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# Fluid Evolution at the Batu Hijau porphyry Cu-Au deposit, Indonesia: Hypogene Sulfide Precipitation at Low-Temperature from a Single-Phase Aqueous Magmatic Fluid during Chlorite-White Mica Alteration

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1 **Fluid Evolution at the Batu Hijau porphyry Cu-Au deposit, Indonesia:**  
2 **Hypogene Sulfide Precipitation at Low-Temperature from a Single-Phase**  
3 **Aqueous Magmatic Fluid during Chlorite – White Mica Alteration**

4  
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11  
12 **Abstract**

13 Mineralization at the Cu-Au porphyry deposit of Batu Hijau, Indonesia, was previously reported  
14 to be associated mainly within stockwork quartz veins associated with pervasive biotite-  
15 magnetite alteration. We use cathodoluminescence (CL) imaging of vein textures followed by  
16 microthermometry and LA-ICP-MS microanalysis of fluid inclusions to decipher the spatial-  
17 temporal evolution of the hydrothermal system. Our results indicate that sulfide precipitation  
18 largely postdated the main stockwork veining. Chalcopyrite and bornite were found in three  
19 textural positions: (1) within conspicuous quartz-poor veinlets (“paint veins”) that post-date  
20 quartz stockwork veins and which also appear to account for the bulk of seemingly disseminated  
21 sulfides; (2) as centerlines in B-type veins; and (3) as interstitial grains in A-type veins. In all three  
22 textural positions, the sulfides occur together with a volumetrically minor, dull-luminescent  
23 quartz generation interstitially to the granular quartz dominating the stockwork veins. All three  
24 positions are associated with chlorite ± variable phengitic white mica with 3 – 6 wt.% FeO + MgO.

25 In the barren core of the deposit, quartz veins host, almost exclusively, fluid inclusions of  
26 intermediate density (~0.6 g/cm<sup>3</sup>) and near-constant salinity of ~3.7 wt.% NaCl<sub>eq.</sub>, representing  
27 the input magmatic fluid. This fluid subsequently separated into a highly saline brine<sup>1</sup> and low-  
28 density vapor during quartz vein formation in the mineralized parts of the deposit; however, we

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<sup>1</sup> ‘brine’ is used here for a hypersaline liquid saturated with halite at room temperature and homogenizing to a liquid upon heating, ‘intermediate density fluid’ homogenizes near-critically by meniscus fading, whereas ‘vapor inclusions’ homogenize to vapor (Heinrich, 2005, tab. 2; Liebscher & Heinrich 2007).

29 found no textural or fluid-chemical evidence that brine + vapor already reached saturation in  
30 sulfides. Within the studied samples, Cu-Fe-sulfides are invariably associated with the dull-  
31 luminescent quartz hosting only low-salinity (~2 to 8 wt.% NaCl<sub>eq</sub>) aqueous fluid inclusions with a  
32 density of ~0.8 g/cm<sup>3</sup> and minimum formation temperatures of 360° to 300°C, in agreement with  
33 Ti-in-quartz and chlorite thermometry indicating trapping conditions only slightly above the  
34 boiling pressure of these liquids. On average, this mineralizing aqueous fluid is compositionally  
35 similar to the initial magmatic fluid, suggesting a common source, but some inclusion assemblages  
36 deviate to significantly higher or lower salinities (25 and 0.5 wt.% NaCl<sub>eq</sub>, respectively).

37 We propose a formation model for the Batu Hijau porphyry Cu-Au deposit in which mostly  
38 barren quartz veins formed at high-temperature (> 400°C) in the central part of the system, while  
39 sulfide mineralization commences to form peripheral to this zone. The economic ore shell was  
40 growing in- and downwards as zones of active sulfide precipitation at 360°-300°C shifted in  
41 response to progressive retraction of isotherms, while barren quartz vein formation continued in  
42 the system's core at higher temperature. Some of the ore-forming aqueous liquid may have  
43 contracted directly from intermediate-density magmatic fluid, but the large salinity variations  
44 above and below the average are interpreted as evidence for re-homogenization of magmatic  
45 brine and vapor that had previously separated and became miscible again after cooling and  
46 contraction of the dominant former vapor. Retrograde solubility of quartz near 400°C and ~ 200  
47 bar, which lead to partial dissolution of earlier formed quartz veins and created cavities and cracks  
48 for sulfide deposition. This evolution of a single magmatic fluid, with transient phase separation,  
49 but limited physical segregation of transient brine and vapor as they flow through the ore shell,  
50 may explain the observed positive correlation between ore grades and quartz vein density in the  
51 ore shell, even though sulfide precipitation overprints high-temperature quartz precipitation on  
52 the vein scale. A brief comparison with published observations indicates that this re-  
53 interpretation may apply to many other porphyry Cu-(Au-Mo) deposits worldwide.

54

## Introduction

55 Porphyry-style deposits are major sources of Cu, Mo, and Au, and their magmatic-  
56 hydrothermal systems have been studied in detail over many decades (Sillitoe, 2010 and  
57 references therein). Early detailed mapping of porphyry deposits (e.g., Lowell and Guilbert, 1970;

58 Lowell, 1974; Gustafson and Hunt, 1975) led to similarities of porphyry systems being identified  
59 worldwide, indicating similar physicochemical conditions of ore formation (e.g., Henley and  
60 McNabb, 1978; Seedorff et al., 2005; Sillitoe, 2010). The deposits form around a central  
61 porphyritic stock or dike swarm, leading to a common sequence of veining associated with a  
62 coaxially arranged alteration pattern and a commonly bell-shaped ore body (e.g., Lowell and  
63 Guilbert, 1970) with a distinct temporal sequence of igneous and hydrothermal features  
64 (Gustafson and Hunt, 1975; Redmond and Einaudi, 2010; Sillitoe, 2010). Fluid inclusions within  
65 most porphyry-related hydrothermal quartz veins typically consist of two types, low-salinity vapor  
66 and high-salinity brine (Roedder, 1971, 1984; Nash, 1976; Cline and Bodnar, 1991; Bodnar, 1995;  
67 Landtwing et al., 2005; Redmond and Einaudi, 2010); these two coexisting fluids formed by  
68 separation of a single-phase magmatic fluid of low- to medium salinity (Henley and McNabb,  
69 1978) commonly preserved as intermediate-density fluid inclusions in the deep core of the  
70 deposits (Redmond et al., 2004; Landtwing et al., 2005). High metal grades spatially correlate with  
71 potassic alteration and hydrothermal quartz veins hosting predominantly brine and vapor fluid  
72 inclusions (Gustafson and Hunt, 1975; Reynolds and Beane, 1985; Ulrich et al., 2002). This  
73 association of alteration, quartz veins and inclusion style, in combination with the truncation of  
74 mineralized quartz veins by the next younger porphyry intrusion (e.g., Gustafson and Hunt, 1975;  
75 Proffett, 2003), imply that at least some Cu-Fe-sulfide precipitation occurred during early quartz  
76 vein formation from a single-phase or two-phase fluid of highly saline brine coexisting with low-  
77 salinity vapor (Proffett, 2009; Sillitoe, 2010), probably at temperatures above 500° (Eastoe, 1978;  
78 Heithersay and Walshe, 1995; Harris et al., 2003; Proffett, 2003; Seedorff et al., 2005; Hezarkhani,  
79 2009).

80 Cathodoluminescence imaging of porphyry quartz veins has become decisive to clarify the  
81 relationship between quartz vein formation and sulfide precipitation, and it indicates that quartz  
82 can be repeatedly dissolved and re-precipitated within veins of porphyry Cu deposits (Rusk and  
83 Reed, 2002). Detailed petrographic studies of several deposits with cathodoluminescence  
84 imaging have found evidence for similar episodes of quartz dissolution and precipitation over a  
85 large range of temperatures and pressures (Redmond et al. 2004; Landtwing and Pettke, 2005;  
86 Landtwing et al., 2005, 2010; Klemm et al., 2007; Müller et al., 2010; Vry et al., 2010; Stefanova  
87 et al., 2014; Gregory, 2017). In all cases studied so far, Cu-Fe sulfides are associated with a



88 volumetrically minor quartz generation, typically marked by distinctively low CL intensities. These  
89 studies imply that most sulfide precipitation occurred after the main mass of quartz vein  
90 formation at any one location, during or after partial quartz re-dissolution, from a fluid that was  
91 cooler than that which deposited the original quartz veins. Sulfide precipitation is commonly  
92 associated with re-opening of early veins, but the paragenetic sequence might be obscured if only  
93 macroscopic observations are taken into account. Evidence for high Cu-Fe sulfide solubility above  
94 400°C (Crerar and Barnes, 1976; Hezarkhani et al., 1999; Liu and McPhail, 2005) and for a region  
95 of retrograde quartz solubility below approximately 1kbar (Fournier, 1983) are consistent with  
96 sulfide precipitation at low temperatures and pressures (Landtwing et al., 2010; Monecke et al.,  
97 2018). The aim of this paper is to use CL-petrography and fluid inclusion analysis to define the  
98 conditions and fluid state of Cu-Fe-sulfide precipitation at Batu Hijau, and to understand more  
99 generally the deposit-scale fluid evolution that leads to the — apparently conflicting but common  
100 — observation that a high density of early quartz veins coincides with high Cu grades deposited  
101 later in the paragenetic sequence.

102

## **Geological Framework**

103 Batu Hijau is an Au-rich porphyry Cu deposit following the definitions of Sillitoe (1979; >0.4 g/t  
104 Au) and Kesler et al. (2002 ; Cu/Au <40,000), with total mineable reserves estimated at 914 Mt of  
105 ore averaging 0.525 wt.% Cu and 0.403 g/t Au (Clode et al., 1999). It is located in the  
106 southwestern corner of Sumbawa, an island within the E trending active intra-oceanic Sunda –  
107 Banda Arc in Indonesia (Fig. 1A). The Pliocene deposit lies at a major structural discontinuity  
108 cutting the arc, caused by the subduction of the Roo Rise, an oceanic plateau on the Indian Ocean  
109 plate (Garwin, 2000). Slab tearing due to subduction of the buoyant oceanic plateau is indicated  
110 by whole-rock geochemical and radiogenic isotope data, suggesting an asthenospheric mantle  
111 contribution to arc magmatism (Fiorentini and Garwin, 2010). This perturbation of the subduction  
112 zone is marked by a crustal-scale arc-transverse fault zone, which focused magma ascent and  
113 controlled the distribution of upper-crustal igneous activity (Garwin, 2002). The low K content of  
114 intrusive rocks in the vicinity of the mine area is consistent with rapid intrusion through thin  
115 oceanic crust approximately 14 to 23 km thick (Hamilton, 1979; Barbieri et al., 1987).

116 Batu Hijau is centered on a complex of pipe-shaped low-K calc-alkaline tonalite porphyritic  
117 intrusions, which intruded along the contact between gently westward-dipping volcanoclastic  
118 sequences of andesitic composition and a precursor equigranular quartz-diorite pluton (Fig. 1C;  
119 Meldrum et al., 1994; Garwin, 2000). The complex consists of three different porphyry intrusions;  
120 the mineralized and hydrothermally altered Old and Intermediate Tonalite are both crosscut by  
121 the Young Tonalite that is weakly mineralized and altered and post-dates the formation of  
122 economic ore grades (Clode et al., 1999). All tonalite intrusions consist of quartz, plagioclase,  
123 biotite, and hornblende phenocrysts in a fine-grained quartz – plagioclase matrix. The size and  
124 abundance of quartz phenocrysts increase from the Old to the Young Tonalite. Distinction  
125 between the Old Tonalite and the Intermediate Tonalite is only possible in a few places where the  
126 latter truncates quartz veins in the former, or by the presence of mineralized quartz vein xenoliths  
127 in the Intermediate Tonalite (Proffett, pers. commun., 1998). Due to the almost fresh appearance  
128 and much lower density of hydrothermal quartz veins, the relatively barren Young Tonalite is easy  
129 to distinguish from both mineralized porphyries (Fig. 2). Amphibole – plagioclase  
130 thermobarometry indicates minimum crystallization depths for phenocrysts of 8 km, significantly  
131 deeper than the emplacement depths of the tonalite porphyries at  $\leq 2$  km (Garwin, 2000). Recent  
132 chemical abrasion – isotope dilution – thermal ionization mass spectrometry (CA-ID-TIMS) U-Pb  
133 zircon dating of these intrusions indicate a maximum duration of the main hydrothermal  
134 mineralization of less than  $90 \pm 32$  ky, bracketed by the emplacement of the three porphyries  
135 between  $3.736 \pm 0.023$  Ma and  $3.646 \pm 0.022$  Ma as determined by the youngest zircon grain from  
136 each sample (Large et al., 2020). Earlier U-Pb SHRIMP (Garwin, 2000) and LA-ICP-MS ages  
137 (Maryono et al., 2018) are consistent with these high-precision results.

## 138 **Vein Sequence, Alteration and Mineralization**

139 Distribution of hydrothermal mineralization, crosscutting relationships of various vein types  
140 and alteration patterns at Batu Hijau (Fig. 1, 2 and 3) are similar to other porphyry systems  
141 worldwide regarding spatial and temporal relationships (e.g., Gustafson and Hunt, 1975). Batu  
142 Hijau shows a regular concentric and vertical zonation of a high-grade ore shell overlying and  
143 surrounding a barren core. Veins and alteration types are categorized by Mitchell et al. (pers.  
144 commun., 1998) and Clode et al. (1999) into four types following the nomenclature of Gustafson

145 and Hunt (1975). Truncations of earlier quartz veins and sharp breaks in ore grade at intrusive  
146 contacts define timelines, indicating an early stage of bornite mineralization in some areas of the  
147 deposit (Setyandhaka et al., 2008); however, clear examples of such relations between the Old  
148 and Intermediate Tonalite were not encountered in the present study and have so far not been  
149 studied by CL-petrography. Here, we emphasize the importance of ‘paint veins’ (after Fournier,  
150 1967), which are thin fractures coated by sulfides, truncating most quartz veins and intrusive  
151 contacts and presently hosting most of the ore associated with the earlier two porphyries. We  
152 focus on a N-S section through the deposit (Fig. 1C; mine data supplemented by own  
153 observations) and refer to three deposit parts as barren core (centered on the tonalite complexes  
154 at depths of 750 to 1200 m relative to the pre-mining surface), mineralized flanks (roughly  
155 following the margins of the tonalite complex surrounding the barren core) and the mineralized  
156 center (from the pre-mine surface to the top of the barren core). Observations were made during  
157 three visits in 2009 (Zwyer, 2010), 2016 and 2017, studying mine outcrops and 7 drill holes and  
158 taking more than 100 samples from which 62 thin or doubly-polished sections were cut. The  
159 following description is based on own observations of mineralogy and local timing relations in  
160 core, largely using the nomenclature of Fournier (1967) and Gustafson and Hunt (1975), and  
161 building on earlier observations by Mitchell et al. (pers. commun., 1998), Code et al. (1999), Arif  
162 and Baker (2004) and Setyandhaka et al. (2008). A summary description of the samples and main  
163 vein types is given in Table 1 and 2, respectively.

#### 164 *Quartz-dominated veins and biotite – magnetite alteration*

165 The earliest veins are discontinuous, wavy, magnetite-dominated quartz veins with biotite-rich  
166 alteration envelopes, called Am-type veins (Fig. 3C and D). They are commonly crosscut by  
167 discontinuous and undulating thin veins consisting entirely of anhedral quartz (A veins, Fig. 3A)  
168 and less common B veins with parallel walls, a main fill of euhedral quartz crystals and a  
169 characteristic sulfide centerline (Fig. 3E). AB-type quartz veins are thicker and straighter than A  
170 veins but lack a centerline (Fig. 3B and D), with local crosscutting relationships always showing A  
171 veins cut by AB and finally B veins. From A to B veins the quartz grains change from granular and  
172 anhedral to euhedral shapes, growing from the wall-rock into open space, which was eventually  
173 filled by sulfides. Within all quartz-vein types, minor feldspar, magnetite, anhydrite, biotite,  
174 chlorite, and rutile, as well as bornite, chalcopyrite, pyrite and molybdenite can occur. Magnetite

175 is commonly enriched along the vein walls, indicating reopening of early Am veins. Open spaces  
176 within quartz veins are abundant to depths of up to 1000 m below the pre-mining surface and  
177 might be the result of anhydrite dissolution by ground water as purple anhydrite variably  
178 hydrated to gypsum is present in deeper parts of Batu Hijau. In general, A veins are the dominant  
179 vein type whereas B veins are minor but slightly increasing in abundance in the shallower parts  
180 of the deposit.

181 Biotite – magnetite alteration is associated with quartz-rich stockwork veins (type Am, A, AB  
182 and B) and characterized by pervasive replacement of primary mafic minerals by brown shreddy  
183 biotite and magnetite (Fig. 3G). Primary Ca-rich plagioclase (andesine to oligoclase) is partially  
184 replaced and overgrown by more Na-rich compositions (Clode et al., 1999). Hydrothermal K-  
185 feldspar is generally absent. Biotite – magnetite alteration assemblages are preserved in the  
186 barren core, where quartz vein densities are high but Cu-Fe sulfides are rare (Clode et al., 1999;  
187 Setyandhaka et al., 2008). The biotite – magnetite alteration zone grades outwards to a propylitic  
188 alteration zone, which is subdivided into an inner actinolite and an outer epidote-chlorite domain  
189 (Garwin, 2000; Wilkinson et al., 2015).

#### 190 *Veins and fractures with chlorite – white mica alteration*

191 Chalcopyrite – bornite veinlets associated with feldspar-stable chlorite – white mica alteration  
192 envelopes, called C veins, are < 0.5 cm wide and rare compared to quartz-dominated vein types  
193 at Batu Hijau (Clode et al., 1999; Arif and Baker, 2004). By far more abundant are hairline fractures  
194 coated with chalcopyrite and minor bornite or pyrite, which can be inconspicuous in cut sample  
195 surfaces and appear as ‘disseminated’ sulfides (Fig. 2A and B). However, they are ubiquitous in  
196 the ore shell, as mineralized rocks almost invariably break during blasting or hammering along  
197 these greenish, sulfide-coated fractures. Because of this appearance on broken surfaces, we call  
198 them “paint veins” following Fournier (1967). Both, C veins and paint veins crosscut all quartz vein  
199 types, although actual timing relationships can be obscured by the presence of open spaces and  
200 the interruption of their thin alteration halo at intersections with unreactive vein quartz (e.g., Fig.  
201 3B). Commonly, only a crack propagating through earlier quartz veins or a few aligned sulfide  
202 grains document the real time relationships. Paint veins can merge with the centerline infill of B  
203 veins, commonly introducing chlorite or white mica with sulfides into these veins and adjacent  
204 wall rock (Fig. 3L). Themselves they contain little or no quartz. Due to their similar timing and

205 mineralogy, both vein types have probably formed at similar times and conditions, with C veins  
206 just being thicker and more distinctly filled paint veins. Based on their mineralogy there appears  
207 to be a spectrum of paint veins ranging from bornite-dominated, chlorite-rich to chalcopyrite-  
208 dominated, white mica-rich versions. A sharp decrease in ore grade and drastic change in chlorite  
209 – white mica alteration intensity at intrusive contacts, and locally discernable truncations of paint  
210 veins (Fig. 2) indicate that they mostly pre-date the emplacement of the Young Tonalite, but there  
211 are also paint veins (as well as a few A-type quartz veins) postdating it. No sample material was  
212 available to establish timing relations at intrusive contacts between Old and Intermediate  
213 Tonalite. In general, paint vein truncations at intrusive contacts might be rarely visible due to the  
214 thin and porous appearance of those veins, which renders them more prone to be reworked by  
215 the infiltrating melt close to the contact. Conversely, paint veins might be re-opened during final  
216 cooling and extend into the younger intrusion. Although paint veins have been reported in  
217 previous studies as fracture coatings (Clode et al., 1999; Setyandhaka et al., 2008), their  
218 importance for Cu introduction has not been evaluated. The volumetric abundance of paint veins  
219 is difficult to determine but the characteristic pale-green color of broken ore (e.g., Fig. 2A), the  
220 ubiquitous presence of seemingly disseminated sulfides on planar surfaces down to cm-sized  
221 fragments and their connection to sulfide-filled centerlines and fractures in earlier quartz veins  
222 indicate that paint veins are in fact the predominant vein type in the ore shell in terms of vein  
223 surface per ore volume. The grade jumps at contacts of Young Tonalite indicate that they probably  
224 introduced most of the Cu-Fe-sulfide (chalcopyrite and bornite) present in the mineable ore.

225 Chlorite – white mica alteration with secondary albitic plagioclase is concentrated along C and  
226 paint veins (Fig. 3B, D and Fig. 4) and pervades the ore system, overprinting biotite – magnetite  
227 alteration (Fig. 1B and 4B). In the sample material used for this study hardly any completely  
228 unaltered biotite grain was observed, except in the barren core where paint veins appear to be  
229 less common. In addition to partial or complete replacement of biotite by chlorite and white mica,  
230 igneous Ca-rich plagioclase is altered to fine-grained intergrowths of white mica and calcite,  
231 whereas the Na-rich rims remain stable (Fig. 3H to J). Excess Ti and Ca, released from the  
232 breakdown of Ti-rich biotite and Ca-rich plagioclase, are fixed as rutile or anatase (Fig. 3K) and as  
233 calcite, epidote, apatite and anhydrite. In addition, titanite and barite are present, albeit rare, as

234 euhedral crystals in open spaces. Earlier magnetite was partially transformed to hematite along  
235 internal fractures and grain boundaries.

