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1 **Disentangling the compositional variations of lawsonite in**
2 **blueschist-facies metasediments (Schistes Lustrés, W. Alps)**

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5 Benjamin Lefeuvre^{1,2*}, Benoît Dubacq¹, Anne Verlaquet¹, Clément Herviou^{1,3,4}, Stephanie
6 Walker⁵, Benoît Caron¹, Ethan Baxter⁵, Philippe Agard¹

7 ¹ Sorbonne Université, CNRS-INSU, Institut des Sciences de la Terre de Paris, IStEP UMR 7193, F 75005 Paris,
8 France

9 ² Laboratoire des Fluides Complexes et leurs Réservoirs – IPRA, E2S-UPPA, TotalEnergies, CNRS, Université
10 de Pau et des Pays de l'Adour, UMR5150 Pau, France

11 ³ Department of Earth, Marine and Environmental Sciences, University of North Carolina, Chapel Hill, NC 27599-
12 3315, USA

13 ⁴ Université de Lyon 1, CNRS UMR5276, ENS de Lyon, LGL-TPE, Villeurbanne Cedex, France.

14 ⁵ Department of Earth and Environmental Sciences, Boston College, 140 Commonwealth Ave, Chestnut Hill,
15 MA02467, USA

16 *Corresponding author: benjamin.lefeuvre@univ-pau.fr

17
18 Keywords: lawsonite, metasediments, fluid/mass transfer, Schistes Lustrés, REE (LA-ICP-
19 MS), Strontium isotopes (TIMS)

25 **Abstract**

26 *Lawsonite is important as a carrier of H₂O in subduction zones and as a petrological tracer. The trace*
27 *element content of lawsonite in mafic rocks has been used as a record of fluid-rock interactions but has*
28 *received less attention in metamorphosed oceanic sediments. This study documents the major and trace*
29 *element composition, together with ⁸⁷Sr/⁸⁶Sr isotopic ratios, of the different lawsonite types identified in*
30 *the upper units of the Schistes Lustrés complex of the Western Alps, a paleo-accretionary prism of the*
31 *Liguro-Piemont slow-spreading ocean subducted up to 40 km. The lawsonite-rich upper units of the*
32 *Schistes Lustrés complex are principally composed of metamorphosed pelitic sediments and carbonates.*
33 *Lawsonite content reaches 40 vol.% in the rock matrix and in veins. All lawsonite types originate from*
34 *prograde metamorphic reactions which occurred up to peak metamorphism. Lawsonite compositions*
35 *have been measured in-situ with electron microscopy, microprobe and laser-ablation mass*
36 *spectrometry. Lawsonite separates have been measured for Sr content and ⁸⁷Sr/⁸⁶Sr isotopic ratios using*
37 *TIMS. Bulk rock compositions have been measured with ICP-OES and ICP-MS. Analysis contamination*
38 *by minute retrograde white mica inclusions in lawsonite crystals precluded using univalent cations for*
39 *petrological interpretations. For other trace elements, the variability of lawsonite appears extremely*
40 *high, with crystal compositions varying between samples and between crystals in individual samples.*
41 *However, clear patterns emerge between lawsonite types, reflecting equilibrium and out-of-equilibrium*
42 *processes unrelated to pressure-temperature conditions. At crystal scale, textural hourglass zoning*
43 *predominates in lawsonite from the schist, while growth zoning is found in both schists and veins. The*
44 *combination of both mechanisms results in spectacular zoning in Ti and in rare earth element contents*
45 *spanning four orders of magnitude. Over time, the La/Yb ratio decreases strongly (from ~100 to ~1), as*
46 *La appears much more sensitive to surface effects leading to textural hourglass zoning. Interface-*
47 *coupled dissolution-precipitation also contributes to decoupling of less mobile elements in the schist.*
48 *This is best observed for Ti, but rare earth element and chromium contents are affected too. These*
49 *processes are considered to occur near closed-system conditions. Late lawsonite overgrowths with*
50 *higher Sr contents are interpreted as reflecting system opening in the veins, yet sometimes observed in*
51 *the schists. Strontium isotopic ratios measured on separated lawsonite crystals and on bulk rocks also*
52 *show very large spreads. This indicates that the system did not equilibrate during blueschist-facies*
53 *metamorphism, due to bulk rock heterogeneities during deposition and limited mineral reactivity at the*
54 *local scale. Strontium isotopic ratios do not correlate with Sr content which rules out limestone*
55 *devolatilization as the main driver of lawsonite overgrowths. In places, lawsonite is far more radiogenic*
56 *than the host rocks, indicating either the destabilization of local Rb-rich phases (such as mica, illite and*
57 *smectite) or the infiltration of externally-derived fluids. The overall compositional evolution of lawsonite*
58 *records incremental system opening. However, lawsonite offers no evidence of large-scale mass transfer*
59 *and external fluid influx in the Schistes Lustrés complex, even though the vein network is abundant and*
60 *extends over time.*

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INTRODUCTION

66 Sediments undergoing subduction contain large amounts of H₂O (up to 7 wt. % entering the trench
67 globally, Plank and Langmuir, 1998; Plank, 2014) and release water via metamorphic reactions through
68 burial (e.g., Schmidt and Poli, 1998; Hacker et al., 2003; Van Keken et al., 2011). At depth, fluids play
69 a major role as their migration induces dramatic changes in the mechanical and chemical properties of
70 rocks. The amount of fluids (e.g., H₂O, CO₂) produced can be estimated (e.g., via thermodynamic
71 modeling as by Hacker and Abers, 2004; Baxter and Caddick, 2013; Ague and Nicolescu, 2014; Kleine
72 et al., 2014; Piccoli et al., 2021, among many others), but pathways and scales of fluid migration remain
73 hard to assess, especially in sedimentary lithologies where alternating horizons show hugely variable
74 permeability, and where connection of vein networks may be transient and poorly recorded. Trace
75 elements, fluid inclusions and/or isotopic signatures of minerals growing in subducted rocks,
76 metamorphic veins or shear zones are useful tracers of fluid migration during prograde metamorphism.
77 Lawsonite is of particular interest as it is stable over an extended pressure-temperature range (Schmidt
78 and Poli, 1994), generally growing as prisms, and can be locally abundant. Lawsonite is also intimately
79 linked to both the water and carbon cycles as it is a major H₂O host (~12 wt. %) and its formation
80 reactions involve devolatilization and dissolution/crystallization of carbonates (Nitsch, 1972; Cook-
81 Kollars et al., 2014; Lefeuvre et al., 2020).

