues of
 the larger scale laterite profiles. In such profiles, the inherited fracture networks filled by
 serpentine developed in earlier stages and depend on magmatic microtextures and
 subduction-obduction history. They constitute the Ni-trap and show fractal dimensions.

The Ni-redistribution results from a hierarchical and multi-scale organization related to the
 previous deformation. The self-similarity of spatial distribution patterns implies scaling laws.
 The evaluation of upscaling laws would, in consequence, open up great opportunities for
 designing reliable large-scale model geometry and better predictive models of Ni mineralization location.

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715 Appendix A

716 CT acquisition

The CT methodology drives the resolution of each sample. Indeed, it is mainly determined by the fixed number of detectors and the size of the analyzed sample. The bigger the sample is, the lower the resolution is since each detector analyses signal for a more significant part of the sample. Thus, a resolution of 65 μ m/vx was obtained by using a voltage of 120 kV and a current of 110 μ A for the core sample, and 12.5 μ m/vx using a voltage of 85 kV and a current of 95 μ A for S_2.

722 Features extraction

1. Image stacks analysis

724 The first step of the structural features extraction is based on image analyses on FIJI. A local auto 725 threshold computed for each pixel within a set radius was applied using the Niblack method (Niblack, 726 1986). Based on the standard deviation calculation, this method highlights significant changes in grey 727 values and thus reveals the microfracture network. Then, successive median filters were applied to 728 reduce the noise until the visual aspect of the images was estimated to be correct. The resulting images 729 were then skeletonized to describe the microfractures better. Indeed, this method removes pixels 730 repeatably from the edges of objects (in current work, the fractures) until they are reduced to single 731 pixel-wide skeletons, allowing them to be identified.

732 2. Point cloud analysis

On CloudCompare, the point cloud was first sub-sampled randomly to reduce its size for computing costs. Then, the normal vector for each point was calculated through the Hough Normal Computation Plugin (Boulch and Marlet, 2012). This plugin estimates the normal by finding the best one in a neighbourhood around a chosen point. Then, the normal is converted into a dip/dip direction scale factor to colorize the point cloud. Distinct fracture sets were identified based on the dip and dip direction distribution, and each point was assigned to a given set. Finally, planes were adjusted from points using the Ransac Shape Detection Plugin (Schnabel et al., 2007) and exported to analyze theirspatial and orientation distribution.

741 Fractal dimension analysis

742 Since fractal analysis is noise sensitive, a prerequisite for evaluating the potential fractal organization 743 of fractures, is to reduce unwanted noise, hence allowing a better extract of the structural features 744 (Pant, 2013; Reiss et al., 2015). The background noise of CT images was thus treated with FIJI by 745 applying a local auto threshold with the Niblack method, followed by an outlier removal through the 746 analyze particle plugin and a skeletonize step. Then the fractal dimension of the fracture network was 747 calculated by the box-counting method (Roy et al., 2007) using the FracLac plugin (Karperien, 2013). 748 The box sizes vary from 1 pixel to 45% of the height of the images to avoid errors in the estimation 749 (Walsh and Watterson, 1993). Furthermore, to reduce the bias related to the grid location (Roy et al., 750 2007), 50 different positions were used for each image, and the final fractal dimension corresponds to 751 their mean value. The calculation was restricted to only a central portion of S_2 to avoid disturbances 752 from part of the image outside the sample.

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