#### 236 *Pyrite-dominated veins and feldspar-destructive white mica – clay alteration*

237 Pyrite-dominated D veins are associated with distinct halos of white mica ± clay alteration  
238 characterized by pervasive feldspar destruction (Fig. 3A and F). These veins crosscut all previously  
239 formed vein types and can contain minor quartz and chalcopyrite in addition to pyrite. D-type  
240 veins occur in all rock types and, in contrast to most previous veins, invariably cut across the  
241 contacts to the Young Tonalite.

242 Clay alteration is characterized by the formation of smectite, white mica and minor chlorite  
243 after feldspar destruction along base metal veins filled with sphalerite, galena, tennantite, pyrite  
244 and minor amounts of chalcopyrite and bornite (Clode et al., 1999). Intense clay alteration only  
245 occurs in a limited zone at the eastern top of the deposit (Maryono et al., 2018) and was not  
246 studied here.

#### 247 *Metal distribution*

248 The ore body has the shape of an inverted cup with a barren core in the deep center  
249 surrounded by mineralized flanks (Fig. 1D and E). At the mine scale, the ore shell overlaps with  
250 the biotite – magnetite alteration zone (Clode et al., 1999; Arif and Baker, 2004), which is variably  
251 overprinted by chlorite – white mica alteration. At the resolution of mine blocks and blasthole  
252 assays the ore shell is strikingly regular with generally decreasing ore grades with distance from  
253 the intrusion center, excluding the weakly mineralized Young Tonalite. Sharply decreasing ore  
254 grades at the contact from the Old Tonalite to the Intermediate Tonalite, are locally documented  
255 by Setyandhaka et al. (2008), but are not generally recorded in the ore shell model of the  
256 operating mine and could not be confirmed in this study due to the scarcity of samples where the  
257 contact is clearly visible. Although Cu and Au both have similarly regular distribution patterns (Fig.  
258 1D and E), the Cu/Au ratio varies systematically within the deposit (Fig. 1B), generally decreasing  
259 with depth and laterally from the core to the periphery (Arif and Baker, 2004). The Young Tonalite  
260 has ~10 times lower metal grades (typically < 0.3 wt.% Cu; Fig. 1D) and a significantly higher Cu/Au  
261 ratio based on blasthole assays from the -30m level, documenting a weak late phase of  
262 dominantly Cu introduction or dispersion. In general, Ag contents are atypically high in Batu Hijau  
263 ore (up to 15 g/t) compared to other Au-rich porphyry deposits (<3 g/t; Sillitoe, 1997) and

264 correlate better with Cu grades than Au in the investigated drill holes. Economic amounts of  
265 molybdenum are absent in Batu Hijau and molybdenite was only encountered as small flakes  
266 within B-type quartz veins from the barren core.

267 Chalcopyrite and bornite are the main Cu-Fe sulfides, with minor amounts of digenite and  
268 chalcocite (Arif and Baker, 2004). Digenite and chalcocite intergrown with bornite is reported to  
269 only occur within A-type quartz veins and was therefore interpreted to represent an early low-  
270 sulfidation assemblage that was subsequently transformed to the higher sulfidation assemblages  
271 chalcopyrite – bornite and eventually chalcopyrite – pyrite (Clode et al., 1999). Native gold and  
272 electrum are typically attached to or enclosed in the Cu-Fe sulfides (Arif and Baker, 2004). Only a  
273 few  $\mu\text{g/g}$  of Au are present as homogenous solid solution within Cu-Fe sulfides (Kesler et al.,  
274 2002), which therefore cannot host the bulk of the Au ore grade. Cu-Fe sulfides are most  
275 conspicuous in quartz veins and Cu and Au grades are highest in areas of high quartz vein density,  
276 indicating that ore minerals are mostly hosted in A, AB and B veins (Clode et al., 1999; Arif and  
277 Baker, 2004; Setyandhaka et al., 2008). Sulfide grains in quartz veins are apparently randomly  
278 distributed, but even macroscopically are commonly seen to line up along small cracks, follow a  
279 centerline or cement open spaces between euhedral quartz crystals that grew into cavities. In the  
280 barren core, where vein densities are high, but Cu-Fe sulfides are either rare or absent, the lower  
281 ore grades do not correlate with the high vein density.

282

## Sampling and Methods

283 Due to the regular concentric vein and ore distribution at Batu Hijau, sampling was conducted  
284 largely on one N-S section (000) covering the extent of mineral zonation (Fig. 1). We thereby  
285 missed an area in the Western part of the deposit where Setyandhaka et al. (2008) found high-  
286 grade ore with early bornite in A-veins hosted by Old Tonalite and truncated by Intermediate  
287 Tonalite, which we acknowledge as a possible bias of the present study. Petrographic  
288 observations, secondary electron microscope – cathodoluminescence (SEM-CL) imaging, fluid  
289 inclusion microthermometry, and laser ablation – inductively coupled plasma – mass  
290 spectrometry (LA-ICP-MS) analysis of fluid inclusion assemblages were carried out at ETH Zürich,  
291 focusing on 19 samples from five boreholes. In total, 27 thin (30  $\mu\text{m}$ ) and 35 double-polished thick  
292 (300  $\mu\text{m}$ ) sections were prepared for petrographic work and analysis.

293 We used SEM-CL imaging and conventional petrography of polished thin sections to document  
294 the textural relationships and sulfide distribution relative to quartz, and to determine fluid  
295 inclusion assemblages (FIAs) within quartz veins. Several thousand SEM-CL images were obtained  
296 using a JEOL JSM 6390 LA Secondary Electron Microscope equipped with a Deben Centaurus CL  
297 detector at an acceleration voltage of 15-kV. Spot size, and hence energy density, was adjusted  
298 according to the CL-intensity of quartz in the different vein types. Images were taken with 80x to  
299 200x magnification, 60 s acquisition time and a resolution of 1280 x 960 pixels.

300 The same SEM device was used for spectral imaging and quantitative analysis of alteration  
301 minerals, using iSpectra (Liebske, 2015) to create phase maps based on X-ray spectral images  
302 obtained by energy-dispersive spectroscopy (EDS). Images were taken at 15-kV acceleration  
303 voltage, 20 minutes acquisition time, a magnification between 80x and 100x, and a resolution of  
304 256 x 192 or 512 x 384 pixels, resulting in pixel sizes of approximately 10 or 5  $\mu\text{m}$ . Quantitative  
305 EDS analyses of chlorite was conducted after calibration for Na, Mg, Al, Si, K, Ca, Ti, Mn and Fe at  
306 15-kV acceleration voltage and dead times between 25 and 30%. The reported compositions are  
307 average values obtained by 5 to 15 point measurements per individual chlorite crystal.

308 For microthermometric measurements, chips were cut out of double-polished thick sections,  
309 with 1878 fluid inclusions in 339 FIAs analyzed by a Linkam THMSG 600 heating-freezing stage.  
310 Heating and freezing experiments were conducted on FIA as defined by Goldstein and Reynolds  
311 (1994) to avoid unrepresentative results from fluid inclusions that entrapped mixtures of fluids or  
312 were significantly modified after their formation. Timing of fluid entrapment relative to the  
313 growth of the host mineral is based on petrographic criteria, following Goldstein and Reynolds  
314 (1994). Synthetic fluid inclusions of known compositions were used for calibration of the heating-  
315 freezing stage within  $\pm 0.3^\circ\text{C}$  at  $-56.6^\circ\text{C}$ ,  $\pm 0.05^\circ\text{C}$  at the melting point of ice and  $\pm 2^\circ\text{C}$  at  $374.1^\circ\text{C}$ ,  
316 the critical point of pure water. Freezing experiments were conducted first to avoid decrepitation  
317 of lower-temperature fluid inclusions during heating runs. Microthermometric results were  
318 processed with HokieFlinCs (Steele-MacInnis et al., 2012) based on Driesner and Heinrich (2007)  
319 to obtain minimum entrapment temperatures, salinities, and densities of entrapped fluids.

320 The chemical composition of 600 fluid inclusions from 105 FIAs were subsequently analyzed  
321 by LA-ICP-MS using the ETH-prototype Excimer 193 nm (ArF) laser ablation system coupled to a  
322 PerkinElmer NexION 2000, fast-scanning quadrupole ICP-MS. The sample introduction system,



323 including the in-house, fast-washout (ca. 6 cm<sup>3</sup>) glass ablation cell, was cleaned to minimize  
324 contamination, following the recommendations of Schläglová et al. (2017). We used a laser  
325 repetition rate of 10 Hz, energy density of ca. 15 to 20 J/cm<sup>2</sup> and spot size set as to entirely ablate  
326 the targeted inclusion (typically between 5 and 40 µm inclusion size at depths up to approximately  
327 100 µm). A progressive spot diameter increase up to 40 µm was applied using an iris aperture  
328 whenever necessary, to prevent quartz cracking upon ablation (Pettke et al., 2012). The detailed  
329 procedure is described by Günther et al. (1998), Audétat et al. (1998) and Heinrich et al. (2003).  
330 Element quantification was based on external calibration to the NIST SRM610 glass reference  
331 material (Jochum et al., 2011) and to the Sca-17 scapolite reference material for S, Cl, and Br (Seo  
332 et al., 2011), both ablated with laser spot diameters of 40 µm and energy densities of ca. 5 J/cm<sup>2</sup>.  
333 The SILLS software (Guillong et al., 2008a) was used for data reduction. Each time-integrated  
334 signal was first critically scrutinized for elemental peaks that clearly belong to the fluid inclusion  
335 signal, to avoid misinterpretation of the results with analytical artifacts such as spikes or matrix  
336 fluctuations. Absolute concentrations were calculated from measured element ratios using Na as  
337 internal standard, based on total salinities obtained by microthermometry (wt.% NaCl<sub>eq.</sub>)  
338 corrected for the effects of other salts (Heinrich et al., 2003; Steele-MacInnis et al., 2016). This  
339 procedure possibly introduces systematic errors, so that the background-corrected element  
340 ratios were preferred here for data interpretation (e.g., Seo et al., 2009; Schläglová, 2018). Ratios,  
341 concentrations, and their standard deviations (1σ) reported here are based on average values of  
342 typically 4-7 fluid inclusion analyses belonging to a common FIA. This procedure allows  
343 identification of outliers within a group of fluid inclusions that represent a common fluid  
344 according to petrographic observations and microthermometric measurements, and to assess  
345 analytical uncertainties including degrees of heterogeneous entrapment and post-entrapment  
346 modification (Pettke et al., 2012).

347 We also obtained trace element compositions of hydrothermal quartz by LA-ICP-MS, either  
348 directly from the matrix signals of fluid inclusion analyses (as implemented in SILLS; Guillong et  
349 al., 2008a) or from individual spot analyses along profiles through veins, using Si as internal  
350 standard for trace element quantification. In addition to elements monitored during fluid  
351 inclusion analysis, P, Ge and Ga were included in the elemental setup of trace element analysis of  
352 quartz.

353 Data obtained by microthermometry, LA-ICP-MS analysis of fluid inclusions and quartz as well  
354 as SEM analysis of chlorite grains is available as electronic repository.

## 355 Results

### 356 *Cathodoluminescence of hydrothermal veins*

357 Based on CL-intensities and the inferred relative chronology of the different vein types (table  
358 2), four major quartz generations,  $Q_1$  to  $Q_4$ , can be differentiated. In general, the brightness of CL  
359 images decreases from early A-vein quartz ( $Q_1$ ) over AB ( $Q_2$ ) and B ( $Q_3$ ) veins to almost non-  
360 luminescent paint-vein quartz ( $Q_4$ ). Anhedral early A- and AB-vein quartz commonly shows  
361 blurred zoning patterns, whereas euhedral quartz grains of B and paint veins show oscillatory  
362 growth zoning. Early A-type quartz locally contains rutile needles. The second generation of  
363 quartz ( $Q_2$ ), characteristic of AB veins, has a slightly weaker luminescence intensity than A-type  
364 quartz veins. The crystals are anhedral to subhedral and have diffusive or sharp growth zonation  
365 (Fig. 5A). The third generation of quartz ( $Q_3$ ), typical of B veins, is characterized by a medium gray  
366 luminescence and euhedral growth zoning (Fig. 5B). The intensity of gray luminescence can vary  
367 moderately between individual growth zones. Quartz crystals have commonly overgrown earlier  
368 generations of quartz (Fig. 5B). The fourth generation of quartz ( $Q_4$ ) has dull luminescence and  
369 occurs either as filling of a network of micro-cracks that follow and crosscut grain boundaries and  
370 growth zones of earlier generations, or as euhedral quartz grains growing into open spaces (Fig.  
371 5B-D, 6 and 7). The transition between  $Q_4$  and earlier  $Q_{1-3}$  is typically abrupt and irregular, caused  
372 by quartz dissolution preferentially along crystal boundaries and micro-fractures prior to  $Q_4$  re-  
373 precipitation. Locally, straight grain boundaries show that euhedral bright-luminescent quartz  
374 grains were overgrown by  $Q_4$  without intervening quartz dissolution.  $Q_4$  can contain fragments of  
375  $Q_{1-3}$  (e.g., Fig. 5A). Dull-luminescent  $Q_4$  occurs within paint veins, within earlier veins at  
376 intersections with paint veins (Fig. 5A) and along sulfide centerlines of B veins (Fig. 5B to D).

377 In all investigated samples, quartz vein-hosted sulfide grains, including bornite displaying  
378 digenite exsolution lamellae, are systematically associated with a fracture network filled by  $Q_4$   
379 (Fig. 6). Sulfide grains are aligned along growth zones in  $Q_4$  or were deposited over the newly  
380 grown  $Q_4$  grains (Fig. 7). Only a few bornite grains ( $\ll 1\%$  of all imaged sulfides) have been found  
381 without any contact to  $Q_4$  on the imaged surface (e.g., the small bornite grain in Fig. 6C) but even

382 these apparently isolated sulfide inclusions in earlier quartz generations appear to be associated  
383 with grain boundaries or cracks in the third dimension that are not visible by CL imaging of the  
384 sample surface. In addition to sulfides, Q<sub>4</sub> is spatially associated with, and contains inclusions of  
385 minerals typical for chlorite – white mica alteration associated with paint veins, i.e. chlorite, white  
386 mica, calcite, apatite, rutile, barite and anhydrite (Fig. 6, 7, 10C and 13). Based on luminescence,  
387 quartz from paint and D veins is not distinguishable and thus some of the quartz classified as Q<sub>4</sub>  
388 may have precipitated during D-vein formation.

#### 389 *Trace elements in hydrothermal quartz*

390 Trace elements were analyzed along profiles through veins containing all quartz generations  
391 (Fig. 7C). Commonly detected elements are Li, B, Na, Mg, Al, K, Ti, and Ge, while other elements  
392 such as Cl, Ca, Fe, Cu, Sr, and Ag were rarely above detection limit, which may relate to silicate,  
393 sulfide or fluid inclusions.

394 In general, concentrations of Li, B, Al, and Ti have the largest systematic variations between  
395 different quartz generations (Fig. 8 and 9, table 3), while other trace elements like Na, Mg, P, and  
396 K have no systematic differences. With up to 100 µg/g for Na and K, these trace elements are high  
397 in all quartz generations compared to most other trace elements. From Q<sub>1</sub> to Q<sub>4</sub> Li, B, and Al  
398 systematically increase, whereas Ti shows the opposite trend (Fig. 8 and 9). However,  
399 discrimination of the different vein types based on trace element compositions is not  
400 straightforward since most element concentrations overlap. Titanium concentrations are highest  
401 in Q<sub>1</sub> and Q<sub>2</sub> (50 to 150 µg/g), consistent with the presence of thin rutile needles. In contrast, the  
402 Li, B, and Al concentrations in Q<sub>1</sub> and Q<sub>2</sub> are the lowest, 1 µg/g, 0.5 to 1 µg/g, and 100 µg/g,  
403 respectively. In Q<sub>3</sub>, Ti contents (10 to 80 µg/g) are lower than Q<sub>1</sub> and Q<sub>2</sub>, whereas Li and Al  
404 concentrations are higher but still show a significant overlap (respectively 2 and 22 µg/g on  
405 average). The average Ti concentrations in Q<sub>4</sub> (ca. 9 µg/g, with some values < 1 µg/g) are the  
406 lowest among all quartz types, whereas Li, B, and Al contents are the highest, reaching  
407 concentrations of 10 µg/g, 5 µg/g and a few thousand µg/g, respectively. Due to the appearance  
408 of Q<sub>4</sub> as thin cracks, spot sizes overlapped with the surrounding, earlier quartz resulting in a  
409 mixture of trace elements from different quartz generations that resulted in a wide range of Li,  
410 Al, and Ti concentrations (Fig. 9). The outliers of low Ti concentrations of cracks filled by Q<sub>4</sub> are  
411 lower than Ti concentrations in quartz-dominated veins but overlap with the concentration of Ti

412 in paint veins, indicating that mainly Q<sub>4</sub> was analyzed during these measurements. The two  
413 outliers in AB veins with Ti concentrations below 10 µg/g, also reflect the presence of Q<sub>4</sub>,  
414 potentially due to ablation very close to euhedral dull-luminescent quartz, which could be  
415 dominant at depth.

#### 416 *Mineral thermometers*

417 Based on experiments above 600°C, Wark and Watson (2006) proposed that Ti concentrations  
418 in quartz correlate with its formation temperature and developed a thermometer. This  
419 thermometer was later recalibrated and corrected for the effect of pressure by Huang and  
420 Audédat (2012), who also warned of overestimating precipitation temperatures due to high and  
421 variable growth rates in hydrothermal environments. Based on the ubiquitous presence of rutile,  
422 the activity of Ti was set to 1 (Wark and Watson, 2006). Pressures varied between 0.75 and 0.2  
423 kbar depending on estimated depth and rock behavior during vein formation.

424 Applying the pressure-corrected formula of Huang and Audédat (2012), the Ti-in-quartz  
425 temperatures range from 700°C in Q<sub>1</sub> to ~ 330°C in Q<sub>4</sub>. In the barren core, high Ti concentrations  
426 in Q<sub>1</sub> from A veins yield temperatures between 620° to 700°C, comparable with Ti-in-quartz  
427 temperatures from early quartz veins in other porphyry systems (e.g., Rusk et al., 2008a; Müller  
428 et al., 2010). Based on lower Ti contents, Q<sub>2</sub> in AB veins from the mineralized ore zone formed  
429 between 550° and 650°C. Q<sub>3</sub> points to formation of B veins in a narrow temperature range  
430 between 410° and 500°C, in agreement with the ductile-brittle transition region for porphyry  
431 systems (Fournier, 1999). Low Ti concentrations in Q<sub>4</sub> indicate formation temperatures mostly  
432 between 330° and 380°C.

433 Chlorite composition has been proposed as a geothermometer (McDowell and Elders, 1980;  
434 Walshe, 1986; Cathelineau, 1988). We used the major element composition of hydrothermal  
435 chlorite from the ore zone at Batu Hijau to calculate the temperature range of chlorite – white  
436 mica alteration using the adjusted calibration of Jowett (1991), which considers the effect of Fe  
437 and Mg on the temperature dependency of incorporation of tetrahedrally coordinated aluminum.  
438 Following the suggestions of Inoue et al. (2010), chlorite with (CaO + Na<sub>2</sub>O + K<sub>2</sub>O) > 0.5 wt.% was  
439 regarded as contaminated with other mineral phases such as white mica or calcite and were  
440 therefore avoided. Most analyzed chlorites plot in the pycnochlorite and ripidolite fields (Hey,  
441 1954). Iron numbers (FeO/FeO+MgO) are between 0.35 and 0.60 for chlorite replacing biotite

442 along paint veins and between 0.60 and 0.85 for chlorite rosettes within dull-luminescent quartz  
443 (Fig. 10A). Specifically, the rims of chlorite rosettes are Fe-rich and plot close to the daphnite field  
444 (Fig. 10). Tetrahedral silicon varies from 5.36 to 5.82 a.p.f.u. Electron microprobe analyses of  
445 chlorites from Batu Hijau by Mitchell et al. (pers. commun., 1998) and Wilkinson et al. (2015)  
446 overlap with our data (Fig. 10). Calculated temperatures range from 290° to 380°C with an  
447 average of 340°C. Applying the T-R<sup>2+</sup>-Si diagram of Bourdelle and Cathelineau (2015), which is  
448 best at low temperatures (< 350 °C), low pressures (< 3-4 kbar) and assumes chlorite – quartz  
449 equilibrium (Inoue et al., 2009; Bourdelle et al., 2013), also yields mostly 300° to 350°C (Fig. 10B).