82 Lawsonite has been shown to be remarkably stable around its ideal composition (CaAl₂Si₂O₇(OH)₂·H₂O)
83 regardless of pressure-temperature conditions (e.g., Okamoto and Maruyama, 1999), but it can host
84 various trace elements. Lawsonite incorporates Pb, Sr and rare earth elements (REE), especially light
85 rare earths (LREE), preferentially to other minerals in classical assemblages (e.g., reviews in Fornash et
86 al., 2019; Whitney et al., 2020; Kang et al., 2022). Previous studies of the minor and trace element
87 composition of lawsonite in mafic blueschists, eclogites, and metasomatic rocks have shown that
88 variations of lawsonite composition may reflect reactions between trace element-rich minerals and/or
89 changes in fluid compositions, with implications for element cycling in shallow to deep parts of
90 subduction zones (e.g., Tribuzio et al., 1996; Ueno, 1999; Spandler et al., 2003; Tsujimori et al., 2006;
91 Martin et al., 2011, 2014; Vitale Brovarone et al., 2014; Vitale Brovarone and Beyssac, 2014; Fornash
92 et al., 2019; Whitney et al., 2020; Kang et al., 2022).

93 Studies on metasedimentary lawsonite are scarce (Martin et al., 2014; Whitney et al., 2020), although
94 lawsonite is abundant in blueschist-facies calcschists such as in the Schistes Lustrés (SL) complex of
95 the Western Alps, where lawsonite locally represents up to 40 vol.% of the metasediments (Agard et al.,
96 2001; Lefeuvre et al., 2020; Manzotti et al., 2021). The SL units correspond to fragments of oceanic
97 lithosphere successively buried along a cold geothermal gradient (~ 8°C/km, Agard et al., 2001; Agard,
98 2021; Herviou et al., 2022) during Alpine subduction, exhumed sliced from the downgoing plate at

99 various depths. The SL provide a well-exposed example of a deep accretionary complex (Platt, 1986;
100 Marthaler and Stampfli, 1989; Agard et al., 2009; 2018; Agard, 2021). Metasediments are volumetrically
101 dominant and crop out continuously over large distances, allowing studies of the production,
102 composition and behavior of fluids in subducted sediments (Agard et al., 2000; Bebout et al., 2013;
103 Herviou et al., 2021). Oxygen and carbon isotopic data in the metasediments suggest that most of the
104 fluid present during subduction was locally-derived (Henry et al., 1996; Bebout et al., 2013; Cook-
105 Kollars et al., 2014; Jaeckel et al., 2018; Epstein et al., 2020), consistently with the composition of fluid
106 inclusions in carpholite and lawsonite-bearing veins (Agard et al., 2000; Herviou et al., 2021; see also
107 Raimbourg et al., 2018; Lefeuvre, 2020). Therefore, metasediments of the SL complex mostly behaved
108 as a rock-buffered system, with volumetrically large but transient and restricted fluid migration. External
109 fluid infiltration was limited, but the scales of fluid migration and element mobility remain poorly
110 known. Lawsonite is found in these rocks in pelitic layers and in a network of high-pressure veins,
111 locally with carpholite (Agard et al. 2000, 2001; Vitale Brovarone et al., 2020; Lefeuvre et al., 2020;
112 Herviou et al., 2021, 2023; Manzotti et al., 2021). Lawsonite is thought to grow during prograde
113 metamorphism of the SL units, with veins providing evidence for incremental lawsonite growth during
114 crack-seal events (Herviou et al., 2023). Do trace element compositional variations in lawsonite record
115 variations in fluid signature along the prograde path?

116 In light of the very large variability of minor and trace element composition of lawsonite (Fornash et al.,
117 2019; Whitney et al., 2020; Kang et al., 2022), this study provides a petrological model to refine the
118 crystallization history of lawsonite and to shed light over the scale of element mobility during prograde
119 and peak metamorphism at blueschist-facies conditions. To address these questions, lawsonite crystals
120 from the SL calcschists and high-pressure veins have been analyzed for major and trace elements,
121 coupled to bulk rock analyses and strontium isotopic analyses on bulk samples and on lawsonite
122 separates.

123 **GEOLOGICAL SETTING AND PREVIOUS WORK**

124 **The Schistes Lustrés complex in the Western Alps**

125 The Western Alps result from an east-dipping, slow and cold subduction ($\sim 8^\circ\text{C}/\text{km}$ at a
126 maximum of 2 cm/year) of the Valais and Liguro-Piemont slow-spreading oceans below Adria/Apulia
127 microcontinents (Le Pichon et al., 1988; Lagabrielle and Cannat, 1990; Agard et al., 2001; Lapen et al.,
128 2003; Schmid et al., 2017). Subduction of these slow-spreading oceanic domains lasted around 60 Myr
129 (Stampfli et al., 1998; Agard, 2021) and was followed by continental subduction and collision (e.g.,
130 Mendes et al., 2023).

131 Remnants of Liguro-Piemont oceanic lithosphere are now exposed in the internal domain of the Western
132 Alps and form a nappe-stack known as the Liguro-Piemont domain or SL complex (Caron, 1974;
133 Lagabrielle and Lemoine, 1997; Agard et al., 2001, 2002; Agard, 2021; Herviou et al., 2022) owing to
134 their characteristic sedimentary features (Fig. 1a; for more details on the overall Alpine structure, see:
135 Coward and Dietrich, 1989; Lagabrielle and Lemoine, 1997; Rosenbaum and Lister, 2005; Handy et al.,
136 2010). Former oceanic sediments (marls, shales and limestones, e.g., Lemoine, 2003) correspond to an
137 initial ~200-400 meter thick sequence of Upper Jurassic to Upper Cretaceous pelagic and hemipelagic
138 seafloor deposits (De Wever and Caby, 1981; Lemoine et al., 1984; Lemoine and Tricart, 1986; Deville,
139 1986; Deville et al., 1992; Lemoine, 2003). Minor and variable amounts of mafic and ultramafic rocks
140 are also found in the SL complex (e.g., Lagabrielle et al., 2015; Herviou et al., 2022).