#### 450 *Fluid inclusion distribution and microthermometry*

451 Four main fluid inclusion types were distinguished at Batu Hijau based on phase proportions  
452 at room temperature: intermediate density (ID: Fig. 11A), brine (B: Fig. 11C to F), vapor (V: Fig.  
453 11B and F), and aqueous (AQ: Fig. 11G to J) fluid inclusions. Vapor, ID and AQ inclusions are two-  
454 phase fluid inclusions, distinguished by bubble sizes of 70-95 (V), 35-70 (ID) and 10-35 vol.% (AQ).  
455 Most ID and V inclusions have negative-crystal shapes, and many contain small opaque daughter  
456 crystals with tetragonal shapes, thought to be chalcopyrite. In contrast, AQ inclusions are typically  
457 more rounded or irregular in shape and rarely contain solids. Brine inclusions contain up to 7  
458 different crystals; cubic halite and opaque crystals are always present, and sylvite, hematite, and  
459 either anhydrite or calcite are common. Liquid CO<sub>2</sub> at room temperature and clathrate melting  
460 during freezing experiments was not observed in any fluid inclusion type.

461 Most FIA are of secondary origin, and any rare ID, B, and V inclusions on clear growth zones  
462 are mostly too small for further investigations ( $\leq 1 \mu\text{m}$ ) with a few exceptions (e.g., Fig. 11J).  
463 Microthermometry results are shown in Figure 12.

464 ID inclusions are dominant in A, AB, and B veins of the barren core of the deposit but are rare  
465 to absent at depths shallower than around 800 m below the pre-mining surface (Fig. 1C). They  
466 typically form secondary FIAs along fracture planes within Q<sub>1</sub> to Q<sub>3</sub> and homogenize by meniscus  
467 fading (critical behavior) at temperatures between 370° and 390°C in A-veins and 340° to 370°C  
468 in B-veins. Ice melting over a narrow temperature range between -2.4° to -2.1°C yields a constant  
469 salinity of around 3.7 wt.% NaCl<sub>eq.</sub> for all ID inclusions (Fig. 12). Calculated densities of ID  
470 inclusions range from 0.55 to 0.65 g/cm<sup>3</sup>.

471 The V and B inclusions dominate in vein quartz from the ore shell of the deposit (Fig. 1C). Due  
472 to the clustering of inclusions, distinguishing individual FIAs is difficult. All B and V assemblages  
473 are therefore characterized as secondary inclusions relative to their bright-luminescent host  
474 quartz (mainly Q<sub>2</sub> and Q<sub>3</sub>). The apparent disappearance of the last liquid in V inclusions is assumed  
475 to be generally well below homogenization temperature (Sterner, 1992). Final ice melting in V  
476 inclusions was only occasionally observable and yielded total salinities between < 1 to > 10 wt.%  
477 NaCl<sub>eq.</sub>

478 The B inclusions are separated into three subgroups. Subtype 1 (B<sub>1</sub>, Fig. 11C) is rare and  
479 dominantly occurs in the barren core, characterized by a small halite crystal (30 to 35 wt.% NaCl<sub>eq.</sub>)  
480 and a bubble of approximately the same size as the halite crystal (10 to 20 vol.%). Inclusions of  
481 this type always homogenize by bubble disappearance to the liquid at temperatures of ~ 320°C.  
482 Subtype 2 (B<sub>2</sub>, Fig. 11D) is more abundant and describes B inclusions with larger bubbles (20 to  
483 30 Vol.%) and higher salinities (35 to 45 wt.% NaCl<sub>eq.</sub>), usually with a small opaque crystal, thought  
484 to be chalcopryite, and rarely hematite, sylvite or anhydrite. Like subtype 1, homogenization is  
485 always to the liquid at temperatures between 350° and 500°C by bubble disappearance. Subtype  
486 3 (B<sub>3</sub>, Fig. 11E) describes B inclusions with up to seven crystals present and small bubble volumes  
487 (~ 20 vol.%). Apart from halite and chalcopryite that are identified in all inclusions, hematite,  
488 sylvite, and anhydrite are variably present. Ryan et al. (2001) identified other Ca, Sr, and Ba-  
489 bearing daughter crystals within poly-phase B inclusions from Batu Hijau using a nuclear  
490 microprobe. Inclusions of subtype 3 always homogenize by halite dissolution after bubble  
491 disappearance, yielding high salinities of > 50 wt.% NaCl<sub>eq.</sub>. The temperature difference between  
492 halite dissolution and bubble disappearance in B<sub>3</sub> inclusions is 65°C on average. Overall, the  
493 salinity measured in B inclusions and the amount of daughter crystals increase with decreasing  
494 depth, from 900 m (on average 34.2 wt.% NaCl<sub>eq.</sub>) to 300 m (on average 47.5 wt.% NaCl<sub>eq.</sub>) below  
495 the pre-mining surface (Fig. 1C). Fluid inclusions trails that host B and V inclusions (Fig. 11F) are  
496 common and called “boiling trails”.

497 The AQ inclusions are typically small (< 5 to 10 μm) and occur throughout the deposit (Fig. 1C).  
498 They are the only inclusion type that occur in Q<sub>4</sub>-filled cracks that cut earlier quartz veins and  
499 euhedral Q<sub>4</sub> grains that grew into open spaces (Fig. 13). In addition, rectangular AQ inclusions are  
500 also present in anhydrite within D-type veins (Fig. 11H). Homogenization temperatures, salinities,

501 and inclusion shapes allow the distinction of three inclusion subtypes: medium-salinity high-  
502 temperature (AQ<sub>1</sub>), low-salinity high-temperature (AQ<sub>2</sub>), and low-salinity low-temperature (AQ<sub>3</sub>).  
503 The AQ<sub>1</sub> inclusions are rare and undistinguishable from AQ<sub>2</sub> inclusions at room temperature.  
504 During freezing experiments, hydrohalite or ice melting at -25° to -17°C indicate salinities of 20 to  
505 25 wt.% NaCl<sub>eq.</sub> in AQ<sub>1</sub> inclusions. In AQ<sub>2</sub> inclusions, ice melting temperatures between -5.0° to -  
506 0.4°C indicate lower salinities of 0.7 to 8 wt.% NaCl<sub>eq.</sub>. Typically, AQ<sub>1</sub> inclusions occur as primary  
507 or pseudosecondary FIA within the cores of euhedral Q<sub>4</sub>, whereas AQ<sub>2</sub> inclusions are present as  
508 pseudo- or secondary trails within the outer rims of these crystals or Q<sub>4</sub>-filled cracks terminated  
509 by sulfide grains (Fig. 13). Homogenization temperatures for both subtypes AQ<sub>1</sub> and AQ<sub>2</sub> range  
510 from 290° to 390°C, with an average of 324°C. In contrast to the rounded-shaped AQ<sub>1</sub> and AQ<sub>2</sub> FI  
511 subtypes, AQ<sub>3</sub> inclusions are flat and irregular in quartz (Fig. 11I) or rectangular in anhydrite (Fig.  
512 11H) and usually occur within D veins. Ice melting temperatures vary between -3.0 to -1.0°C  
513 indicating salinities between 1 and 5 wt.% NaCl<sub>eq.</sub> with homogenization temperatures between  
514 230° and 300°C.

#### 515 *Chemical composition of inclusion fluids*

516 Elemental mass ratios to Na of analyses of 625 individual fluid inclusions within 107 FIAs are  
517 plotted as box plot diagrams for comparison between different fluid inclusion types (Fig. 14).  
518 Average concentration ratios, the second and third quartile (bottom and top of boxes, Fig. 14,  
519 respectively), and average concentrations of the different fluid inclusion types are reported in  
520 table 4. All values are based on averages of FIAs. Element ranges for all FIAs are interpreted to  
521 represent variations of the natural fluid compositions rather than analytical uncertainties.

522 In all types of fluid inclusions K, Fe, and S are the most abundant elements besides Na. Copper  
523 ratios to Na are high in ID, B, and V FIAs. However, Cu concentrations in ID and V inclusions are  
524 known to significantly exceed original Cu contents of the fluids due to post-entrapment  
525 modifications by coupled exchange of H<sup>+</sup> and Cu<sup>+</sup> between the inclusion and the ambient fluid,  
526 facilitated by excess S in the inclusions (Zajacz et al., 2009; Lerchbaumer and Audétat, 2012; Seo  
527 and Heinrich, 2013). Other elements are minor except for Mn with ratios relative to Na as high as  
528 0.69 in some B FIAs. In general, K/Na, Rb/Na, and Cs/Na are uniform among all fluid inclusion  
529 types (overall average 0.42, 0.02 and 0.0005, respectively), whereas most other elements for the  
530 ID, B, and V FIAs have different ratios relative to Na. Metal/Na mass ratios in brine inclusions for

531 example are higher than in vapor inclusions. Intermediate density and AQ FIAs overlap with  
532 respect to all element ratios except for Cu/Na.

533 The ID inclusions contain all elements detected in other fluid inclusion types, with the  
534 composition of ID FIAs having values intermediate between B and V FIAs compositions. The  
535 highest concentrations in ID inclusions are S and Cu, with average S/Na of 2.4 and Cu/Na of 1.2.  
536 Average ratios of K and Fe to Na are 0.25 and 0.3, respectively, making them the second most  
537 abundant elements found in ID inclusions, followed by minor concentrations of Li, B, Mg, Mn, Zn  
538 and Ba with ratios to Na below 0.1, and As, Rb, Sr, Mo, Ag and Pb with ratios to Na below 0.01.  
539 The least abundant, yet frequently detected element is Cs, with an average Cs/Na ratio of 0.0006.  
540 Gold was rarely above the detection limit and locally detected values are interpreted to be either  
541 unidentified analytical artifacts (spikes) or tiny gold particles within the ablated inclusions.

542 Element mass ratios to Na in B and V FIAs are generally distinct from ID-type fluid compositions  
543 with Li, B, Cu, As, and Ag enriched in V inclusions and Mn, Fe, Zn, and Pb enriched in B inclusions  
544 relative to Na (Fig. 14). Exceptions are K, Rb, Sr, and Cs ratios to Na, which are comparable in B  
545 and V inclusions and overlap with those in ID inclusions. Because base metals, except Cu, were  
546 not part of the elemental setup for V inclusion analysis, direct comparison with V inclusions is not  
547 possible. Concentrations of all other measured element ratios in V inclusions are comparable to  
548 those in ID fluid inclusions. Compositions of V inclusions can be contaminated by the accidental  
549 entrapment of disproportionate amounts of brine as the wetting phase (Bodnar et al., 1985).  
550 Therefore, mass balance calculations following the procedure described by Klemm et al. (2007)  
551 and Landtwing et al. (2010) were used to obtain the original vapor composition (Table 5).  
552 Corrected element to Na ratios for V inclusions are shown as smaller boxes partially overlapping  
553 with analyzed element ratios for V inclusions in Figure 14.

554 The AQ inclusion compositions overlap with the ID fluid inclusions in most element ratios. The  
555 exception is Cu/Na that, which is more than an order of magnitude lower (mostly between 0.01  
556 and 0.06) than in ID inclusions (mostly > 1). Although a small sulfur peak was commonly present  
557 in the time-integrated signals, usually S was below the detection limit because of the high and  
558 fluctuating S signal intensity during quartz ablation (Guillong et al., 2008b). AQ inclusions from D  
559 veins were too small to be analyzed.

560

## Discussion



561 Batu Hijau, with its apparently predominant biotite alteration and dense quartz veining with  
562 bornite, chalcopyrite and gold grains, has been described as a pristine porphyry deposit that was  
563 not widely overprinted by feldspar-destructive alteration postdating ore formation. Our  
564 observations confirm this for silicate alteration and quartz veining, but we find that metal  
565 precipitation was mostly associated with chlorite – white mica alteration and decoupled from  
566 most of the quartz deposition, and occurred at significantly lower temperature, as summarized in  
567 Figure 15. The following discussion aims at linking vein-scale textural evolution and deposit-scale  
568 zonation with successive fluid inclusion generations. We argue that the present-day make-up of  
569 the deposit was dominated by an evolving fluid evolution path across a steep gradient in pressure  
570 and temperature in the outermost parts of a magmatic fluid plume, sourced by a cooling,  
571 subjacent, fluid-producing magma reservoir. The strong gradients developed in response to the  
572 formation of an interface between the hot magmatic fluid plume and externally convecting  
573 meteoric water as suggested by the numerical modelling of Weis et al. (2012). As a result, the  
574 fluid evolution path changed through time (Fig. 16), from an early prograde stage establishing the  
575 magmatic fluid plume to a retrograde stage of gradual magmatic fluid plume retraction, during  
576 which much, if not most, of the metals were introduced.

577 *Vein-forming conditions and fluid evolution in space and time*

578 Reconstructing the conditions of hydrothermal quartz vein formation based on fluid inclusions  
579 requires estimating temperature as well as pressure. The physical state of the magmatic fluid  
580 provides an estimate of the minimum pressure within the barren core at the time of A vein  
581 formation. The solvus of the ID fluid considering the binary H<sub>2</sub>O-NaCl system at formation  
582 temperatures of early quartz veins (~ 500°C or more; Fig. 12, 15A) requires a minimum pressure  
583 of 0.69 kbar, corresponding to a depth of ca. 2.5 km based on lithostatic conditions, consistent  
584 with the partly ductile deformation of A veins (Fournier, 1999). Minor CO<sub>2</sub> of less than 2 mol%  
585 (the maximum based on lack of evidence for CO<sub>2</sub> phases in ID inclusions; Collins, 1979; Rosso and  
586 Bodnar, 1995) would shift the two-phase boundary to slightly higher pressures of approximately  
587 0.75 to 0.8 kbar (Gehrig et al., 1986). The depth estimate agrees well with the geologically  
588 reconstructed depth of 2 ± 0.5 km for the top of the Young Tonalite (Garwin, 2000), indicating

589 that the exposed orebody extended from  $\geq 1.5$  to  $\geq 2.5$  km below the paleosurface at the time of  
590 ore formation.

591 By contrast, the highest pressure indicated by boiling assemblages, i.e., fluid inclusions  
592 entrapped along the two-phase curve in the H<sub>2</sub>O-NaCl system model (Roedder and Bodnar, 1980),  
593 is  $\sim 0.42$  kbar in AB veins from the lower parts of the mineralized center (SBD 21 468m,  
594 approximately 500 m below the pre-mining surface), corresponding to a lower fluid pressure than  
595 lithostatic. Other boiling assemblages (excluding those homogenizing by halite disappearance,  
596 which probably suffered post-entrapment H<sub>2</sub>O loss; Audétat and Günther, 1999; Becker et al.,  
597 2008; Lecumberri-Sanchez et al., 2012) yield even lower pressures between 0.25 and 0.10 kbar  
598 (Fig. 12), indicating significant pressure variations during quartz vein formation, consistent with  
599 expansion and transition of a single-phase fluid initially at lithostatic conditions to a vapor phase  
600 condensing out a minor amount of brine at low pressures approaching hydrostatic or even  
601 vaporstatic conditions. Such a range of rather low fluid pressures is typical for Au-rich porphyry  
602 Cu deposits (e.g., Panguna: Eastoe, 1978; Grasberg: Weiland and Cloos, 1996; Far Southeast:  
603 Hedenquist et al., 1998; Bingham Canyon: Redmond et al., 2004; Landtwing et al., 2005).  
604 Murakami et al. (2010) and Landtwing et al. (2010) proposed that this observation indicates rapid  
605 decompression in a hot fluid plume at this stage of quartz vein evolution and suggest co-  
606 precipitation of both metals as a reason for the Au-rich nature of these porphyry Cu deposits.

607 *Quartz veins in the barren core* formed from a single-phase magmatic fluid as indicated by ID  
608 inclusions as the dominant inclusion type (Fig. 1C). Pressure-corrected homogenization  
609 temperatures suggest formation temperatures for A veins of approximately 450-510°C (Fig. 12).  
610 Brine assemblages in A veins from the barren core are rare, show the lowest homogenization  
611 temperatures of this inclusion type (300-330°C) and do not occur as boiling pairs (Fig. 12). Due to  
612 the steepness of brine isochores, any pressure correction is small so that these brines must have  
613 been significantly cooler than the earlier ID fluids in A veins of the barren core ( $< 370^\circ\text{C}$  at 0.7 kbar  
614 but more likely  $< 340^\circ\text{C}$  at a hydrostatic pressure of  $\sim 0.3$  kbar). Likewise, the straight B veins in  
615 the barren core, suggesting more brittle rock conditions, formed close to hydrostatic pressures,  
616 and pressure-corrected homogenization temperatures of ID inclusions are between  $390^\circ$  and  
617  $460^\circ\text{C}$ . In summary, the fluid inclusions of the barren core show most clearly that successive fluids

618 record decreasing pressures from lithostatic to hydrostatic pressures and thereby cooled from >  
619 510°C to ~ 300°C.

620 *Quartz veins in the ore shell* formed during phase separation of the ascending magmatic fluid,  
621 as documented by boiling trails in AB and B veins. Co-existence of hypersaline liquid and low-  
622 density vapor implies entrapment along the two-phase boundary, which means that measured  
623 homogenization temperature and pressure are equal to the original entrapment conditions  
624 (Roedder and Bodnar, 1980). In the mineralized center, these assemblages record mostly higher  
625 temperature and higher salinity in the brines, implying a larger degree of phase separation  
626 compared to boiling trails in the mineralized flanks (Fig. 12). Mass balance based on conservation  
627 of H<sub>2</sub>O and NaCl shows that a ID fluid that initially contained 4 wt.% NaCl can split into vapor with  
628 1 wt.% NaCl and brine with 40 wt.% NaCl at a weight proportion of approximately 92% to 8%  
629 (Table 5), showing that the vapor phase dominated by weight (and even more so by volume)  
630 during AB and B vein formation, as argued for other shallow porphyry systems (Henley and  
631 McNabb, 1978; Hedenquist and Lowenstern, 1994; Hedenquist et al., 1998; Williams-Jones and  
632 Heinrich, 2005; Seo et al., 2009; Landtwing et al., 2010; Weis et al., 2012). The assumed salinity  
633 of the vapor phase co-existing with a hypersaline liquid was calculated using the SoWat software  
634 (Driesner and Heinrich, 2007) and corresponds to the maximum salinity expected upon phase  
635 separation at pressures of 0.15 to 0.25 kbar. Higher salinities of up to 10 wt.% NaCl<sub>eq.</sub> in V FIAs  
636 are probably the result of small amounts of co-existing hypersaline liquid that was coincidentally  
637 entrapped resulting in erroneously high salinities (Roedder, 1984; Bodnar et al., 1985). Vapor  
638 dominance is further supported by the chemical composition of the vapor, which is similar to that  
639 of the intermediate density fluid implying condensation of a minor amount of brine from the  
640 dominating vapor. Highest temperatures obtained from boiling assemblages in AB veins are 530  
641 ± 10°C, while the lower range overlaps with those of B veins at 410 to 360°C (Fig. 12). Similar  
642 formation temperatures of both vein types might derive from secondary boiling assemblages  
643 trapped in AB veins during B-vein formation. The Ti-in-quartz thermometer yields slightly higher  
644 formation temperatures for Q<sub>2</sub> and Q<sub>3</sub> between 600° to 500°C and 450° to 400°C, respectively.  
645 Entrapment pressures from boiling assemblages in B veins vary between 0.13 and 0.18 kbar,  
646 indicating a hydrostatic to vapor-static pressure regime (Fournier, 1999). Subsequent cooling of  
647 the vapor phase below ~400°C led to a continuous increase of its density and resulted in partial

648 dissolution of previously formed quartz veins (Fournier, 1983), creating secondary pore space for  
649 subsequent fluid flow.

650 *Paint vein formation and Cu-Fe sulfide precipitation* occurred during and after partial re-  
651 dissolution of the dominant vein quartz at any one point in the deposit. Minor Q<sub>4</sub> quartz  
652 associated with paint veins and filling micro-fractures in earlier quartz veins exclusively host  
653 single-phase AQ inclusions (e.g., Fig. 13). Overlap between homogenization temperatures of 310  
654 - 360° (Fig. 12), chlorite and Ti-in-quartz thermometry (290 - 410°C, Fig. 10) and the destruction  
655 of biotite to muscovite + chlorite indicate that the AQ FIs associated with paint vein formation in  
656 this temperature range require no significant pressure correction. The indicated temperature  
657 range also agrees well with the low concentrations of Au within bornite and chalcopyrite,  
658 suggesting precipitation temperatures of approximately 300°C at Batu Hijau (Simon et al., 2000;  
659 Kesler et al., 2002) and includes the pressure-temperature window of retrograde quartz solubility  
660 (400° to 350°C at 0.2 kbar; Fournier, 1999; Monecke et al., 2018) that precedes sulfide  
661 precipitation. The salinity of the AQ inclusions overlaps with the single-phase ID inclusions (3.7 ±  
662 0.5 wt% NaCl<sub>eq</sub>) but extends to notably lower as well as higher salinities (0.5 - 23.5 wt% NaCl<sub>eq</sub>;  
663 Fig. 12). This large salinity range of the AQ fluid inclusions contrasts with most element ratios that  
664 largely overlap with those of the ID fluids of the barren core within less than one order of  
665 magnitude, with the exception of the significantly lower Cu content of the paint vein inclusions  
666 (Fig. 14). We therefore suggest that the paint vein-forming AQ fluid was derived from re-  
667 homogenization of brine and vapor phases from which the earlier quartz-dominated vein types  
668 formed, as explained in the next subsection. The intermediate saline AQ<sub>1</sub> inclusions can occur as  
669 primary FIAs in the cores of euhedral Q<sub>4</sub> (e.g., inset of Fig. 13 referring to Fig. 11J), indicating that  
670 Q<sub>4</sub> precipitation was initiated with the re-homogenization of brine with increasing amounts of  
671 contracting vapor.

672 *The formation of D veins* mostly post-dates the emplacement of the Young Tonalite (Fig. 15).  
673 Quartz- and anhydrite-hosted AQ FIAs indicate temperatures between 300° and 220°C (Fig. 12).  
674 The transition from paint to D veins might have been gradual based on the alteration mineralogy  
675 associated with these veins that progresses from chlorite – white mica over white mica – chlorite  
676 to feldspar-destructive white mica – clay alteration, suggesting increasingly reactive fluids with  
677 decreasing temperatures.

678 *Changing fluid evolution during a downward-retracting mineralization front*

679 On the scale of individual quartz veins, precipitation of Cu-Fe sulfides post-dates the main  
680 quartz vein event (Fig. 5, 6, 7 and 13). However, on the deposit scale, hydrothermal quartz and  
681 sulfides both precipitated from a common magmatic fluid, mostly at overlapping times but  
682 spatially separated under different physical conditions in different parts of the system. The main  
683 mass of vein quartz formed at higher temperatures, within the core of the magmatic fluid plume,  
684 while sulfides predominantly precipitated in the peripheral and shallower parts of the magmatic  
685 fluid plume where steep temperature and pressure gradients occurred in response to cooling  
686 effects of convecting meteoric fluids (Weis et al., 2012). As the magmatic fluid production  
687 gradually decreased with the progressive cooling of the magma reservoir, the magmatic fluid  
688 plume gradually retracted and, accordingly, zones of sulfide precipitation shifted in- and  
689 downward and overprinted the previously precipitated high-temperature quartz veins. This mine-  
690 scale evolution of the hydrothermal system (Fig. 16) can therefore be explained by three stages:  
691 1) A prograde stage in which volcanic rocks are heated by a magmatic fluid plume until the  
692 maximum temperature and spatial extent of the hydrothermal system is reached; 2) a retrograde  
693 stage of sustained fluid flow through inward and downward retracting mineral precipitation  
694 fronts; and 3) a waning stage marked by an evolution path of magmatic fluids at overall lower  
695 temperatures at all depths.

696 *The prograde stage* (Fig. 16A) is characterized by the outward expansion of isotherms as the  
697 surrounding rocks are heated up by the porphyry intrusion and by the displacement of cold  
698 meteoric water by a focused magmatic fluid plume. Initial vein and alteration types form  
699 depending on P-T conditions. A-type quartz veins and biotite – magnetite alteration develop in  
700 the region of the present ore deposit around the top of the tonalite complex (~ 2 km paleodepth).  
701 Early bornite within A-type veins, as described by Clode et al. (1999), may precipitate due to rapid  
702 cooling of hot fluid against initially cold rock (e.g., McInnes et al., 2005). The observation by  
703 Setyandhaka et al. (2008) that such potentially early, high-grade bornite ore is mainly restricted  
704 to marginal relicts of Old Tonalite, supports such a quenching mechanism (Proffett, pers.  
705 commun., 2020). However, due to the high specific heat content of the fluid phase in combination  
706 with the focused fluid flux, the host rock heats up quickly to reach thermal equilibrium with the  
707 fluid phase, which may prevent further sulfide precipitation from the high-temperature magmatic

708 fluid. The total quantity of this early bornite mineralization is unknown as textural studies with  
709 CL-petrography are still lacking but likely to be minor, even though it may be of high grade locally.  
710 Once the host rock has been heated up to > 500°C, quartz may still precipitate due to gradual  
711 cooling and phase separation of the ascending magmatic fluid, contributing to A and AB vein  
712 formation. At near-hydrostatic pressures and temperatures of approximately 0.25 kbar and  
713 450°C, phase separation produces a low-density vapor and a highly saline brine phase close to the  
714 vapor-liquid-halite surface in the salt – water system, which could explain the generally higher  
715 salinity B FIAs within shallower quartz veins (Fig. 1C; Fig. 12). However, most of the contained Cu,  
716 Fe and S was dispersed into the low-grade halo around the present deposit, where these  
717 components may have partly precipitated in early paint veins.

718 *The retrograde stage* dominated the development of the present-day ore shell (Fig. 16B) and  
719 reflects the downward retreat of isotherms due to overall cooling of the subjacent intrusive  
720 complex, leading to decreasing magmatic fluid production at progressively greater depth  
721 (Williams-Jones and Heinrich et al., 2005; Lamy-Chappuis et al, 2020). The resulting gradual  
722 thermal retraction of the hydrothermal system ('telescoping') could be triggered by syn-  
723 hydrothermal erosion or collapse of an overlying volcanic edifice (Sillitoe, 1994), but is a natural  
724 consequence of the generation of a stable interface between the hot magmatic fluid plume and  
725 hydrologically separated ground water convection cells (Fournier, 1999; Weis et al., 2012; Weis,  
726 2015). During this hydrologically stabilized stage isotherms moved inward only slowly while the  
727 bulk of available magmatic fluid evolved along a path shown in Figure 16B, based on the inclusion  
728 record summarized in the lower part of Figure 15. Except for somewhat reduced brine salinity  
729 produced by phase separation at greater pressure, this fluid evolution path is the same as in the  
730 prograde stage (Fig. 16A). In the region of the hot barren core, ID fluids injected from below  
731 separated into brine and vapor, contributing additional barren A and AB type quartz. Although  
732 brine and vapor are both expelled from the lithostatically-pressured core and flow outward  
733 together, we speculate that the vastly different wetting properties and viscosities of the two fluid  
734 phases and the locally different but fast flow rates in narrow veins could prevent the two fluids  
735 from staying in chemical equilibrium at all times and locations in the complex vein network. As  
736 sulfur species (H<sub>2</sub>S, SO<sub>2</sub>) are preferentially enriched in the vapor while metals (Cu, Fe) are mainly  
737 transported in the brine (e.g., Drummond and Ohmoto, 1985; Pokrovski et al., 2005; Lerchbaumer

738 and Audétat, 2012; Zajacz et al., 2017), any small-scale isolation of the fluids will inhibit  
739 precipitation of Cu-Fe-sulfides and thus help maintaining high concentrations of these  
740 components in the respective fluids. As the two fluids flow outward into the hydrological contact  
741 region, they both cool below the critical temperature at hydrostatic pressures ( $\lesssim 370^{\circ}\text{C}$  at  $\sim$   
742 0.2kbar, Fig. 16B). As a result, the initially immiscible vapor contracts to a low-salinity aqueous  
743 liquid (Heinrich, 2005; Driesner and Heinrich, 2007) and becomes miscible again with the brine.  
744 This re-homogenization can explain the observation that the dominant aqueous fluid inclusions  
745 in paint veins have overall similar salinity and metal ratios as the original magmatic intermediate-  
746 density fluid inclusions (see Fig. 12 and 14), but with a greater salinity spread towards either side  
747 of 4 wt.%  $\text{NaCl}_{\text{eq.}}$ , i.e. FIAs with lower salinity would be dominated by contracted vapor, whereas  
748 those with higher salinities up to 25% wt.%  $\text{NaCl}_{\text{eq.}}$  originated by a locally greater admixture of  
749 brine. Transient physical isolation of brine and vapor in the barren core, followed by re-  
750 homogenization at lower temperature, will lead to re-combination of reduced sulfur and ore  
751 metals, which may explain the precipitation of most sulfides in paint veins and related cracks  
752 cutting earlier quartz stockwork veins. This idea, although difficult to prove quantitatively, can  
753 explain why Cu deposition was hampered during quartz vein formation and why it mainly  
754 precipitated at relatively low temperatures from a low-salinity, presumably  $\text{H}_2\text{S}$ -rich fluid. The  
755 range of Cu concentrations analyzed in AQ FIAs ( $\sim 0.01$  to 0.21 wt.%) are in good agreement with  
756 the modelled compositional range of primary fluids exsolved from calc-alkaline magmas (0.02 to  
757 0.21 wt.% Cu, Chelle-Michou et al., 2017), concentrations of presumably non-modified ID-type  
758 fluid inclusions (0.01 to 0.05 wt.% Cu, Fiedrich et al., 2020) and overlap with the lowest  
759 concentrations found in ID FIAs from Batu Hijau (Fig. 14), indicating that no significant loss of Cu  
760 from the fluid occurred during its previous evolution. This is in line with the textural observation  
761 that many AQ FIAs appear to immediately pre-date Cu-Fe sulfide precipitation (Fig. 13). The  
762 limiting element for ore formation was Cu, as both, Fe and S, are present in higher quantities (up  
763 to 2 wt.%, respectively in AQ FIAs, although S was rarely quantifiable). Chemical transport and  
764 mineral precipitation is dominated by magmatic fluids, but in the absence of stable isotope data  
765 for Batu Hijau, minor input of low-salinity meteoric liquid cannot be excluded (cf. Fekete et al.,  
766 2018).

767        *The waning stage* (Fig. 16C) is marked by a fluid evolution path through an overall cooler  
768 system where the ductile to brittle transition shifted to even greater depth. Within the barren  
769 core at ~ 3 km paleo-depth, phase separation was suppressed at lower temperatures (< 450°C).  
770 The general absence of B and V inclusions in B veins within the barren core, but dominance of ID  
771 inclusions, indicate B vein formation from a single-phase fluid. The straight-walled B veins suggest  
772 formation during the waning stage under brittle conditions with no ductile deformation after  
773 emplacement. Compositionally, the B vein-hosted ID fluid inclusions are identical to those from  
774 earlier A-type veins, except for lower K/Na ratios. The similarity in fluid composition and the  
775 continuous fractionation of the inferred source magma indicated by falling temperature and  
776 evolving trace element composition of progressively younger zircons from the Old Tonalite to the  
777 Young Tonalite (Large et al., 2020) exclude major changes in the composition of the degassing  
778 magma chamber and the input fluid. Retrograde quartz dissolution did not occur anymore at the  
779 present mine level and dull-luminescent precipitated directly from an intermediate density fluid  
780 upon cooling, as indicated by continuous growth zoning from euhedral Q<sub>3</sub> to oscillatory zoned Q<sub>4</sub>  
781 (Fig. 13). Some chalcopyrite probably still precipitates during the waning stage but the major part  
782 of the present-day ore shell was completed during the retrograde stage.

783        *The relative importance of the three stages* for mineral precipitation cannot be quantified  
784 because of the difficulty of assigning veins to the Old and Intermediate Tonalite and the likelihood  
785 that veins of the older intrusion were reworked by the later intrusion and its fluid plume. We also  
786 do not know whether and to what degree a cycle of these stages already occurred between the  
787 emplacement of the Old and Intermediate Tonalite (Fig. 15, top). Nevertheless, the ubiquitous  
788 observation of quartz dissolution and re-precipitation prior to sulfide precipitation suggests that  
789 a greater proportion of vein quartz in the ore shell was formed in the prograde stage, whereas  
790 the bulk of the economic sulfide ore surrounding the present barren core formed during the  
791 retrograde stage of the hydrothermal system. This mainly occurred after emplacement of the  
792 Intermediate Tonalite but before the Young Tonalite, which was followed by a much weaker cycle  
793 of sparse A-type and paint veins.

794        *The duration and total fluid quantities* involved in these stages can be estimated from the rates  
795 of fluid production and expulsion simulated numerically for a magma chamber of suitable size for  
796 the formation of Batu Hijau. Assuming a parental magma containing 5 wt.% H<sub>2</sub>O and an extraction



797 efficiency of ~ 50% (Chelle-Michou et al., 2017) of an ID fluid with ~ 2000 µg/g Cu (~ maximum Cu  
798 concentration detected in AQ FIAs), about 40 km<sup>3</sup> of source magma are required to generate Batu  
799 Hijau. These dimensions are similar to parameters used by Weis (2015) to model fluid flow  
800 through a generic porphyry deposit and by Lamy-Chappuis et al. (2020) to simulate its extraction  
801 rate from an upper-crustal magma chamber. The latter study predicts ~ 80 ky of focused fluid flow  
802 collected from an upper-crustal intrusion of this size. During the first ~ 20 ky the model predicts  
803 an advance of the 350 – 450°C isotherms, followed by more than 60 ky of steady state and slowly  
804 retracting isotherms while the largest fraction of available fluid is expelled. The total duration of  
805 this magma-chamber process is similar to the duration of mineralization bracketed by zircon  
806 dating of the successive tonalites at Batu Hijau, to have been completed within ~ 90 ky or less  
807 (Large et al., 2020). These considerations are consistent with our interpretation that the  
808 formation of retrograde paint veins have introduced the dominant fraction of metals at Batu  
809 Hijau.

### 810 **Comparison with and implications for other porphyry Cu systems**

811 Textural evidence for sulfide precipitation after quartz vein and veinlets similar to the paint  
812 veins noted in this study are widely reported from other porphyry systems. Although variations  
813 exists, similar veins are described as pale-green sericitic veins (Butte; Meyer, 1965; Rusk et al.,  
814 2008b), bornite-chalcopryrite-quartz-anhydrite veinlets with chlorite-sericite alteration halos (Los  
815 Pelambres; Sillitoe, 1973), discontinuous sulfide veinlets (Panguna; Eastoe, 1978), ore-bearing  
816 veins (Santa Rita; Reynolds and Beane, 1985), C-type veins (El Salvador; Gustafson and Quiroga,  
817 1995; Batu Hijau; Arif and Baker, 2004), copper-gold veins (Grasberg; Pollard and Taylor, 2002),  
818 transitional to late chalcopryrite veins (Bajo de la Alumbreira; Proffett, 2003), type 2 chlorite  
819 veinlets (El Teniente; Cannell et al., 2005; Vry et al., 2010), sulfide-only veins (Bingham Canyon;  
820 Porter et al., 2012), or chalcopryrite hair-veinlets with chlorite selvages (Nuevo Chaquiro, Bartos  
821 et al., 2017). Based on sulfide-hosting vein types, two mineralization events are described for  
822 some deposits, including high-temperature sulfide precipitation during quartz vein formation  
823 followed by low-temperature sulfides along thin sulfide veinlets with chlorite alteration halos  
824 (e.g., Panguna, Eastoe, 1978; Nuevo Chaquiro, Bartos et al., 2017), whereby some paint veins  
825 crosscutting late-mineral intrusions were interpreted to indicate local metal remobilization  
826 (Proffett, 2003). For other deposits like Sierrita-Esperanza (Preece and Beane, 1982), Santa Rita

827 (Reynolds and Beane, 1985), Far Southeast (Hedenquist et al., 1998) and Escondida (Garza et al.,  
828 2001) most of the ore was interpreted to post-date high-temperature quartz vein formation  
829 associated with potassic alteration. Detailed fluid inclusion studies at Bingham Canyon (Landwing  
830 et al., 2010) and Grasberg (Mernagh et al., 2020) emphasize an alternative process of fluid  
831 expansion based on brine + vapor inclusions in quartz veins, arguing for gold + sulfide  
832 precipitation due to low-pressure metal de-complexation (cf. Migdisov et al., 2014). But even at  
833 Bingham Canyon (Landtwing et al., 2005, 2010), CL imaging of quartz stockwork veins shows a  
834 clear textural relationship between sulfides and a dull-luminescent quartz generation post-dating  
835 the main quartz veining event, similar to other porphyry systems (e.g., Rusk and Reed, 2002;  
836 Redmond et al., 2004; Klemm et al., 2007; Müller et al., 2010; Vry et al., 2010; Stefanova et al.,  
837 2014; Gregory, 2017).

838 A comparable mineralizing fluid to the one proposed here for Batu Hijau is reported from Far  
839 Southeast in the Philippines (Hedenquist et al., 1998) and El Teniente in Chile (Klemm et al., 2007).  
840 For Far Southeast, fluid inclusion and stable isotopic constraints indicated that a single-phase  
841 magmatic fluid separated into brine and vapor at paleo-depths below ~2 km (Shinohara and  
842 Hedenquist, 1997; Hedenquist et al., 1998). The early hypersaline liquid precipitated quartz veins  
843 followed by later sulfide precipitation from a low-salinity aqueous fluid during illite ± chlorite  
844 alteration (Hedenquist et al., 1998). These authors suggest sequential events of fluid injection  
845 with changing input compositions to explain the successive vein and alteration types. Similarly,  
846 for El Teniente, Klemm et al. (2007) proposed that quartz veins and sulfides have been formed  
847 from a common fluid but at different stages. Klemm et al. (2007) interpreted the Cu-Fe sulfides  
848 to be genetically linked to both, a vapor phase and its contracted single-phase low-salinity liquid  
849 derivative. For both deposits, the estimated temperature ranges of sulfide precipitation are  
850 comparable, 350°C in Far Southeast (Hedenquist et al., 1998) and 410° to 320°C in El Teniente  
851 (Klemm et al., 2007), and overlap with our results for Batu Hijau. However, the interpretation that  
852 quartz vein formation is temporally decoupled from sulfide precipitation on the scale of the  
853 hydrothermal system would imply that Cu contained in the early quartz-precipitating fluids is lost  
854 from the ore body and dispersed – in contrast to the dynamic spatial-temporal evolution of a  
855 single magmatic fluid proposed here for the retrograde stage at Batu Hijau.

856 At the Santa Rita deposit in New Mexico, Reynolds and Beane (1985) concluded that hypogene  
857 mineralization post-dates high-temperature quartz stockwork vein formation and associated  
858 potassic alteration and was formed from a low-salinity (< 9 wt.% NaCl<sub>eq.</sub>) aqueous-like fluid at  
859 temperatures below 360°C. In contrast to Batu Hijau the mineralizing fluid was assumed to be  
860 dominantly of meteoric origin (Reynolds and Beane, 1985). Based on similar bulk chemical  
861 compositions of low salinity inclusions in early quartz and late pyrite veins, Rusk et al. (2008b)  
862 proposed that variable hydrothermal features are the result of changing fluid evolution paths of  
863 a single magma-derived fluid at Butte, Montana. Geochemical modelling of fluid-rock interactions  
864 demonstrated that a common magmatic fluid undergoing various cooling and depressurization  
865 paths could indeed be responsible for variable vein types and alteration mineral assemblages  
866 (Reed et al., 2013).

867 Many features of hypogene mineralization at Batu Hijau are common in descriptions of  
868 porphyry Cu systems worldwide, such as: the relative timing after the formation of quartz veins;  
869 the association with a dull-luminescent quartz generation and pale-green chlorite – white mica  
870 alteration along thin sulfide veinlets; and sulfide precipitation at moderate temperatures  
871 between 420° and 300°C. Consequently, the fluid evolution at Batu Hijau (Figs. 14 and 15) appears  
872 to be common for hypogene ore formation in porphyry systems, even though the physico-  
873 chemical mechanisms driving ore mineral precipitation are still debated and may vary between  
874 deposits (e.g., Williams-Jones and Heinrich, 2005; Kouzmanov and Pokrovski 2012; Migdisov et  
875 al. 2014; Henley and Seward, 2018). .

## 876 **Conclusions**

877 Hypogene ore minerals hosted within quartz stockwork veins at Batu Hijau comprise  
878 chalcopyrite, lesser bornite with minor amounts of digenite and gold in separate but co-  
879 precipitated grains. Textural relationships documented by CL imaging show that ore mineral  
880 precipitation, although spatially associated with quartz veins, temporally post-dates the main  
881 event of quartz deposition at any given point within the hydrothermal system and is related to  
882 superimposed paint veins associated with chlorite – white mica alteration. Thus, most, if not all,  
883 of the studied sulfides that macroscopically appear to be hosted by high-temperature quartz  
884 stockwork veins were introduced during the subsequent formation of paint veins.

885 Aqueous FIAs hosted within the mineralized quartz generation as well as chlorite and Ti-in-  
886 quartz thermometry suggest a narrow temperature window of sulfide precipitation between 360°  
887 and 310°C. The similar chemical composition of the high-temperature magmatic and the low-  
888 temperature mineralizing fluid implies that vein and alteration types are the result of  
889 progressively changing fluid evolution paths due to cooling and depressurization. Based on  
890 crosscutting relationships of different vein types, overprinting alteration mineral assemblages  
891 and the distribution of fluid inclusion types, we propose an evolution of the hydrothermal system  
892 at Batu Hijau that started with quartz vein formation in the high-temperature central parts (>  
893 500°C) coeval with sub-economic sulfide precipitation in the low-temperature periphery (<  
894 400°C). Inward-retreating isotherms led to telescoping of alteration styles, crosscutting of  
895 different vein types and most of the sulfide deposition after partial quartz dissolution within  
896 earlier quartz stockwork veins. Sulfides precipitated from a low-salinity single-phase aqueous  
897 fluid (~ 330°C, 1 to 8 wt.% NaCl<sub>eq.</sub>, 730 µg/g Cu on average) derived by re-homogenization of brine  
898 and vapor that were produced episodically by phase separation of the parental magmatic fluid on  
899 the preceding depressurization path, which thereby precipitated most of the locally predating  
900 vein quartz.