141 During subduction, fragments of the Liguro-Piemont oceanic lithosphere were successively scraped off
142 the downgoing plate, stacked at depths and exhumed to form a deep accretionary complex built by
143 progressive underplating (Platt, 1986; Marthaler and Stampfli, 1989; Agard et al., 2009; 2018; Agard,
144 2021; Agard and Handy, 2021; Plunder et al., 2012., Herviou et al., 2022). Several units corresponding
145 to independent subduction slices were identified in the SL complex (Lagabrielle, 1987; Fudral, 1996;
146 Tricart and Schwartz, 2006; Lagabrielle et al., 2015). Herviou et al. (2022) reappraised the overall
147 structure of the complex, highlighting a trimodal distribution of units with near-continuous eastward
148 increase in peak burial conditions from the Liguro-Piemont Upper units (LPU; 320-400°C – 1.2-1.9
149 GPa) to the Middle units (LPM; 415-475°C – 1.7-2.2 GPa) and to the Lower units (LPL; 500-580°C –
150 2.2-2.8 GPa; Fig. 1c; for peak metamorphic conditions through the domain see also Agard et al., 2001;
151 Beyssac et al., 2002; Gabalda et al., 2009; Plunder et al., 2012; Angiboust et al., 2012; Schwartz et al.,
152 2013; Locatelli et al., 2018). The LPU and LPM units metamorphosed under blueschist-facies conditions
153 are dominated by sediments (> 90 %), whereas the LPL units metamorphosed at eclogite-facies
154 conditions are richer in mafic-ultramafic rocks (> 40 %; Herviou et al., 2022).

155 **Lawsonite-bearing calcschists on the Cottian Alps transect**

156 This contribution focuses on lawsonite-bearing calcschists from the LPU unit of the Cottian
157 Alps (Lago Nero slice; Polino et al., 2002; Herviou et al., 2022) and more specifically on the Strada
158 dell'Assietta transect (Caron, 1974; Lefeuvre et al., 2020; Fig. 1b) between Fraiteve and the Assietta
159 pass, along a ~15 km SW-NE traverse in the French-Italian Cottian Alps (Fig. 1b, c; Caron, 1974; Polino,
160 1984). Metasediments recorded four deformation stages described by Agard et al. (2001; 2002) and
161 Ghignone et al. (2020). Kilometer-scale folding associated with thin-skinned stacking during blueschist-
162 facies peak burial (D1) is dated at 62-55 Ma. These early fabrics have been largely erased by a D2 event
163 (51-45 Ma) with top-to-the-east shear deformation patterns associated with early exhumation under
164 blueschist-facies, and by subsequent west-vergent shear zones (D3) associated with greenschist facies

165 conditions (38-35 Ma; Agard et al., 2002). Late normal faults characterize the final ductile-brittle tran-
166 sition during late exhumation. The metasediments along the transect are composed of alternations of
167 carbonates and pelitic horizons (Fig. 1d, e), which are of varying thickness and proportions, ranging
168 from pelites (described as type 1 calcschists by Lefeuvre et al., 2020, Fig. 1d, e) to thick carbonate beds
169 almost lacking pelitic horizons (type 5 in Lefeuvre et al., 2020, Fig. 1d, e).

170 Lawsonite is the dominant hydrous phase stable at peak conditions in LPU metasediments, but
171 its distribution is very heterogeneous. Lawsonite growth has been described as ‘massive’ by Lefeuvre
172 et al. (2020) because it amounts to about half the volume of large outcrops in the LPU units in type 3
173 calcschists, but it is restricted to fine carbonate-metapelite mixtures (also ubiquitous in type 3
174 calcschists) that allow reaction between Ca, Al, Si and H₂O. Four textural occurrences of lawsonite were
175 identified by Lefeuvre et al. (2020) and were labelled from A to D (LwsA, LwsB, LwsC, LwsD) without
176 implying chronological order. In thin pelitic horizons, lawsonite occurs as millimetric black prisms
177 coined LwsA, by far the most abundant lawsonite type. Cream-colored lawsonite fibers labelled LwsB
178 are found in decimeter to meter-long quartz-rich veins elongated parallel to vein walls and recording
179 incremental crack-seal growth (Fig. 2f; Herviou et al., 2023). LwsC-bearing centimetric tensile veins
180 contain cream-colored lawsonite fibers in textural equilibrium with ankerite and quartz, all growing
181 perpendicular to vein walls. LwsA in the schist is interpreted as the product of the prograde reaction
182 chlorite + aragonite + kaolinite = lawsonite + ankerite + quartz + H₂O, beginning at conditions possibly
183 as low as 0.4 GPa/180 °C (Lefeuvre et al., 2020). Lawsonite veins likely formed all along the prograde
184 path, in response to pressure-solution processes. While LwsC veins likely opened as single stage, short-
185 lived tensile events, LwsB incremental crack-seal fibers may have recorded longer-lived events along
186 the prograde path (Lefeuvre et al., 2020; Herviou et al., 2023). LwsD is restricted in pelitic layers and
187 occurs as white rosette-like prisms in the schist (Fig. 2g). Table 1 presents the mineralogy, rock and
188 lawsonite types for all samples.