901 Although evidence for high-temperature sulfide precipitation has repeatedly been reported on  
902 the basis of macroscopic observations and might be of importance for Cu endowment in some  
903 porphyry systems, the CL observations from Batu Hijau — previously thought to be a dominantly  
904 high-temperature Cu-Au deposit — should be taken as a word of caution that macroscopic  
905 observations alone can be deceptive. Of the two endmember processes of Cu-Fe sulfide  
906 introduction and precipitation that contribute to porphyry Cu mineralization, the low-  
907 temperature process associated with inconspicuous chlorite – white mica-lined paint veins is  
908 commonly underestimated in our opinion. Phase separation of a single-phase hot magmatic fluid  
909 into dominant vapor and minor brine, followed by re-homogenization of these two fluids to an  
910 aqueous liquid of variably low salinity resembling the composition of the initial input fluid, could  
911 contribute to a temperature gap between the two endmember processes of Cu-Fe-sulfide  
912 precipitation.

913

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924

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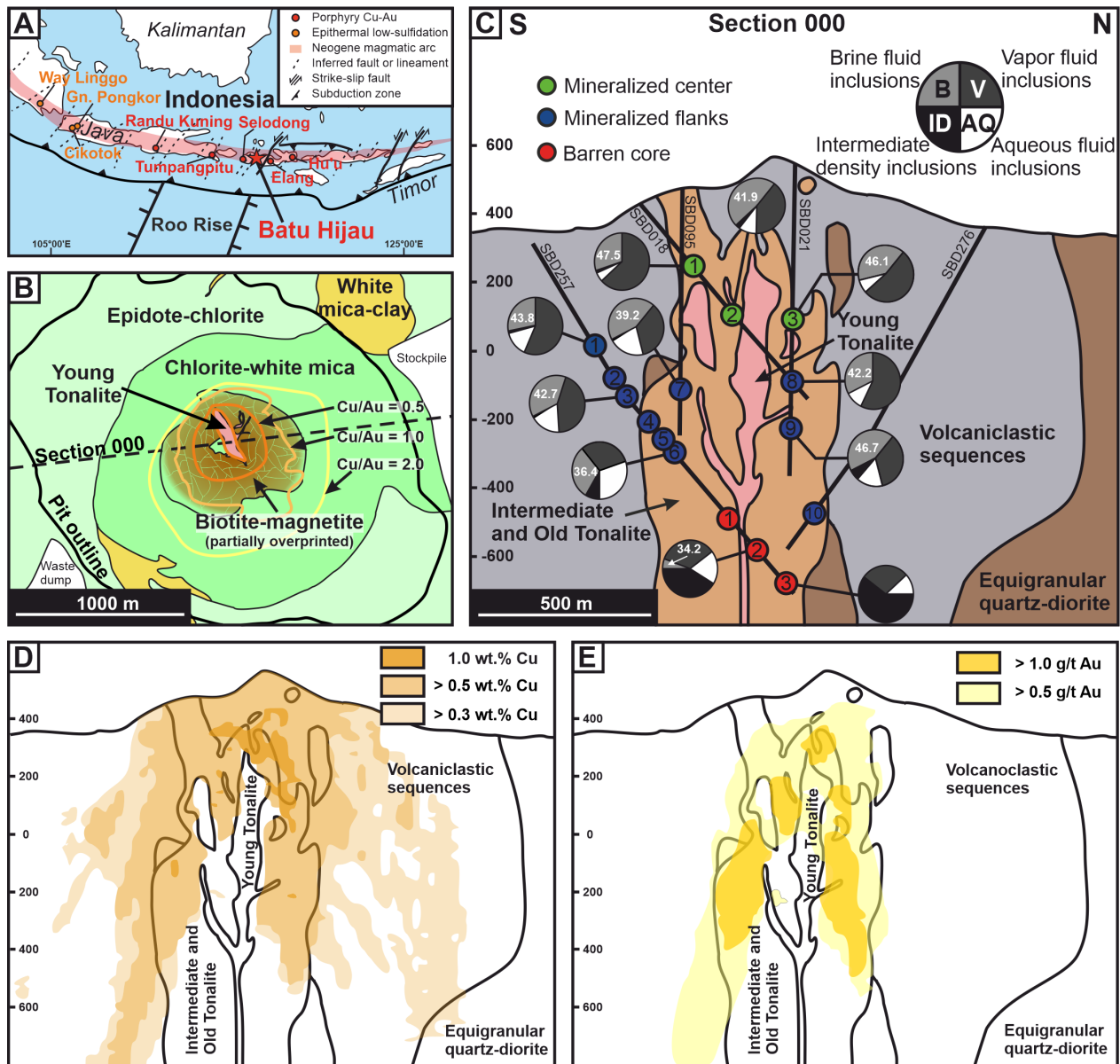
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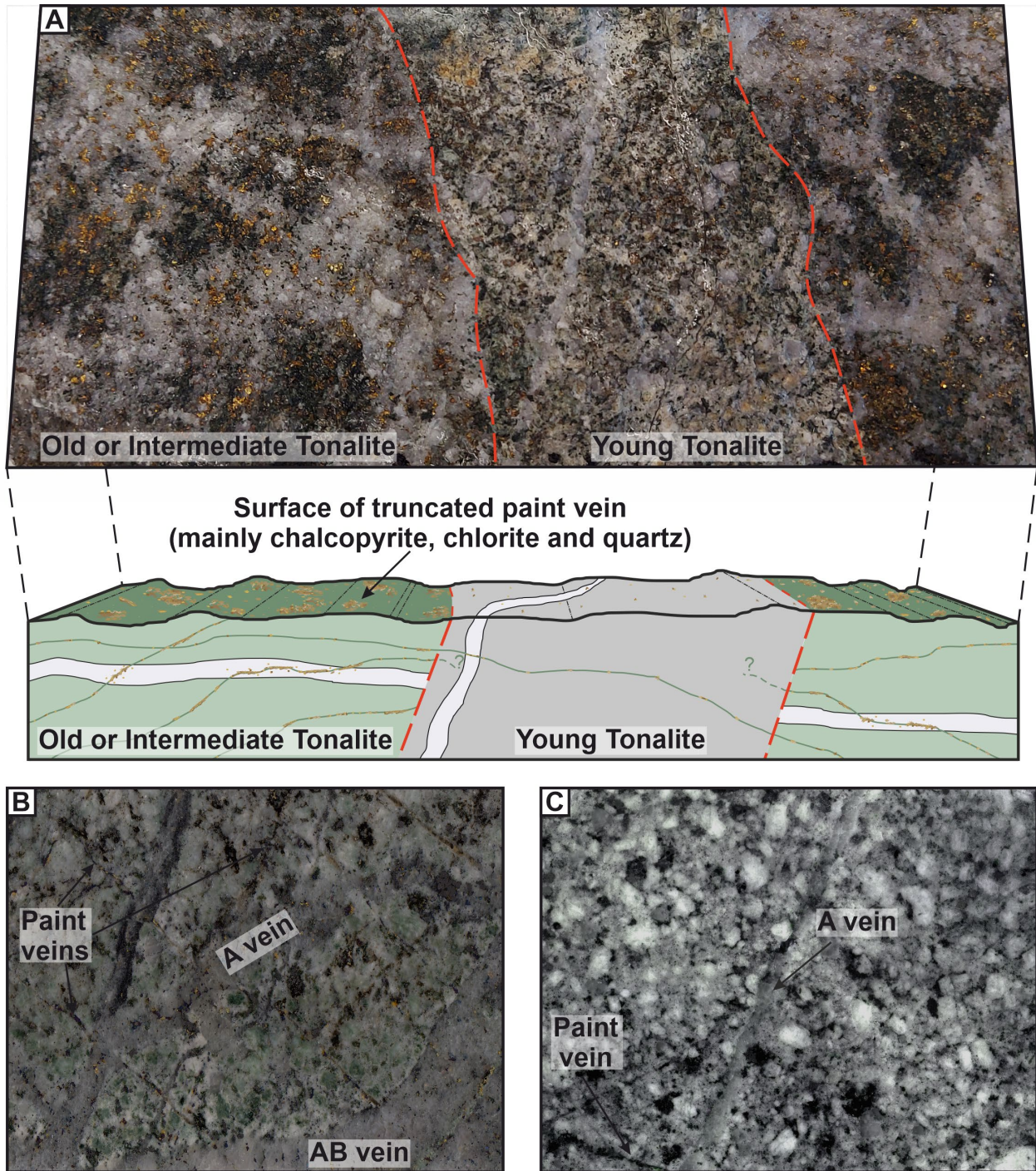


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1221 Figure 1: A) Location of Batu Hijau mine as well as other porphyry and epithermal deposits within the Neogene Sunda-Banda arc  
 1222 system, Indonesia (modified from Garwin, 2000). B) Alteration pattern of Batu Hijau at mine-scale with outline of Cu/Au ratios  
 1223 increasing from the center to the periphery (based on internal company reports provided by PT Amman Mineral Internasional).  
 1224 C) Sample locations along drill holes on cross-section 000 and distribution of fluid inclusion types. Sample numbers refer to  
 1225 samples listed in Table 1. Average bulk salinities (NaCl<sub>eq</sub>) for brine fluid inclusions are listed as small numbers in white in the  
 1226 respective parts of the pie charts. D) Cu ore-grades, and E) Au ore grades along the same cross-section (provided by PT Amman  
 1227 Mineral Internasional).

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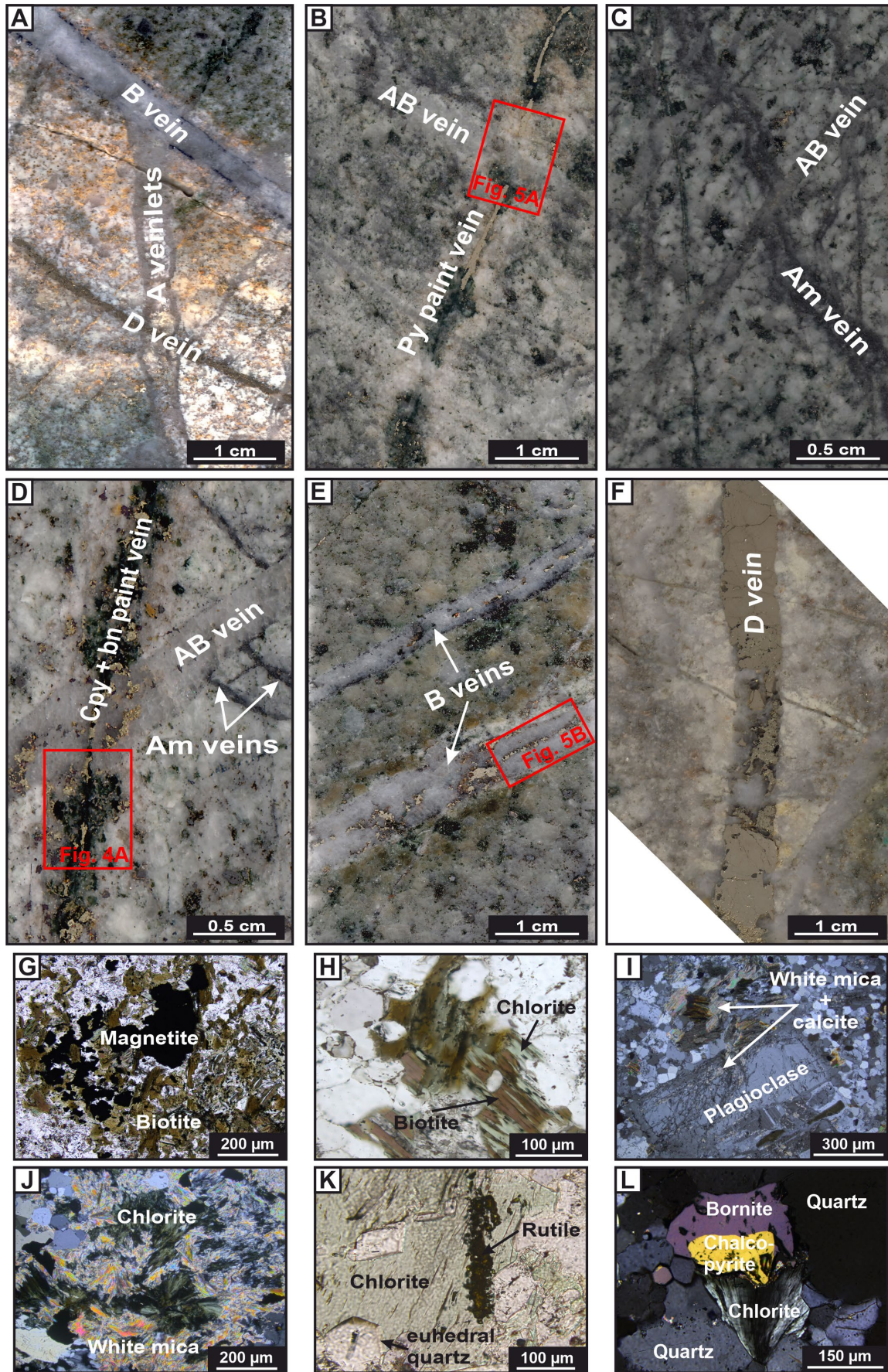


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1230 Figure 2: Intersection relations and petrographic differences between low-grade Young Tonalite (YT) and economically mineralized  
 1231 rocks (OT and IT) at Batu Hijau. A) Surface of paint vein in OT or IT truncated by YT. Although unambiguous truncations of paint  
 1232 veins at the contact to the YT are uncommon, the significantly lower amount of chalcopyrite and chlorite in YT compared to the  
 1233 older OT or IT indicates that the paint vein was truncated by the YT. B) and C) The general abundance of paint veins and color  
 1234 of economically and sub-economically mineralized samples is another indication that paint veins and low-temperature  
 1235 mineralization largely pre-date the YT: in the OT/IT (B, sample SBD 18 419m) paint veins are abundant and chlorite – white mica  
 1236 alteration is intensive resulting in the characteristic green color of highly mineralized rocks, whereas in the YT (C) paint veins are  
 1237 less common resulting in a less intense chlorite – white mica overprint so that the grey-white color of the porphyritic rock is largely  
 1238 preserved.

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1241 Figure 3: Examples of vein types and associated alteration assemblages from Batu Hijau. A) A-veins crosscut by molybdenite-  
1242 bearing B-vein and D-vein associated with feldspar-destructive alteration from the barren core (SBD 257 1226m). B) pyrite-  
1243 dominated paint-vein with characteristic chlorite-white mica alteration below the ore-shell apparently crosscut by barren AB-  
1244 veins (SBD 257 1132m). C) Typical magnetite-rich quartz stockwork veins in the barren core crosscut by AB-vein (SBD 257 925m).  
1245 D) Chalcopyrite (cpy) and bornite (bn)-rich paint vein with characteristic chlorite-white mica alteration halo crosscutting AB-vein  
1246 within the highly mineralized part of Batu Hijau (SBD 18 416m). E) Two parallel B-veins with sulfide centerline (SBD 18 230m). F)  
1247 Thick pyrite-dominated D-vein with quartz associated with feldspar-destructive white mica-clay alteration (SBD 276 1002m). G)  
1248 Transmitted light image of K-silicate alteration in the barren core with shreddy biotite and opaque magnetite replacing mafic  
1249 phenocryst (SBD 257 1132m). H) Partial alteration of presumably primary biotite to chlorite (SBD 257 507m). I) Polarized  
1250 transmitted light image of euhedral biotite and Ca-rich plagioclase with Na-rich rim replaced by white mica and calcite along  
1251 fractures (SBD 18 416m). J) Chlorite rosettes with white mica along paint-vein containing chalcopyrite and bornite in the high-  
1252 grade ore zone under polarized transmitted light (SBD 18 416m). K) Transmitted light image of brown, blocky rutile within  
1253 secondary chlorite as result of biotite replacement (SBD 18 416m). L) Combined reflected and polarized transmitted light image  
1254 of bornite and chalcopyrite associated with chlorite rosette within B-vein in the high-grade ore zone (SBD 95 601m).

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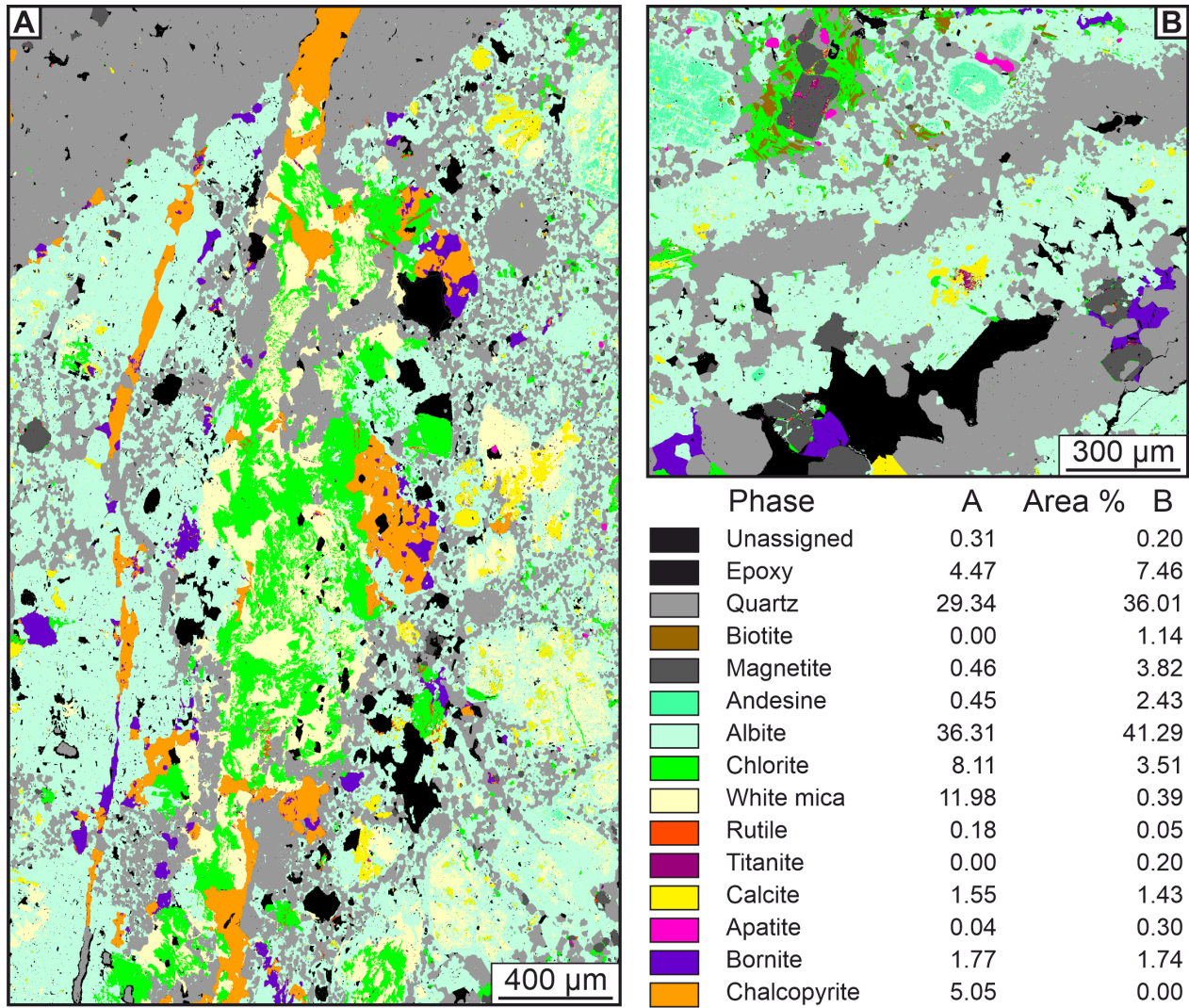
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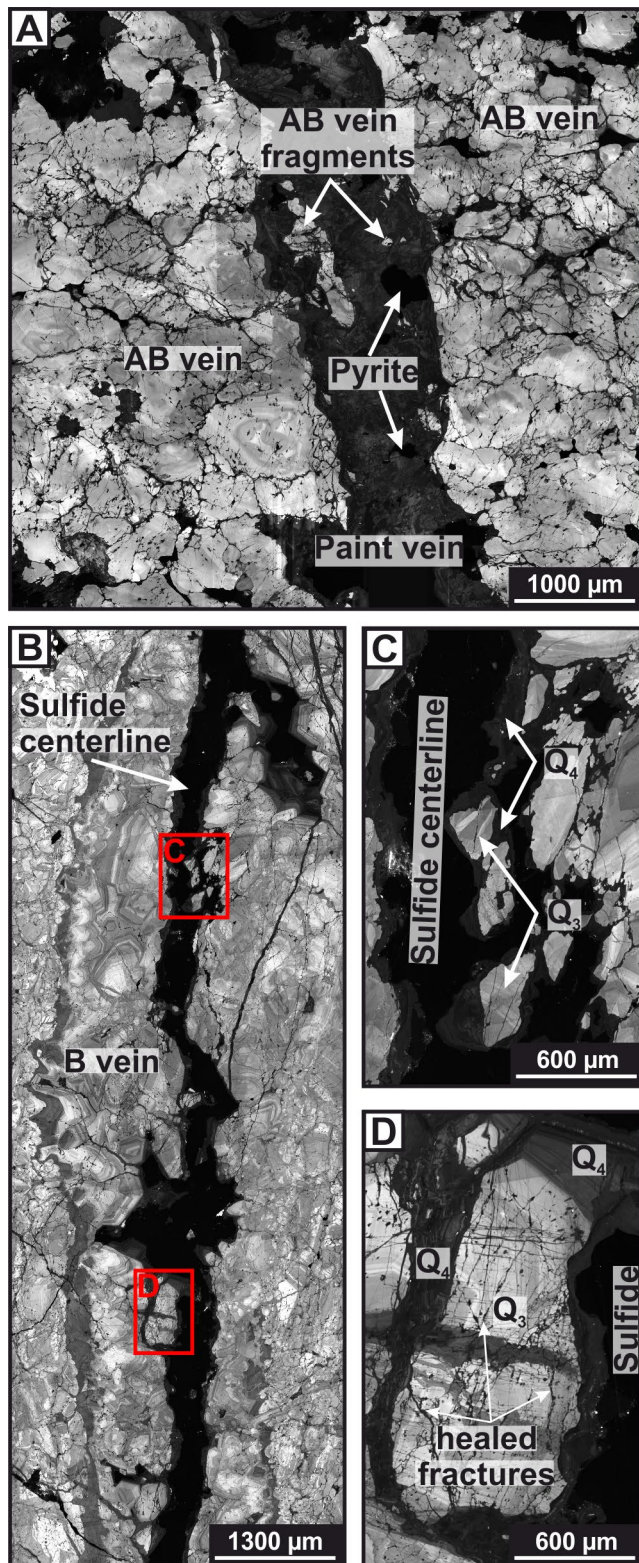
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 1275 Figure 4: Phase maps generated with iSpectra (Liebske, 2015) of paint veins with associated alteration. A) Paint vein containing  
 1276 chalcopyrite and bornite with distinctive chlorite-white mica alteration halo, crosscutting an AB vein as shown in Figure 3D (SBD  
 1277 18 416m). B) Bornite-dominated paint vein associated with incipient chlorite-white mica alteration (more andesine, magnetite  
 1278 and biotite is still present; SBD 21 797). Note the increased porosity along and within these paint veins, now filled with epoxy.