189 **LAWSONITE OCCURRENCE, TEXTURE AND ASSOCIATED MINERALOGY**

190 This study focuses on three representative lawsonite-bearing outcrops along the Strada
191 dell’Assietta transect (Fig. 1): Desertes, Triplex and Assietta. The Fraiteve location also offers an
192 interesting outcrop of the least metamorphosed rocks of the series. At the 200m long Mt. Triplex
193 outcrop, lawsonite is particularly abundant in both schist and veins (Fig. 2). These type 2 (and type 3 to
194 a lesser extent) calcschists are made of pelitic and carbonate-rich layers alternating every few
195 centimeters, highly folded and affected by D2-related penetrative shear bands. D2 shear structures are
196 cross-cut by later west-vergent D3 shear bands (Fig. 2a, b).

197 LwsA crystallized in the thin pelitic layers as millimetric to centimetric black prisms enriched
198 in organic matter (OM). LwsA is systematically associated reddish oxidized ankerite found at the

199 interface between pelitic and carbonate-rich beds (Figs. 2b, c, 3a). Samples Tpx-A1, Tpx-A2 and Tpx-
200 A3 (Table 1) contain large amounts of centimetric LwsA crystals reaching 40 vol. % of the whole rock
201 (Fig. 3a, b), and are hereafter referred to as ‘host calcschists’ (Figs. 2c, 3). LwsA in pelite-rich layers is
202 closely associated with ankerite and quartz (Fig. 3c, d, e, f). Fine mixtures of phengite, chlorite and
203 calcite are surrounding LwsA crystals and interpreted as the retrograde assemblage of lawsonite
204 destabilization (Fig. 3c, e; Sicard-Lochon and Potdevin, 1986). Accessory minerals are scarce in the
205 host calcschists; tiny rutile needles were identified in lawsonite, and apatite occurs in one sample (Fig.
206 3f; Tpx-A2). Optical microscopy reveals complex growth patterns for LwsA. Differences in interference
207 colors under crossed polars clearly indicates the presence of hourglass zoning (Fig. 3b, e). In some LwsA
208 crystals, organic matter (OM) inclusions help to illuminate the zoning and identify the hourglass shapes
209 (Fig. 3b). Such chemical zoning has been previously described in metasedimentary lawsonite (Vitale
210 Brovarone et al., 2014; Fornash et al., 2019; Whitney et al., 2020). LwsA cores include OM in trails
211 aligned along crystal faces (Fig. 3c, d), folded and rotated compared to the surrounding schistosity, or
212 highlighting hourglass zoning (Fig. 3c, e). These OM-rich cores are generally rimmed by OM-free
213 lawsonite (Fig. 3b, c, e), suggesting a series of discrete growth events of LwsA (see Rice and Mitchell,
214 1991 for details about textural zoning and carbon-rich inclusion bands).

215 Rare white crystals of prismatic lawsonite, without preferential orientation, are observed in
216 pelitic layers in the vicinity of LwsB-bearing veins. These have been classified as a fourth lawsonite
217 type: LwsD. LwsD is present in sample Tpx-D1 (Fig. 2e, g) collected 1 meter away from LwsB-bearing
218 Tpx-B1, and in Ft-D1 collected in Fraiteve. As with LwsA, LwsD crystals are locally associated with
219 ankerite and quartz (Fig. 2g), and partially retrogressed by phengite, chlorite and calcite. LwsD contains
220 little OM inclusions compared to LwsA.

221 Veins are abundant, especially at Mt. Triplex where they have been estimated to make up to
222 30 % of the outcrops (Herviou et al., 2023). LwsB-bearing veins are decimeter to meter-long, parallel
223 to the main D1/D2 schistosity, ductility deformed (Fig. 2b, f). LwsB-bearing veins collected at Triplex
224 and at Desertes (Table 1, Tpx-B1, Tpx-B2, Ds-B2, Ds-B3) are composed of quartz and fibers of cream-
225 colored LwsB often fractured and systematically elongated parallel to vein walls (Fig. 4a-c),
226 highlighting their formation by dilatational shearing (Herviou et al., 2023). Ankerite crystals were found
227 at the selvages of some veins but are rarely observed in association with LwsB (Fig. 4a). LwsB crystals
228 do not contain OM but are often pseudomorphed into a mixture of phengite and chlorite (Fig. 4b, c).

229 LwsC-bearing veins are millimetric to centimetric tensile veins crosscutting the schistosity.
230 These veins occur in the vicinity of fold hinges, close to LwsB-bearing veins, and are only found where
231 LwsA is widespread in the schist. LwsC fibers are associated with ankerite and quartz, all growing
232 perpendicularly to the vein selvages (Fig. 4d-f). LwsC-bearing veins are easily missed as they are small,
233 scarce, largely sheared and poorly preserved. In most veins, LwsC fibers are pseudomorphed into

234 phengite, chlorite and calcite (Fig. 4f). The freshest LwsC veins, selected for the geochemical study,
235 were found in the vicinity of the ‘multiquartz aggregates’ described below. Herviou et al. (2023) have
236 shown that the emplacement of the lawsonite vein (LwsB and LwsC) network is continuous and
237 incremental, interpreted as the fossil record of slow slip events. The relative chronology between these
238 events is therefore complicated to establish as the formation of these veins can be related to multiple
239 processes.

240 In places, lawsonite-bearing veins are clustered and intimately folded and sheared with the
241 surrounding schist, resulting in intricate patterns with schistose fragments embedded in high-pressure
242 veins with abundant quartz (Figs. 2d, e). These objects, referred to as ‘multiquartz aggregates’, were
243 sampled both in Triplex (Tpx-AB5 and Tpx-AC4) and Assietta pass (As-AB1 and As-AC2). Lawsonite
244 in these schistose fragments is therefore in direct proximity (< 1 cm) to lawsonite growing in the
245 embedding veins. Lawsonite in schistose fragments (noted LwsA*) forms black prisms including OM
246 trails, similar to those observed elsewhere in the schist, crystallizing with quartz and ankerite (Sup. Mat.
247 1a-c). Veins contain elongated lawsonite fibers similar to LwsB (labelled LwsB*; Figs. 2e, 5a-d),
248 showing multiple growth steps synchronous with deformation (Figs. 2e, Sup. Mat. 1a, c). LwsB* crystals
249 are associated with quartz and are very often pseudomorphosed into phengite and chlorite (Sup. Mat.
250 1c, d). Some multiquartz aggregates display LwsC tensile veins (Figs. 2d, Sup. Mat. 1f), generally at the
251 interface between LwsB*-bearing veins and embedded schistose fragments (Fig. 2e). LwsC fibers are
252 associated with quartz and oxidized ankerite (Sup. Mat. 1g) and are well-preserved compared to LwsC
253 in most veins elsewhere and were therefore selected for analysis (Tpx-AC4, As-AC2).