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1289 Figure 5: Cathodoluminescence images revealing textures of different hydrothermal quartz generations. A) Pyrite-bearing paint  
 1290 vein crosscutting AB vein. Such crosscutting relationships are difficult to interpret by eye or transmitted light observations, as in  
 1291 Figure 2B. Fragments of earlier vein generations can also obscure fluid inclusion distribution along such veins because buried  
 1292 fragments containing fluid inclusions associated with earlier quartz generations are not visible by CL imaging of the surface (SBD  
 1293 257 1132m). B) B vein with large euhedral  $Q_3$  grains and sulfide centerline as shown in Figure 2E (SBD 18 230m). Note that many  
 1294 different quartz generations are visible progressively overgrowing earlier quartz from the rim to the center. C) Enlarged section

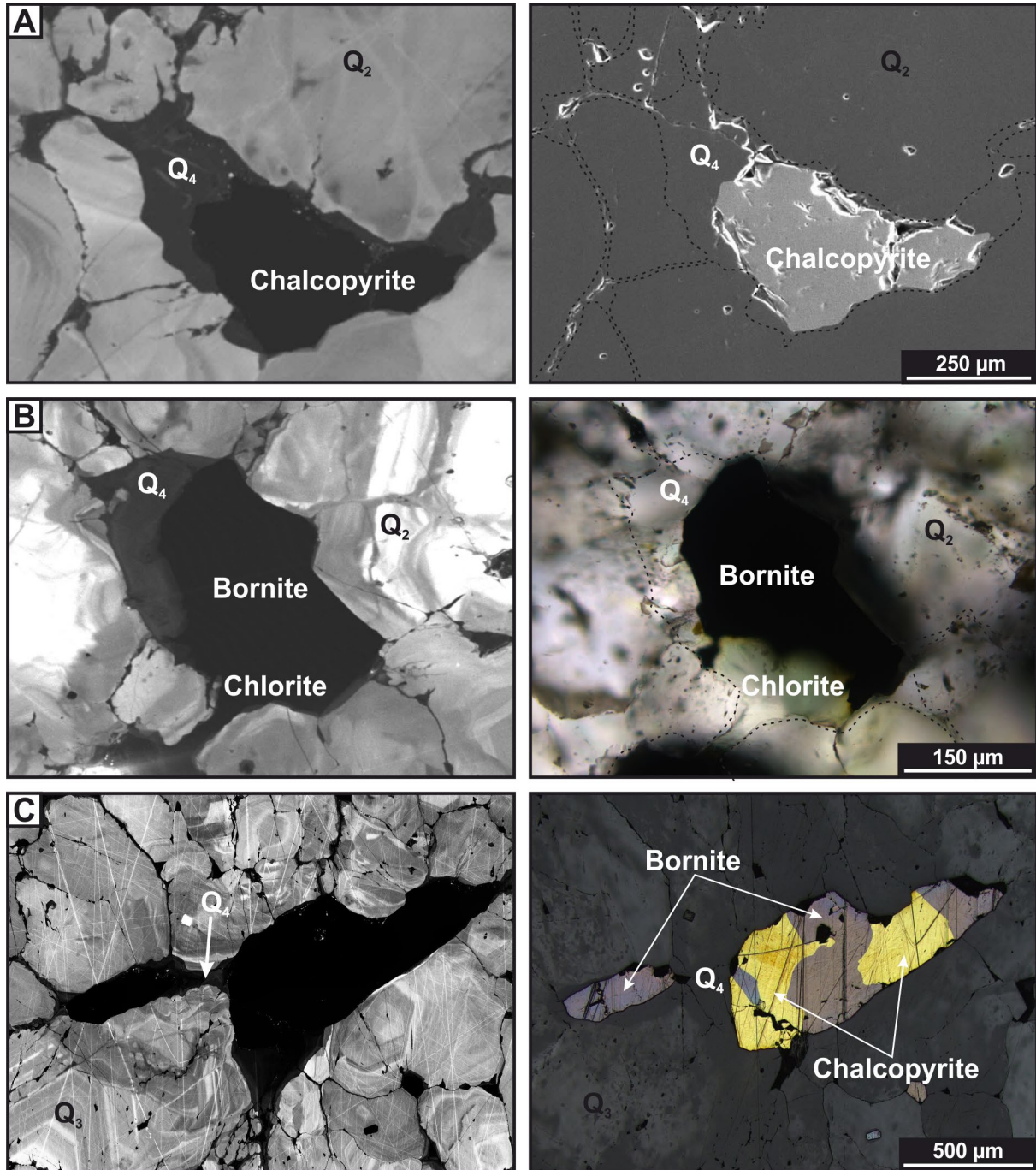
1295 of sulfide centerline showing  $Q_4$  sheathing the sulfides. The wavy contact between  $Q_3$  and  $Q_4$  indicates an interval of quartz  
1296 dissolution before renewed precipitation of quartz. D) Enlarged section of  $Q_3$  grains coated by  $Q_4$  associated with sulfide  
1297 centerline. Note that the micro-fracture network produced by trails of fluid inclusions leaching trace elements from the hosting  
1298 quartz (Lambrecht and Diamond, 2014) are truncated by  $Q_4$ . This is clear evidence for the pseudosecondary origin of the fluid  
1299 inclusions hosted by  $Q_3$  relative to  $Q_4$ .

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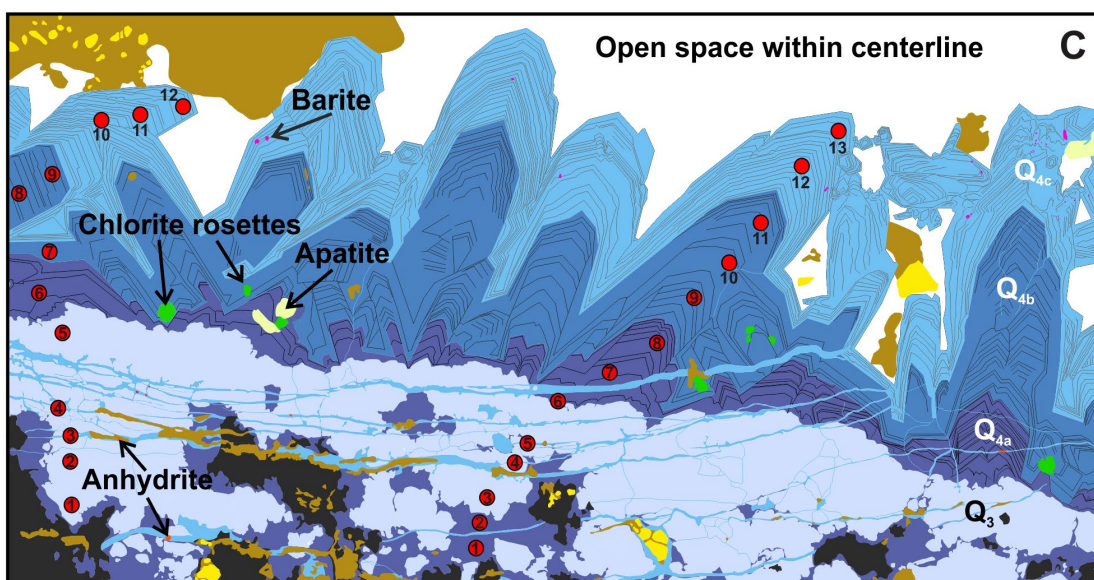
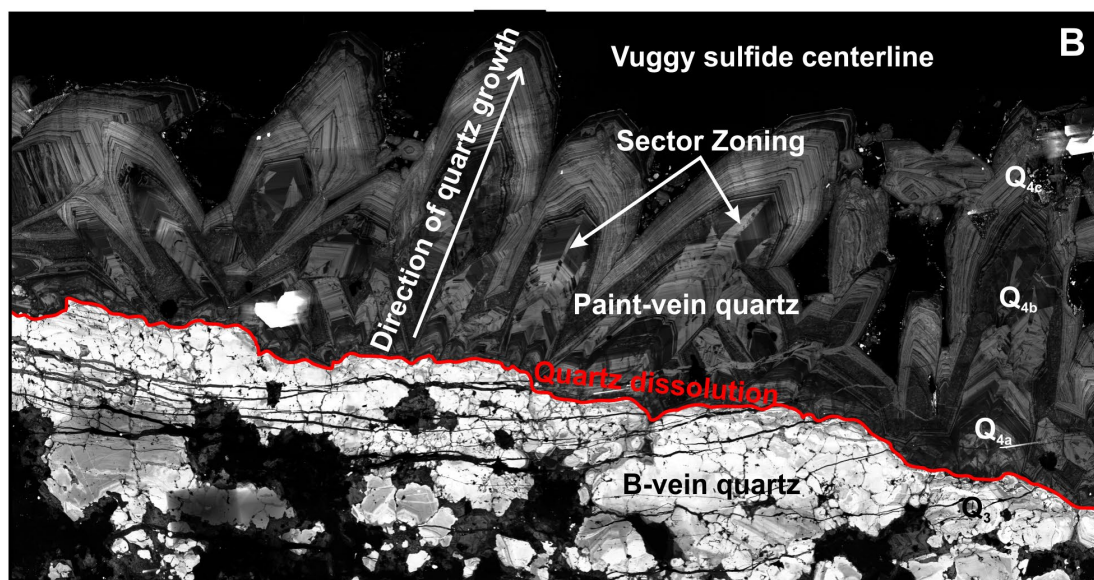
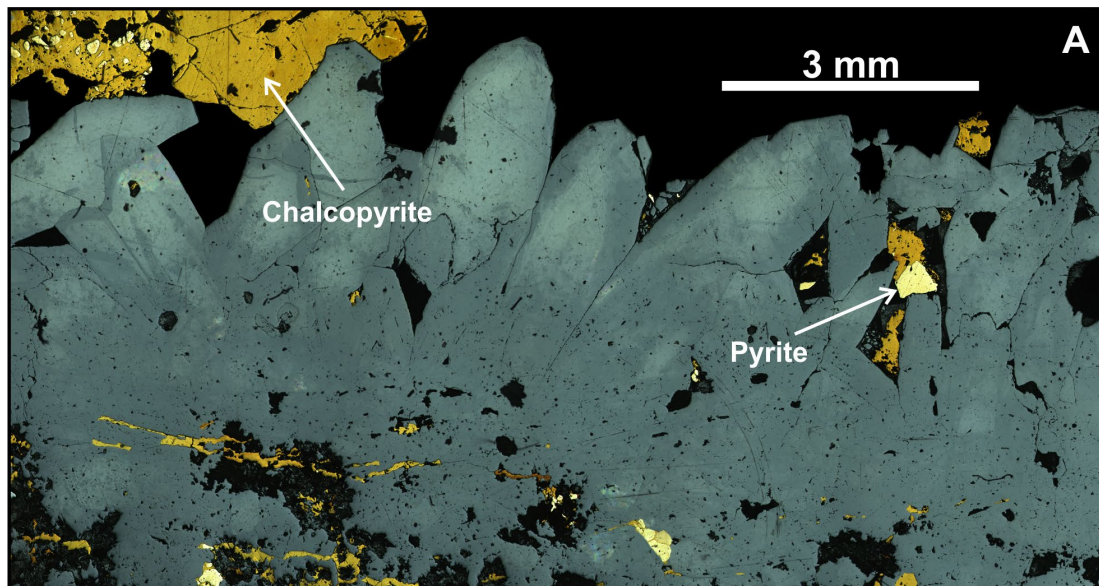




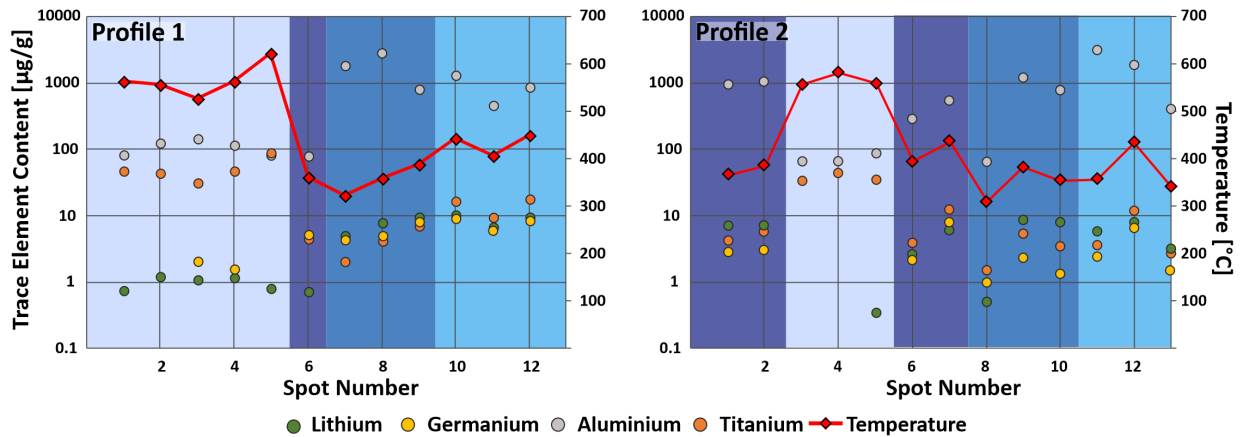
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1304 Figure 6: Textural context of sulfides hosted by hydrothermal quartz veins. A) CL image (left) and back-scattered electron image  
 1305 (right) of chalcopyrite grain hosted in AB vein. The sulfide grain is completely coated by Q<sub>4</sub> associated with paint veins as explained  
 1306 in the text (SBD 257 637m). B) CL image (left) and transmitted light image (right) of bornite grain associated with radial growing  
 1307 chlorite rosette. Both minerals are coated by Q<sub>4</sub> (SBD 21 797m). C) CL image (left) and reflected light image (right) of bornite and  
 1308 chalcopyrite along discontinuous sulfide centerline of B vein. Sulfide grains are connected by Q<sub>4</sub>-filled cracks crosscutting euhedral  
 1309 Q<sub>3</sub> grains (SBD 18 416m).



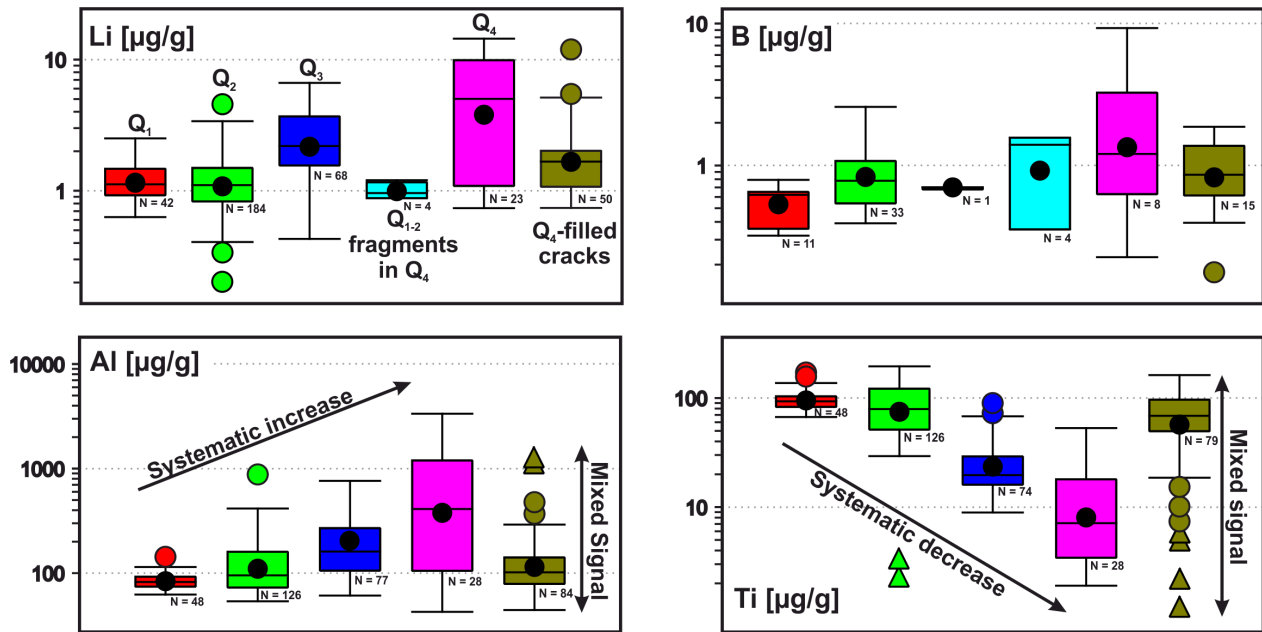


1311 Figure 7: Textural overview of reopened (A)B vein with vuggy sulfide centerline from the high-grade ore shell at Batu Hijau (SBD  
 1312 95 601m). A) Reflected light image of euhedral  $Q_3$  growing from the wall rock (bottom) to the center (top). Sulfides, mainly  
 1313 chalcopyrite with some pyrite, occur mainly in the open spaces or along cracks. Only few grains are within the quartz. B)  
 1314 Cathodoluminescence image of  $Q_{2-3}$  separated by a wavy dissolution front (red line) from the overgrowing  $Q_4$ . Bright luminescent  
 1315 spots represent anhydrite, apatite and barite inclusions. C) Simplified sketch of different  $Q_4$  generations and distribution of sulfides  
 1316 and alteration minerals. B-vein quartz is pure and does not host any alteration or sulfide minerals. The earliest paint-vein quartz  
 1317 generation ( $Q_{4a}$ , purple) hosts anhydrite, apatite and barite crystals as well as chlorite rosettes but apparently no sulfides. The  
 1318 second generation ( $Q_{4b}$ , dark blue) hosts chalcopyrite grains, commonly along growth zones, and chlorite. The last generation  
 1319 ( $Q_{4c}$ , light blue) hosts most of the chalcopyrite and pyrite along cracks crosscutting all earlier quartz generations or in open spaces.  
 1320 Many rounded and small anhydrite, apatite and barite inclusions are present as well within the quartz and chalcopyrite. An  
 1321 evolution of the paint veins from chalcopyrite (potentially  $\pm$  bornite) bearing over chalcopyrite-dominated to chalcopyrite + pyrite  
 1322 is visible here. The final stage of this evolution might be marked by the pyrite-dominated D veins. However, note that the  
 1323 distinction of different  $Q_4$  generations is simplified and somewhat arbitrary here and many more sub-generations could be  
 1324 distinguished. The red dots represent LA-ICP-MS analyses along profiles crossing different quartz generations (Fig. 8).