254 ANALYTICAL METHODS

255 Analytical strategy (Table 1)

256 Major and trace element analysis have been carried out in lawsonite crystals to study whether
257 lawsonite and its zoning patterns record evolving length-scales of element mobility and fluid circulation.
258 Thick sections were made on 20 lawsonite-bearing samples (Table 1) to perform laser ablation ICP-MS
259 and X-ray chemical maps to measure lawsonite trace element content and distribution.

260 Ten rock samples were selected along the Strada dell’Assietta transect (Fig. 5a) for bulk
261 measurements. Nine of them are calcschists, representative of the types identified by Lefeuvre et al.
262 (2020; Fig. 2a, b). One pelitic (Desertes; type 1) and one carbonate-rich calcschist (Gleise; type 4) were
263 selected to represent end-member lithologies, where lawsonite is not abundant. Most samples (7) were
264 selected in type 2 and type 3 calcschists (Fig. 2a, b) in which lawsonite is widespread (Lefeuvre et al.,
265 2020). In order to identify potential interactions with mafic rocks, one ~10 m long basaltic pillow breccia

266 body embedded in the calcschists was sampled at the Bourget site (Bg-mafic; 20 m away from the
267 Bourget calcschist sample) and analyzed.

268 Strontium isotopic ratios were measured on mineral separates and selected bulk samples
269 because Sr is commonly enriched in both carbonates and lawsonite (Enami, 1999; Martin et al., 2014;
270 Hara et al., 2019; Whitney et al., 2020) and rather mobile in aqueous fluids, therefore useful to study
271 fluid-rock interactions (see Hara et al., 2018).

272 **Bulk-rock geochemistry (Table 2)**

273 Samples were finely crushed ($< 2\mu\text{m}$) in an agate mortar. Sample powders were weighed and
274 heated for 1h at 1100°C to ensure destruction of OM. Powders were dissolved at 80°C in closed beakers
275 with HNO_3 and HF overnight. For analysis of major elements, $\text{B}(\text{OH})_3$ was introduced quickly after
276 opening the beakers to avoid loss of silica. Further dilution was carried out with 2% HNO_3 before
277 analysis. Major and trace element compositions were analyzed at ALIPP6 (ISTeP, Sorbonne Université,
278 Paris) using an ICP-OES spectrometer (Agilent 5100 HVDV) for major elements and an ICP-MS/MS
279 (Agilent 8800) for trace elements. Calibration was made using a set of international and in-house
280 standards (RGM-1, BIR-1, AGV-2, LIP, M77-2, GSN, COS, BHVO-2). To characterize the major
281 element budget of these rocks, 10 oxides were measured (SiO_2 , TiO_2 , Al_2O_3 , FeO , MnO , MgO , CaO ,
282 Na_2O , K_2O and P_2O_5). A total of 37 minor and trace elements were selected for this study (Li, Be, Sc,
283 V, Cr, Co, Ni, Cu, Zn, Ga, As, Rb, Sr, Y, Zr, Nb, Cs, Ba, Ta, Pb, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy,
284 Ho, Er, Tm, Yb, Lu, Hf, Th and U).

285 **Major element analysis and chemical mapping of lawsonite (Tables 3, 4, 5, 6)**

286 Major element analysis of lawsonite crystals has been carried out at CAMPARIS (Sorbonne
287 Université, Paris) with a CAMECA SX-Five electron microprobe and the data reduction method of
288 Pouchou and Pichoir (1991). Analytical conditions were 15 kV-10 nA in WDS mode for analysis of
289 major elements. Conditions were changed to 25 kV-100 nA when measuring minor elements (Sr, Ti,
290 and Cr). Fe_2O_3 (Fe), MnTiO_3 (Mn, Ti), diopside (Mg, Si), Cr_2O_3 (Cr), SrSi (Sr), orthoclase (Al, K),
291 anorthite (Ca) and albite (Na) were used as standards for calibration of elements in parentheses.
292 Quantitative mapping was performed using the same analytical conditions in WDS mode. Qualitative
293 element mapping was also carried out with a SUPRA-55VP SEM at ISTeP (Sorbonne Université, Paris)
294 in energy-dispersive spectroscopy (EDS) mode at 15 keV acceleration voltage, 30 nA beam current with
295 a dwell time of $256\ \mu\text{s}$, analyzing each area up to 10 times for increased signal/noise ratio.

296 **Trace element analysis of lawsonite (Table 7)**

297 Trace element lawsonite concentrations were measured at ALIPP6, using a laser ablation ICP-
298 MS/MS (Teledyne Analyte G2 193 nm ArF excimer laser ablation system coupled to an Agilent 8800
299 ICP-MS/MS) on polished thick sections (100 μm). Lawsonite crystals were ablated for 60 s (frequency
300 of 8 Hz) in a He atmosphere using a 40 μm beam and a laser fluency of 3.54 J/cm². Background was
301 measured 18 s before and after sampling ablation. Standardization was carried out using BCR2-G
302 international glass standard, and ²⁷Al as internal standard obtained from electron microprobe analyses.
303 Instrument drift and accuracy were controlled with repeated analyses of the international standards
304 ATHO-G and BHVO2-G. Every ablation pattern was verified with an in-house spreadsheet to check for
305 absence of impurities and/or inclusions during ablation (89 out of 400 analyses were discarded).

306 **Strontium isotopic ratios (Table 8)**

307 Lawsonite strontium isotopic ratios were determined at the Boston College Center for Isotope
308 Geochemistry using an IsotopX Phoenix Thermal Ionization Mass Spectrometer (TIMS). First,
309 lawsonite crystals were separated from the bulk rock after crushing and sieving (see Sup. Mat. 2).
310 Mineral fractions between 500 μm and 125 μm were separated using sodium polytungstate (density of
311 2.9 g.cm⁻³) allowing isolation of the heavy fraction containing lawsonite, ankerite and oxides. Lawsonite
312 grains were separated using magnetic separation, cleaned in ultrasonic baths and handpicked before
313 dissolution for TIMS analysis. Dissolution followed a four-step protocol (Sup. Mat. 2). Lawsonite
314 separates were partially dissolved in 2mL of glacial acetic acid in an ultrasonic bath to remove remaining
315 carbonates. This first step was repeated twice to purify the lawsonite fraction. Samples were
316 subsequently dissolved in HF-HNO₃-HCl. Cleaned lawsonite fractions were attacked with 100 μL of
317 HF together with 2 mL of HNO₃ at 120°C overnight. After 12 hours, the sample was dried down. The
318 same step was repeated with 2 mL of HNO₃ at 120°C. Dissolved samples were prepared for chemistry
319 in 2 mL of HCl at 120°C. Strontium was separated using 100 μL micro-columns and pre-cleaned Sr-
320 spec resin. Approximately 100 ng of Sr were loaded onto Re filaments with a TaF activator solution.
321 Samples were analyzed using a multidynamic method, and repeated analysis of Sr standard SRM987
322 during the duration of the project gave 0.710240 ± 0.000010 (n = 5; 2sd), which is within the long-term
323 average of 0.710242 ± 0.000005 (n=114; 2sd).

324 This procedure has been applied to bulk rock samples (without preliminary partial dissolution).
325 In total, 7 bulk rocks and 17 lawsonite separates have been analyzed (8 LwsA, 6 LwsB, 1 LwsC and 2
326 LwsD samples). These same dissolved lawsonite samples were also aliquoted for ICP-MS analysis to
327 determine the Sr and Rb concentrations and ⁸⁷Rb/⁸⁶Sr ratios reported in Table 8. Uncertainty in ⁸⁷Rb/⁸⁶Sr
328 is 2.8 % RSD based on repeat analysis of standard solutions.

329

330 **RESULTS**

331 **Bulk rock geochemistry**

332 Bulk rock compositions reflect the petrographic variability of the metasediments along the
333 Strada dell'Assietta transect (Fig. 1d-e; Table 2): compositions evolve gradually in ACF (Al_2O_3 -CaO-
334 FeO; Fig. 5a) space between pelitic horizons (Desertes, lithology type 1 in Fig. 1d-e) and carbonate-rich
335 endmembers (Gleise, lithology type 4 in Fig. 1d-e). These rocks exhibit similar FeO contents around
336 20 % in ACF except for the mafic sample (Bg-mafic) described below. CaO and Al_2O_3 contents are
337 inversely correlated (Fig. 5b). Pelitic horizons (Desertes; type 1) contain > 25 wt. % of Al_2O_3 and < 10
338 wt. % of CaO, whereas carbonate-rich calcschists (Gleise; type 4) contain up to 58 wt. % of CaO and
339 ~5 wt. % of Al_2O_3 . Type 2 calcschists contain between 15 and 25 wt. % of Al_2O_3 and 10-25 wt. % of
340 CaO, whereas more carbonated type 3 calcschists contain 7 to 15 wt. % of Al_2O_3 and 25 to 50 wt. % of
341 CaO. At first order, TiO_2 and Ce are anti-correlated to CaO content (as for Al_2O_3), but Sr is positively
342 correlated to CaO content (Fig. 5b). TiO_2 is a major component of the SL calcschists (ranging between
343 0.1 and 1.9 wt. %) and is more concentrated in type 1 pelitic lithologies (> 1 wt. %) than in the more
344 carbonated type 2 (0.5-1 wt. %) and type 3-4 (< 0.5 wt. %) rocks. Most of the calcschists analyzed here
345 contain less than 100 ppm of Sr (types 1, 2) but Sr reaches 180 ppm in carbonate-rich type 4.

346 The most pelitic type 1 and type 2 calcschists are generally the most concentrated in LREE (Ce >
347 100 ppm, Fig. 5c) while the lowest concentrations are found in type 4 calcschists (Ce < 25 ppm, Fig.
348 5c). Chondrite-normalized REE patterns (Fig. 5c) show similarly decreasing trends from LREE to
349 HREE for all calcschists, with La/Yb ratios around 20 and negative Eu anomalies (except for the Bourget
350 sample). These patterns compare well with those of Corsican calcschists metamorphosed around 500-
351 550 °C and ~ 2.3 GPa reported by Piccoli et al. (2016). However, Al_2O_3 -rich calcschists from the present
352 study (types 1 to 3) are one order of magnitude richer in REE (Fig. 5c).

353 Compared to the calcschists described above, the mafic sample from the Bourget locality (Bg-
354 mafic: metamorphosed pillow breccia) shows higher contents in FeO (up to 55 % in the ACF, Fig. 5a),
355 MgO (~15 wt. %), Na_2O (~3 wt. %; Table 2), TiO_2 (~2 wt. %; Fig. 5b) and enrichments in V, Cr, Co
356 and Ni (Table 2), elements typically enriched in mafic rocks. Bg-mafic is depleted compared to
357 calcschists in CaO (5 wt. %), HREE, and in most LILE such as Sr (~10 ppm) Rb, Ba, Cs and Pb (Fig.
358 5c; Table 2).

359 **Lawsonite composition**

360 As major elements of the different lawsonite types are very similar (see tables 3, 4, 5, 6), we
361 focused on the trace element contents. In the following, chemical maps (obtained by electron

362 microscopy) are presented next to corresponding extended trace element patterns from LA-ICP-MS spot
363 analyses for each lawsonite type.