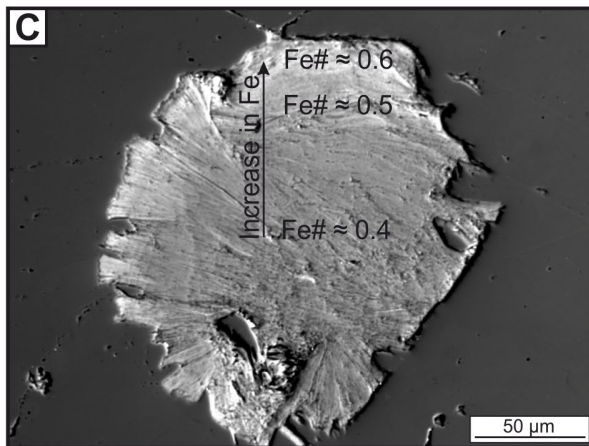
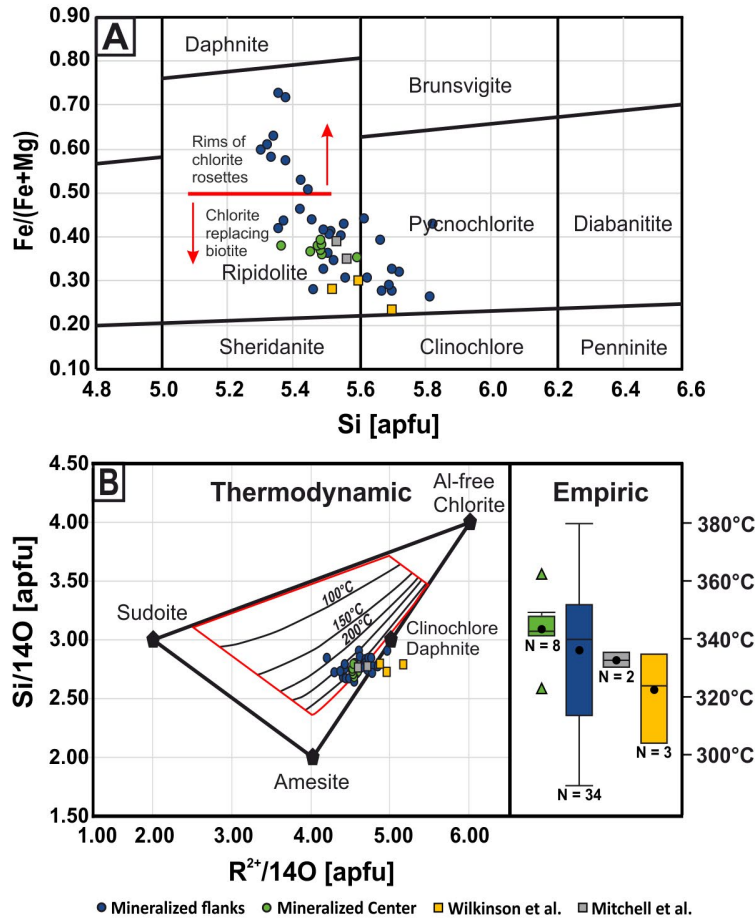
1326 Figure 8: Spot analyses along profiles (Fig. 7C). Profile 1 includes 12 spot analyses (profile on the left in Fig. 7C) and profile 2  
 1327 includes 13 spot analyses (right profile in Fig. 7C) crossing several quartz generations. Color-coding is the same as used in figure  
 1328 6C. In general, all shown trace elements are significantly higher in the  $Q_4$  generations, except for Ti which significantly decreases  
 1329 from up to 100  $\mu\text{g/g}$  in the  $Q_{2-3}$  to below 15  $\mu\text{g/g}$  in  $Q_{4a}$ . The temperature calculation is dependent on Ti content (Wark and  
 1330 Watson, 2006; Huang and Audétat, 2012).





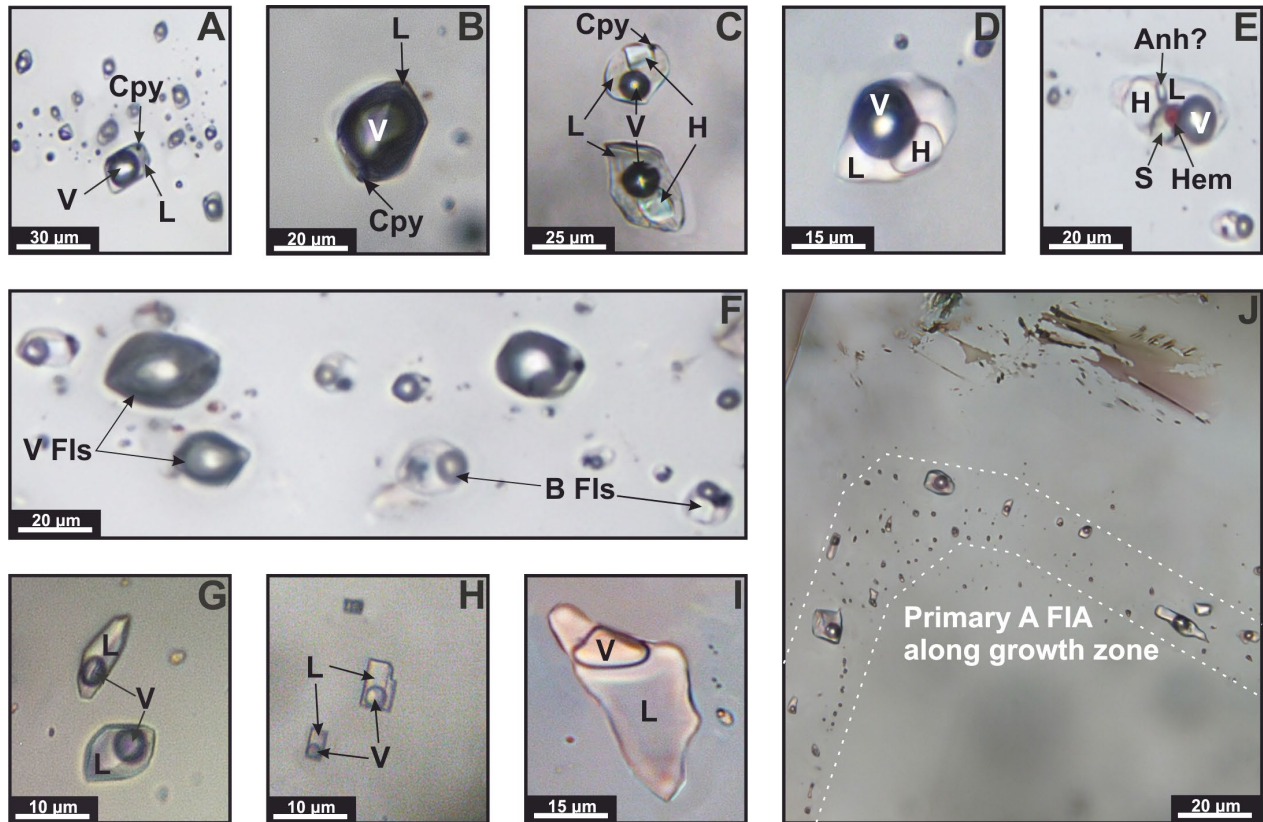
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1332 Figure 9: Box plots of selected trace elements in quartz (Q<sub>1</sub> to Q<sub>4</sub> represented by different colors). The lower whisker represents  
 1333 the first quartile, the bottom and top of the box the second and third quartile, respectively, and the upper whisker the fourth  
 1334 quartile of the data. The line and black dot within the boxes represent the median and average concentrations, respectively.  
 1335 Outliers are marked as colored circles (further away from the box than 1.5 x (value at the top of the box – value at the bottom of  
 1336 the box)) and far outliers are marked as colored triangles (further way from the box than 3.0 x (value at the top of the box – value  
 1337 at the bottom of the box)). Note that outliers have a pronounced effect on calculated averages compared to median values. All  
 1338 quartz generations representing different vein types partially overlap in their trace element concentrations. Nevertheless, on  
 1339 average Li, B, and Al concentrations increase from Q<sub>1</sub> to Q<sub>4</sub>, whereas Ti decreases. Trace elements in Q<sub>4</sub> have the largest variations  
 1340 in composition, almost covering the total range of concentrations.



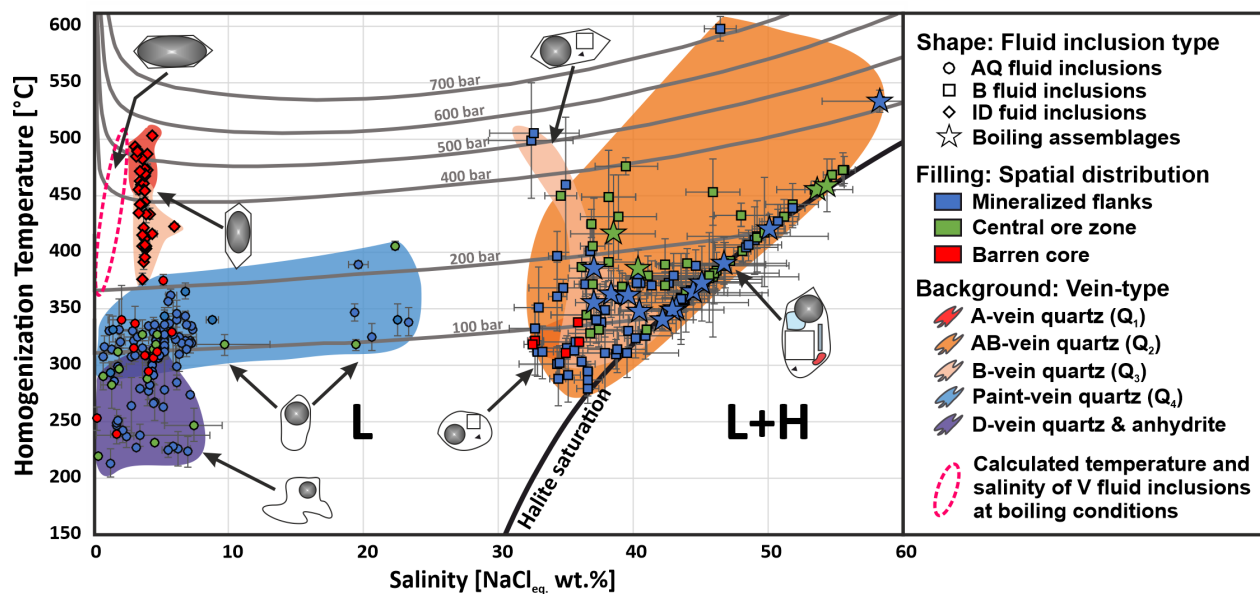
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1342 Figure 10: Results of chlorite analyses and comparison with EPMA data of chlorites from Batu Hijau obtained by Mitchell et al.  
 1343 (pers. commun., 1998) and Wilkinson et al. (2015). A) Classification scheme after Hey (1954) showing that most chlorites plot  
 1344 within the ripidolite and pycnochlorite field. B) T-R<sup>2+</sup>-Si diagram of Bourdelle and Cathelineau (2015) predicting formation  
 1345 temperatures of chlorites based on a semi-empirical approach valid at low temperatures (<350 °C) and low pressures (< 3-4 kbar)  
 1346 considering chlorite-quartz equilibrium (Inoue et al., 2009; Bourdelle et al., 2013). Some points plot outside of the valid  
 1347 temperature field (red area), indicating that formation temperatures were higher than 350 °C or chlorite-quartz was not at  
 1348 equilibrium. However, most points (75%) plot between 300 and 350 °C, overlapping with results obtained by empirical calibrations  
 1349 (Jowett, 1991). C) Back-scattered electron image of chlorite rosette hosted by euhedral dull-luminescent paint-vein quartz (SBD  
 1350 95 601m). Brighter rims indicate higher densities due to higher Fe contents.



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1352 Figure 11: Different types of fluid inclusions hosted by veins. A) Two-phase intermediate density fluid inclusions with vapor (V)  
 1353 and liquid (L) filling the available space in equal proportions. Triangular opaque solids, probably chalcopyrite (Cpy), are present in  
 1354 almost all inclusions of this type. B) Vapor fluid inclusion filled mainly by vapor (~ 90 vol.%) and a thin rim of liquid wetting the  
 1355 fluid-crystal interface. Chalcopyrite daughter crystals are commonly present. C) B<sub>1</sub> fluid inclusions with small bubbles (~ 20 vol.%)  
 1356 and halite (H). D) High-temperature B<sub>2</sub> fluid inclusion with large vapor bubble (~ 30 vol.%). E) Complex B<sub>3</sub> fluid inclusion with  
 1357 several daughter crystals besides halite, such as sylvite (S), red flakes of hematite (Hem), and blueish, birefringent, commonly  
 1358 elongated, transparent anhydrite (Anh). Chalcopyrite is also present but not visible because it is covered by the vapor bubble. F)  
 1359 Boiling trail hosting brine and vapor fluid inclusions. G) Low-salinity aqueous fluid inclusions (AQ<sub>2</sub>) within euhedral dull-  
 1360 luminescent paint-vein quartz. H) Rectangular aqueous fluid inclusions hosted by anhydrite in D-veins (AQ<sub>3</sub>). I) Low-temperature,  
 1361 irregular and flat aqueous fluid inclusions may be associated with D-vein formation (AQ<sub>3</sub>). J) Primary aqueous fluid inclusion  
 1362 assemblage along growth zone in euhedral paint-vein quartz (Q<sub>4</sub>). This assemblage contains high-salinity aqueous inclusions (AQ<sub>1</sub>)  
 1363 in which dissolution of hydrohalite was visible during freezing experiments. High- (AQ<sub>1</sub>) and low-salinity aqueous fluid inclusions  
 1364 (AQ<sub>2</sub>) are indistinguishable at room temperature.



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1366 Figure 22: Microthermometry results of different fluid inclusion types within different parts and hydrothermal veins at Batu Hijau.  
 1367 Each dot represents a fluid inclusion assemblage with, on average, five individual fluid inclusions. Errors in homogenization  
 1368 temperature and salinity represent standard deviations between individual fluid inclusions within each assemblage. Due to the  
 1369 difficulty of conducting microthermometric experiments on vapor inclusions, salinities and homogenization temperatures were  
 1370 inferred from boiling assemblages using the SoWat software (Driesner, 2007; Driesner and Heinrich, 2007) and are shown with a  
 1371 dashed line. Fluid inclusion assemblages plotting on the halite saturation curve homogenized to the liquid phase by halite  
 1372 dissolution. Note that only intermediate density fluid inclusions are pressure corrected.

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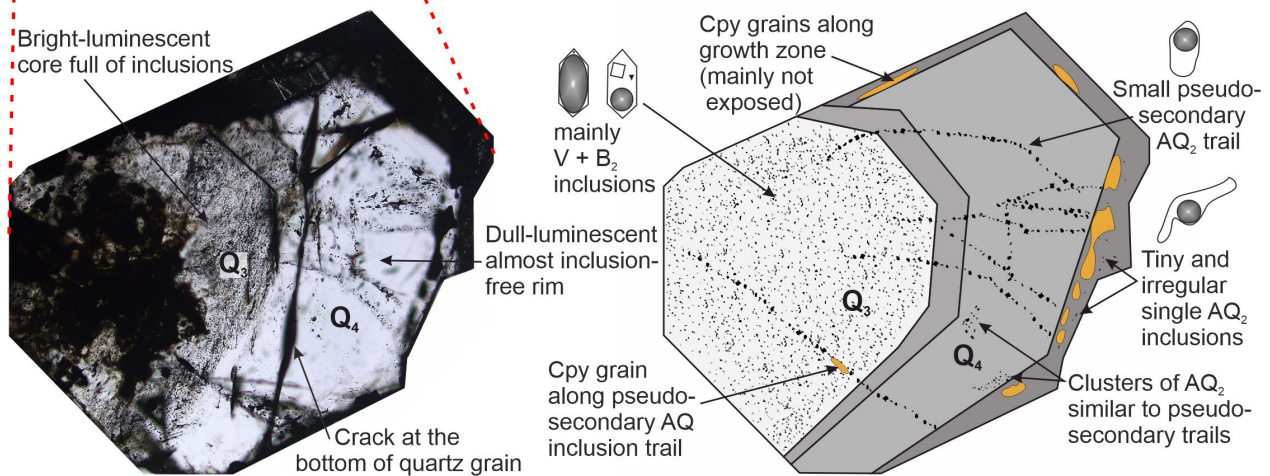
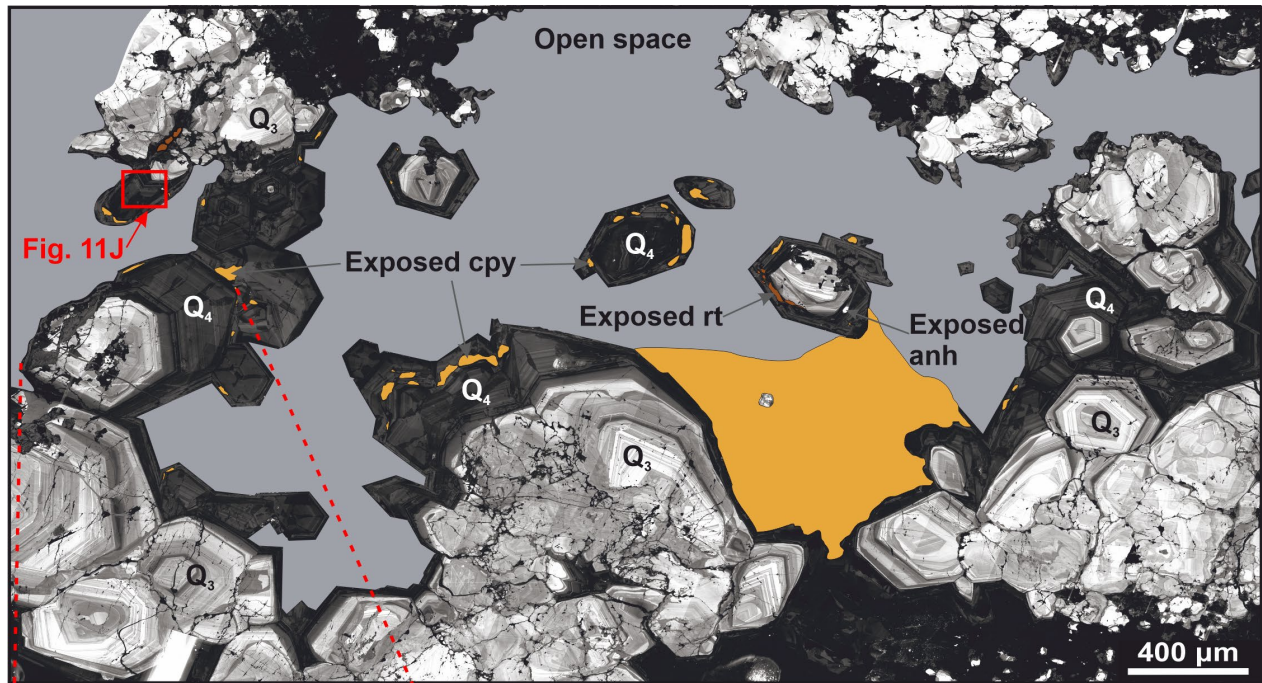
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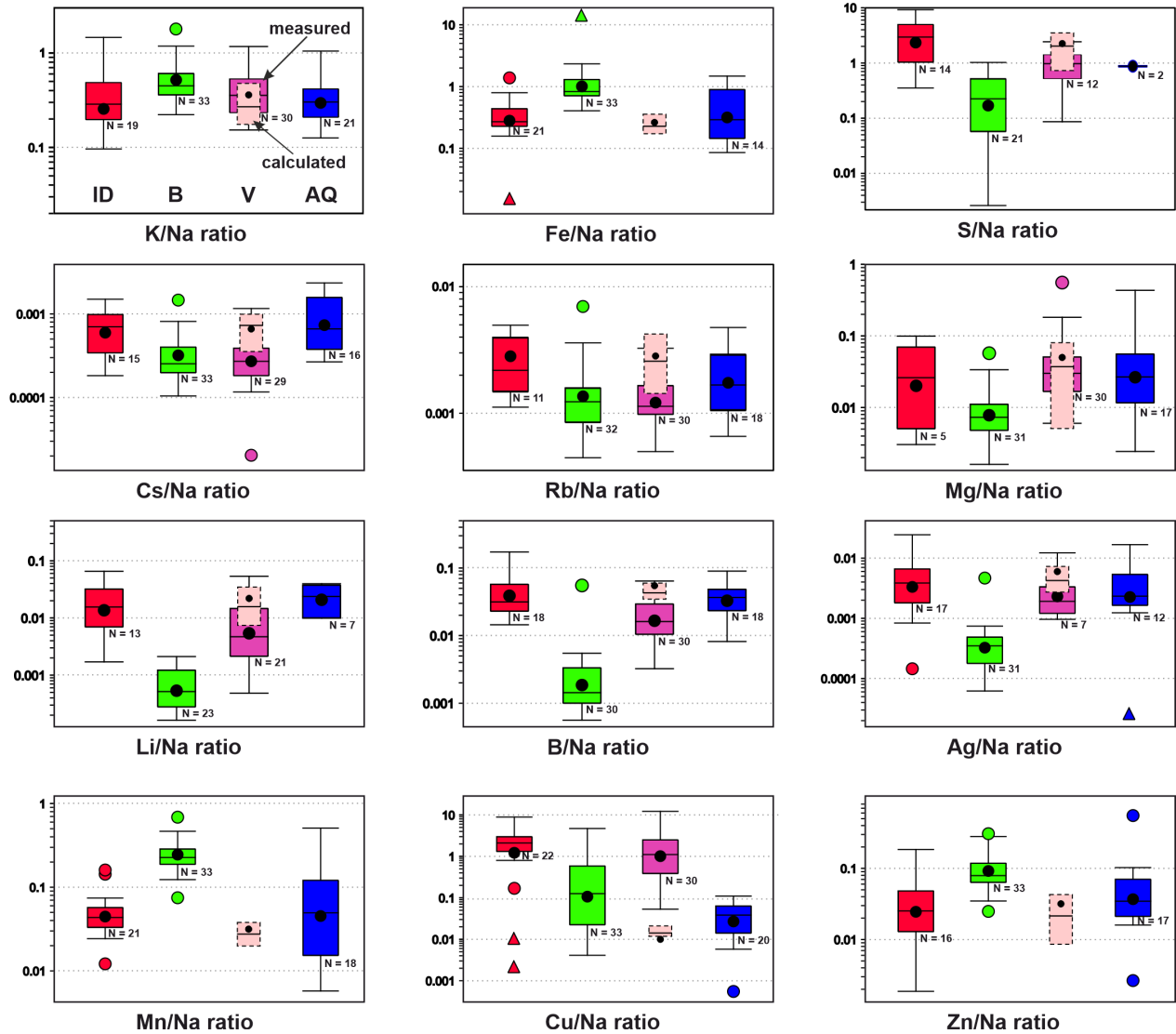