364 *Univalent cations and white mica contamination*

365 Even though preservation of lawsonite crystals has been a prevalent criterion for selection before
366 laser ablation, and each lawsonite ablation pattern has been carefully inspected to detect contamination
367 from inclusions, contamination remains apparent. A strongly positive correlation between K content and
368 Na, Ba, Rb and Cs contents is observed (Sup. Mat. 3). These elements are classically enriched in white
369 mica, in substitution of K. Because lawsonite is generally devoid of K, these specific enrichments are
370 attributed to white mica contamination via minute inclusions in lawsonite (see also Whitney et al., 2020).
371 Therefore, interpretation of analytical results does not incorporate Na, K, Ba, Rb and Cs in the following.
372 It is also noteworthy that Li appears to show a positive correlation with K, yet much weaker than for
373 other univalent cations (Sup. Mat. 3). Significant white mica contamination appears limited to these
374 elements as no other correlation with K is observed.

375 *Lawsonite composition in host calcschists (LwsA)*

376 Five areas have been analyzed in a pelitic schist (sample Tpx-A1). They contain dark LwsA
377 crystals with hourglass zoning, easily recognized on Ti maps (Fig. 6a, c). Crystals may exhibit both
378 sides of the hourglass (Fig. 6c) or only one (Fig. 6a) due to their orientation. In chemical maps (Fig. 6a,
379 c), hourglass zoning is faintly observed for Ce, with enriched domains similar to Ti (blue spots 30-32
380 and 34-35 in C1; spots 13, 14, 16 and 18 in C4) whereas Al is slightly depleted in these domains (C1,
381 Fig. 6a). Ce also shows enrichment in angular sectors in the center of lawsonite crystals, again anti-
382 correlated to Al (Fig. 6a) but not linked to specific Ti enrichment (close to spots 37 and 29 in Fig. 6a,
383 close to spot 15 of Fig. 6c).

384 Ti-rich hourglass domains (blue dots 30-32 and 34-35; Fig. 6a, b) show the highest
385 concentrations in Ti, Zr and Hf and the lowest concentrations in Sr, Y and Mn. REE patterns also show
386 large variations, in terms of slope and absolute values, whether located within or without the Ti-enriched
387 domains. Even though barely seen on elemental maps (Fig. 6a, c), core to rim variations appear
388 superimposed to the hourglass domain, in particular in Ti-rich domains (e.g., blue curves 30-32 and 34-
389 35 in Fig. 6b, blue and green curves in Fig. 6d): LREE content decreases from core to rim, while HREE
390 values remain almost constant, which tends to flatten the REE pattern slope and generate spoon-shaped
391 patterns. Ti also tends to decrease from core to rim in the Ti-rich domain (Fig. 8a). In the crystal core
392 (spot 15 on Fig. 6c), the Ce-enriched Ti-depleted domain shows the highest U, Zr, Y, Hf, REE, Fe, Mn
393 and Cr contents.

394 The LwsA crystal investigated in the third area (Fig. 6e) shows intricate zoning as tentatively
395 schematized (Fig. 6f). Overall, Ti and Sr maps (Fig. 6e) show a lawsonite core (spots 1 and 8) rimmed
396 by Ti-enriched lawsonite, itself rimmed by a Ti-poor, Sr-rich lawsonite border (green and blue zones in
397 Fig. 6f, respectively). In the lawsonite core, hourglass zoning is observed with two domains: Ti-enriched
398 (spots 01 and 08) and Sr-enriched (depleted triangle-shaped domain immediately below). Additionally,
399 the Ti-rich domain shows variable Ti content (Fig. 6e, f, g), the lowest contents in Zr, Hf and Ni, and
400 low LREE contents (spots 01 and 08, Fig. 6g). The enclosing lawsonite (spots 2 to 9) is generally Ti-
401 rich and Sr-poor, with much dispersion and patchy domains. LA-ICP-MS analyses in this zone (spots
402 02, 03, 07, 09) show variable LREE contents, but almost similar HREE contents, resulting in flattened
403 and curved patterns like those observed earlier (Fig. 6b, d). The Sr-rich Ti-poor border (spots 10 and 12)
404 rimming LwsA (Fig. 6f) is enriched in Y and HREE, showing spoon-shaped REE patterns (Fig. 6g).

405 *Lawsonite composition in quartz-rich veins (LwsB)*

406 Cream-colored LwsB crystals have more homogeneous composition than LwsA crystals. None
407 of the investigated LwsB crystals displays hourglass zoning. Other zoning patterns have been identified
408 in LwsB crystals (Fig. 7). The first Ti map (Fig. 7a) features angular Ti-rich domains in crystal cores
409 (LA-ICP-MS spots 38 to 41) surrounded by Ti-depleted rims (spots 42 and 43, Fig. 7a, b). These
410 differences are highlighted in the interpretative sketch and a second map for Ti and Sr (Fig. 7c); it is
411 tempting to interpret these as successive generations of lawsonite. The Ti-poor crystals are enriched in
412 U, Sr, Y and HREE compared to the Ti-rich crystals (Fig. 7b).

413 *Lawsonite composition in multiquartz aggregates (LwsA* & LwsB*)*

414 Compositions of LwsA* and LwsB* from multiquartz aggregate Tpx-AB5 from Triplex are
415 provided in Supplementary material 4. LwsB* shows the same features as LwsB with Ti-rich cores (Sup.
416 Mat. 4a) and Ti-depleted rims corresponding to high Sr contents (Sup. Mat. 4b). The composition and
417 distribution of the other trace elements (Sup. Mat. 4b) compare also well with LwsB.

418 In the schistose fragments included in veins, dark LwsA* cores often show pale rims (Sup. Mat.
419 4d) enriched in Ti and Cr (Sup. Mat. 4c) but not in Ce (spots 2 to 5, Sup. Mat. 4c). LwsA* cores (spots
420 06 to 09) show heterogeneous and patchy Ti-zoning, they are generally enriched in most LILE (and
421 HFSE to a lower extent) but depleted in Pb and Sr (Sup. Mat. 4e).