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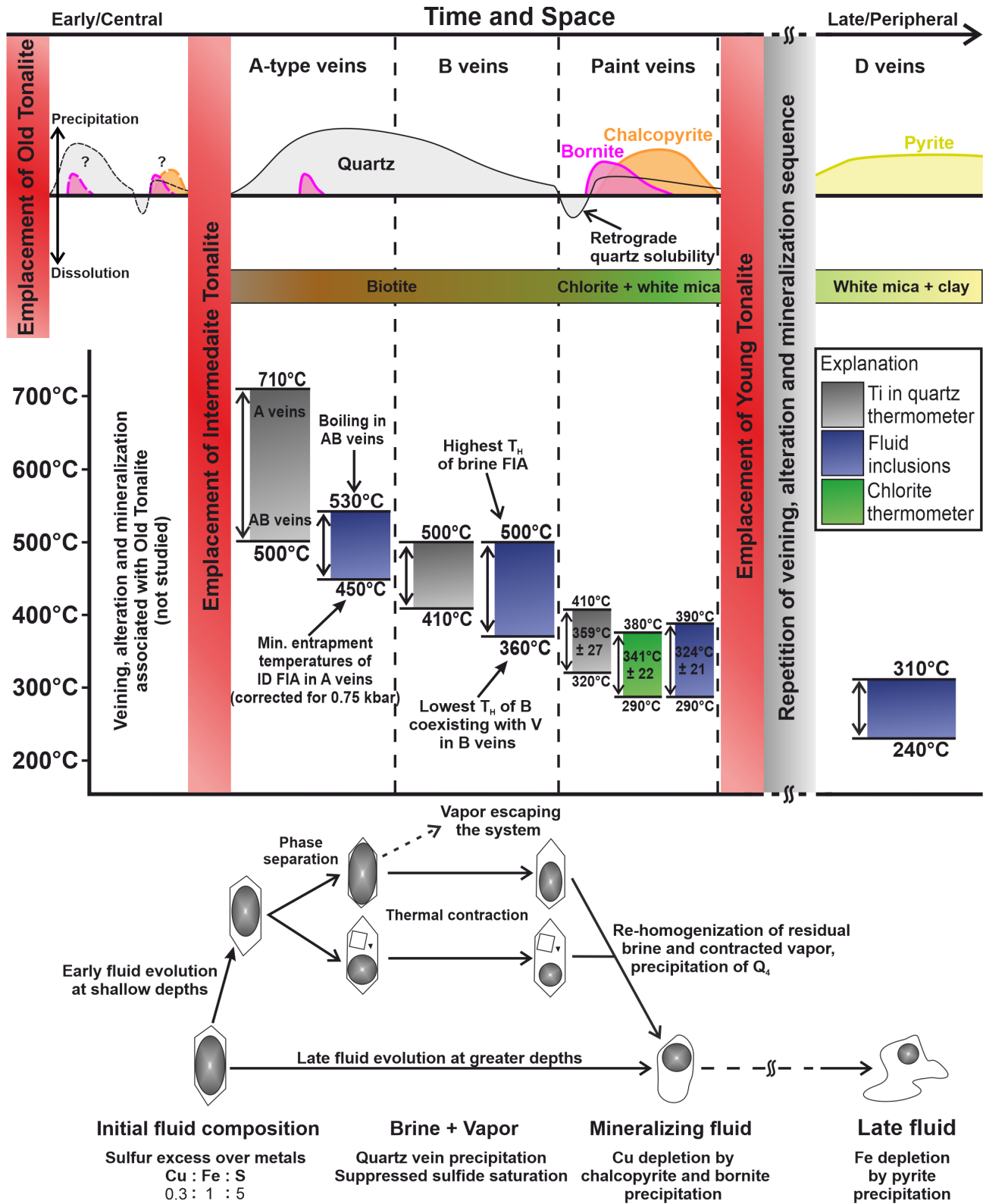
1386 Figure 13: Cathodoluminescence image of paint vein intersecting B vein (top) and fluid inclusion distribution within a selected  
 1387 quartz grain (bottom; SBD 257 694m). Euhedral Q<sub>4</sub> overgrowing earlier euhedral Q<sub>3</sub>, which shows several internal dissolution and  
 1388 re-precipitation textures. Chalcopyrite (cpy), anhydrite (anh) and rutile (rt) grains are exclusively hosted by Q<sub>4</sub>, commonly along  
 1389 growth zones. Note that only exposed chalcopyrite and rutile are colored on the CL map. A transmitted light image and a schematic  
 1390 sketch of a selected quartz grain showing a bright-luminescent core and a dull-luminescent overgrowth with sulfide grains along  
 1391 a specific growth zone is shown in the lower part of the figure. The displayed distribution of fluid inclusions is typical for such  
 1392 composite grains, with countless B and V inclusions in the bright-luminescent core and an almost fluid inclusion free dull-  
 1393 luminescent quartz overgrowth. Fluid inclusions hosted by Q<sub>4</sub> are exclusively of the aqueous type occurring as pseudo-secondary  
 1394 trails that cut the bright-luminescent core and are truncated by the chalcopyrite-hosting growth zone. The outermost rim of the  
 1395 quartz crystal hosts tiny and irregular shaped AQ inclusions.

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1398 Figure 14: Box plots showing the element/Na mass ratios for different types of fluid inclusions (colors defined in the K/Na box).  
 1399 Each box represents the compositional range of FIAs of every fluid inclusion type. The total number of fluid inclusion assemblages  
 1400 (N) is indicated below each box. Both analyzed and calculated ratios for vapor inclusion are shown. Mass balance calculations are  
 1401 based on common boiling assemblages to correct for entrapped portions of brine following the procedure of Landtwing et al.  
 1402 (2010). In the same way, element/Na ratios were calculated for elements not analyzed in vapor inclusions. Calculated ratios are  
 1403 shown as a thinner pink box, commonly partially overlapping with the range of analyzed ratios in V FIAs. For explanation of Box  
 1404 and whisker plots see description of figure 9.



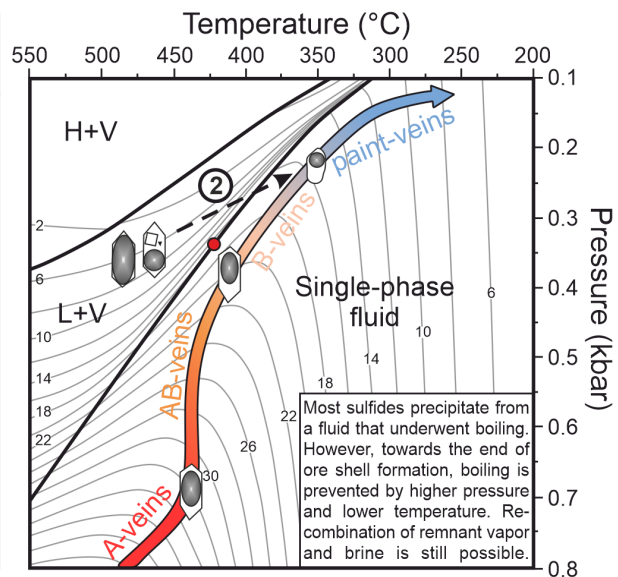
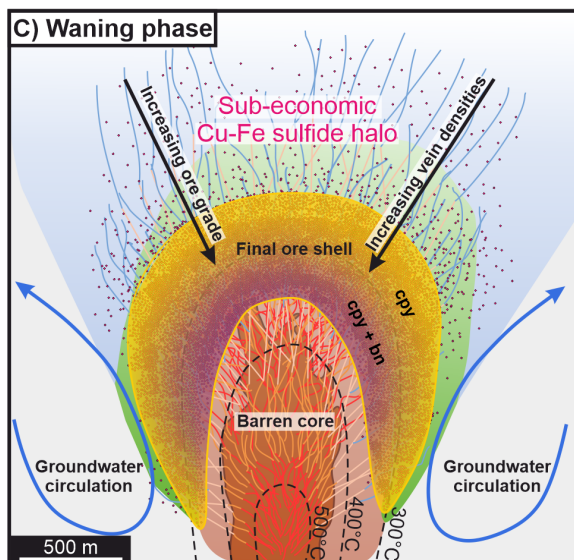
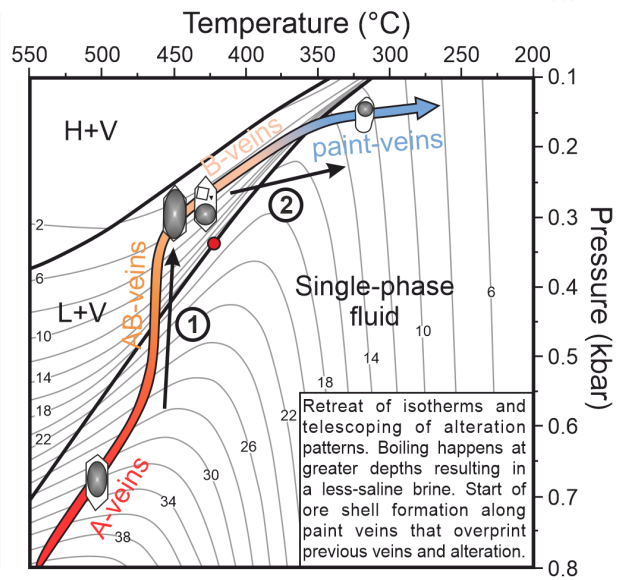
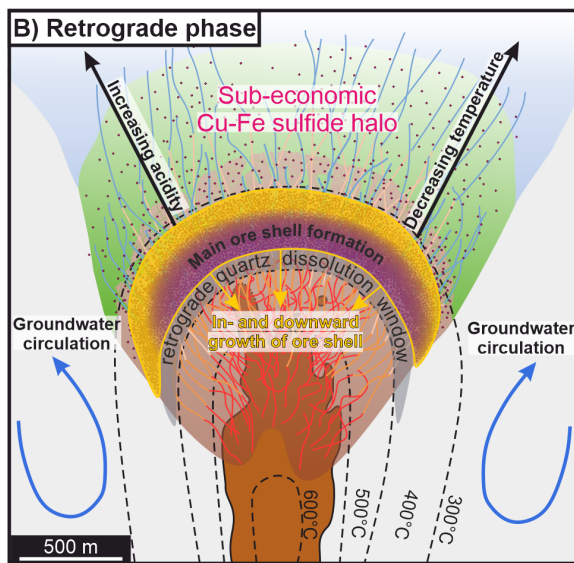
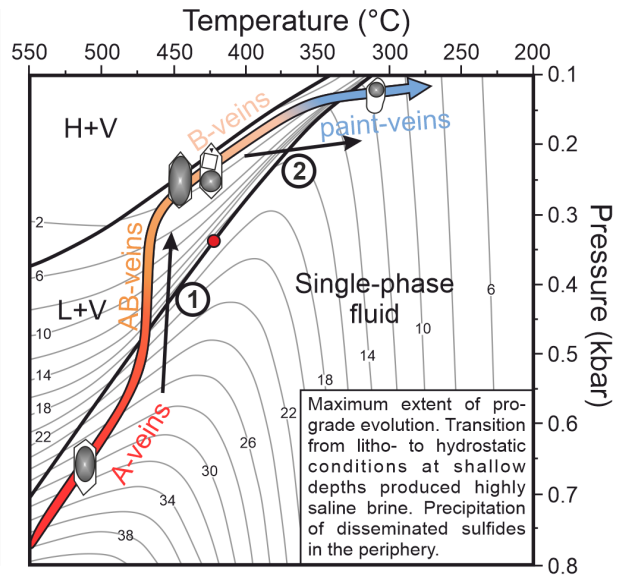
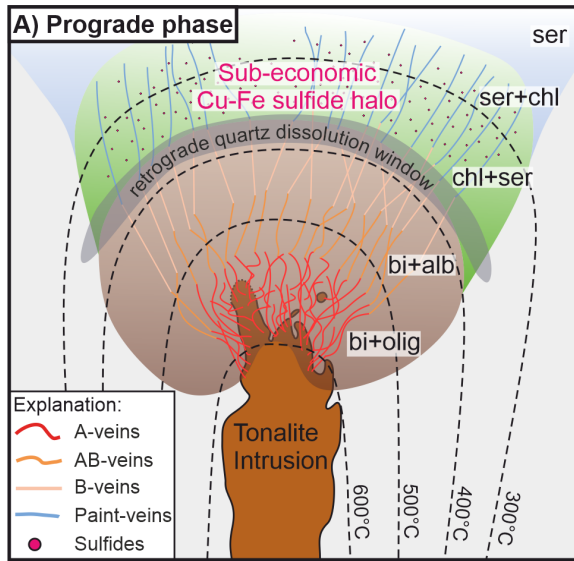
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Figure 15: Summary showing from the top to the bottom the different veining stages, the precipitation or dissolution of selected hydrothermal minerals, the main alteration silicates in the host rock, the temperature evolution as documented by the various geothermometers, and a schematic evolution of the fluid with time based on fluid inclusion observations. Note that a similar veining, alteration and mineralization is expected to have occurred between the emplacement of the OT and IT but was not studied here due to the scarcity of samples that can be clearly identified as OT. Locally, high-temperature bornite formed during

1411 quartz vein formation (Setyandhaka et al., 2008) but since this precipitation event was not documented within our sample  
1412 collection, its exact timing relative to the different quartz-dominated vein types remains unknown. The lower part of the diagram  
1413 summarises the sequence of fluid inclusions, interpreted in Figure 16A and particularly during the main mineralization stage shown  
1414 in Figure 16B.





1416 Figure 36: Hydrothermal evolution at Batu Hijau as schematic sketch in the left column and fluid evolution as function of  
1417 temperature and pressure in the right column. Quartz solubility contours in mmol/kg of fluid are taken from Monecke et al. (2018),  
1418 the red dot represents the critical point of a fluid containing 5 wt.% NaCl. Lateral zonation of propylitic alteration is not shown for  
1419 simplicity. The shown schematic model involves a progressive telescoping process during retreat of isotherms producing  
1420 overprinting alteration, crosscutting vein types and mineralization with the associated evolution of the fluid as recorded by fluid  
1421 inclusions. Arrows labeled 1 and 2 represent phase separation and re-homogenization of previously separated brine and vapor,  
1422 respectively. Sulfide precipitation starts early in the hydrothermal evolution but distal from the present day ore shell, producing  
1423 the wide halo of sub-economically mineralized rocks around Batu Hijau as shown in part A). By reduction of the volume and  
1424 stabilization of the isotherms at depth, sulfide precipitation is more focused, which led to ore shell formation. Part B) shows the  
1425 initiation of ore shell formation during the retrograde stage of the system. The major part of ore shell forms during the retrograde  
1426 phase by transient stabilization of the in- and downward retracting mineralization front. The final shape of the ore shell mimics  
1427 the positions of the 350°C isotherm with time and is shown in part C). During the waning stage only minor amounts of Cu will be  
1428 added to or potentially even removed from the system. Ground water convection through the porous volcanoclastic host rocks  
1429 effectively cools the hydrothermal systems and produces steep isotherms defining the ultimate shape of the ore shell (Weis,  
1430 2015).

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Table 2: Characteristics of vein types and associated alteration at Batu Hijau.

Vein Type	Mineralogy	Alteration Halo	Thickness	Geometry	CL intensity of quartz	Fluid Inclusions	Pressure regime	References
A	Qtz + mag ± bi ± olig ± anh Cpy + bn	Bi + olig ± mag	< 1.5 cm	Irregular, wavy	Bright-luminescent, anhedral granular	ID in barren core, less commonly V and rarely B	Lithostatic	1), 2), 3), 4), this study
AB	Qtz ± mag ± bi/chl ± alb ± anh Cpy ± bn	Alb ± bi	1 – 5 cm	Regular, less wavy	Bright-luminescent, an- to subhedral, granular to prismatic	ID in barren core, B + V in the mineralized zone (boiling)	Lithostatic	2), 3), 4), this study
B	Qtz ± chl ± alb ± anh Cpy ± bn (± mo, barren core)	Alb ± chl	< 2.5 cm	Regular, straight, centerline	Medium-luminescent, comb-like, euhedral with oscillatory growth zoning	ID in barren core, B + V in the mineralized zone (boiling)	Litho- to hydro- or vaporstatic	1), 2), 3), 4), this study
Paint (C)	Chl + ser + rt ± qtz ± anh ± ba ± tit ± cal ± ap Cpy ± bn or cpy ± py or py ± cpy	Chl + ser + rt + cal ± anh ± hem	< 0.1 cm, in rare cases up to 1.0 cm	Indistinct, irregular, vuggy	Dull-luminescent, comb-like, anhedral to euhedral with oscillatory growth zoning	Small rounded A	Hydro- to vaporstatic	this study (4)
D	Py ± cpy ± ser ± anh	Ser + py	0.5 - 2 cm	Regular, straight	Dull-luminescent fragments, euhedral	Irregular A in qtz and rectangular A in anh	Hydro- to vaporstatic	1), 2), 3), 4), this study

Mineral abbreviations: alb – albite, anh – anhydrite, ap – apatite, ba – barite, bi – biotite, bn – bornite, cal – calcite, chl – chlorite, cpy – chalcopyrite, hem – hematite, mag – magnetite, mo – molybdenite, olig – oligoclase, py – pyrite, qtz – quartz, rt – rutile, ser – white mica, tit-titanite.

Abbreviations for vein and fluid inclusion types as explained in text.

References: 1) Clode et al., 1999, 2) Garwin, 2000, 3) Arif and Baker, 2004, 4) Idrus et al., 2009

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1449 Table 3: Characteristics and compositions of the different quartz generations.

Quartz Gen.	Vein Type	CL characteristics	Habitus	Trace Element Concentrations			
				Li	B	Al	Ti
Q <sub>1</sub>	Am, A	Bright luminescent, wavy growth zonation rarely visible	Anhedral, sugary mosaic texture	0.6 - 2.5 Ø: 1.2	0.3 - 0.8 Ø: 0.5	62 - 114 Ø: 84	67 - 144 Ø: 95
Q <sub>2</sub>	AB	Bright luminescent, wavy growth zonation commonly present	An- to subhedral, sugary mosaic texture	0.4 - 3.4 Ø: 1.1	0.4 - 2.6 Ø: 0.8	54 - 415 Ø: 111	29 - 159 Ø: 75
Q <sub>3</sub>	B	Medium luminescent, sharp euhedral growth zoning	Prismatic, euhedral growth zoning, comb-texture	0.4 - 6.6 Ø: 2.2	0.7 - 0.7 Ø: 0.7	61 - 763 Ø: 161	9 - 68 Ø: 24
Q <sub>4</sub> (Q <sub>5</sub> ?)	Paint, D	Dull luminescent, sharp euhedral growth zoning	Like Q <sub>3</sub> in open spaces, anhedral in cracks	0.7 - 14.4 Ø: 3.8	0.2 - 9.3 Ø: 1.3	43 - 3348 Ø: 377	2 - 53 Ø: 8

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