422 *Minor and trace elements in lawsonite types along the Strada dell'Assietta transect*

423 In situ lawsonite compositions (Fig. 8, normalized to C1 chondrite, McDonough and Sun, 1995)
424 are provided in Table 7 for each lawsonite type and each locality.

425 Dark LwsA crystals in host calcschist from Triplex area (Tpx-A1, Fig. 8b) display high values
426 for U, Th and REE. Most LILE (K, Cs, Rb, Ba, Pb, Sr) display large variations between samples, sharing
427 high Pb and Sr contents (10 to 100 times chondritic values) but low contents in the other LILE (down
428 to 10^{-3} chondritic values). The dispersion is much lower for Pb and Sr than for other LILE. High field
429 strength elements (HFSE; Zr, Ti, Hf) are also hugely variable: Zr content varies over four orders of
430 magnitude, Hf and Y over three, Ti over two. These lawsonite crystals show peculiar REE patterns, with
431 marked enrichment in LREE content for some, leading to a four order of magnitude dispersion. HREE
432 are also scattered but enriched between 10 and 1000 times the chondritic values. Two signatures appear:
433 LREE-enriched LwsA with a negative slope from LREE to HREE (La/Yb \sim 100), and LwsA with flat
434 to positive REE profiles, with an inflection point around Gd. Metallic elements (Fe, Mn, Cr and Ni) are
435 generally low with considerably smaller dispersion.

436 LwsB in isolated quartz-bearing veins from Triplex (Tpx-B1, Fig. 8c; Tpx-B3, Fig. 8d) and
437 Desertes (Ds-B3, Fig. 8a) shows very similar patterns for most elements. LwsB shows less dispersion
438 than LwsA, except for LILE in Tpx-B1 (Fig. 8c) that compare well with those of LwsA (Fig. 8b). U and
439 Th contents are either comparable to those in LwsA (Fig. 8a-d) or lower (between 10- and 200-times
440 chondritic values in Tpx-B1). Pb contents are comparable to those of LwsA, and Sr is slightly higher in
441 LwsB (up to 1000 times the chondritic values). For HFSE, Zr and Hf contents are lower than in LwsA
442 (10^{-1} to 10^{-2} chondritic values) while Ti is similar (1-10, Fig. 8c). However, LwsB from Desertes shows
443 higher LILE and HFSE than those from Triplex (Fig. 8a, c, d). REE patterns of LwsB show a slightly
444 negative slope from LREE to HREE, as commonly observed in lawsonite (see compilation of Whitney
445 et al., 2020). These patterns are consistent with some of the LwsA patterns in the host calcschist (Fig.
446 8b). A small positive anomaly in Eu is observed in sample Tpx-B1 only (Fig. 8c). Metallic element
447 contents in LwsB from Triplex are comparable to those in LwsA crystals, although Ni is systematically
448 very low (Fig. 8c, d). The Desertes locality shows LwsB with the highest content in metallic elements
449 (Fig. 8a).

450 LwsD crystals (Tpx-D1, Fig. 8e) show trace element patterns generally like those of LwsA and
451 LwsA*, with somewhat higher Sr and Y values, reaching 2000 times chondritic values. LILE, U, Th
452 and all metallic elements are as high as for LwsA* and LwsC in the multi-quartz aggregate Tpx-AC4.
453 Part of LwsD crystals show relatively flat patterns from LREE to MREE and an enrichment of HREE
454 starting from Ho, resulting in La/Yb \sim 10^{-2} . These patterns are similar to some of LwsA* and LwsC in
455 multi-quartz aggregate Tpx-AC4. (Fig. 8g). The other half of LwsD crystals is comparable with the most
456 REE-enriched LwsA crystals in host calcschist (Fig. 8b, e).

457 Lawsonite crystals in multi-quartz aggregates (LwsA* and LwsB*) in Triplex (Tpx-AB5, Fig.
458 8f) and Assietta Pass (As-AB1, Fig. 8h) show compositions overall similar to those of LwsA and LwsB,
459 respectively (Fig. 8b, c). LILE and HFSE contents are generally consistent with similar variability

460 between lawsonite types (LwsA* are comparable to LwsA and LwsB* to LwsB). Yet LwsA* and LwsC
461 from multiquartz aggregate Tpx-AC4 (Fig. 8g) show higher LILE, U, Th and metallic elements (Cr in
462 particular) than LwsA in host calcschist and LwsA* from other samples. REE patterns are more
463 distinctive: LwsA* and LwsC from multiquartz aggregate Tpx-AC4 (Fig. 8g) show pronounced spoon-
464 shaped REE patterns similar to some of LwsA in host calcschist, while LwsA* from sample Tpx-AB5
465 (Fig. 8f) displays flat REE patterns around 10 to 100 times chondritic values, with one crystal showing
466 a spoon-shaped REE pattern. Most LwsB* crystals from Triplex and sample As-AB1 from Assietta Pass
467 show REE patterns consistent with those of LwsB (Fig. 8a, b, d, h), i.e. with a slightly negative slope
468 and slightly curved. However, in sample As-AB1 from the Assietta pass, LwsB* growing with ankerite
469 (Fig. 8h) shows convex REE patterns (i.e., richer in MREE; Fig. 8h), which is also observed for a few
470 LwsB* crystals from Triplex (Fig. 8f). In the multiquartz aggregate from the Assietta Pass, LwsC
471 crystals show trace element patterns similar to those of neighboring LwsA* (Fig. 8i), with both spoon-
472 shaped and convex REE patterns.

473 Overall, two key observations can be made in the context of expected lawsonite composition
474 from the literature (Martin et al., 2014; Vitale Brovarone et al., 2014; Whitney et al., 2020; Kang et al.,
475 2022). First, enrichment in some elements (LREE, U, Th, Pb, Sr) stands out, in particular for LwsA,
476 LwsA* and LwsD crystals. Second, the dispersion of analyses is huge, even within samples.

477 **Lawsonite $^{87}\text{Sr}/^{86}\text{Sr}$ ratios**

478 Comparing $^{87}\text{Sr}/^{86}\text{Sr}$ ratios from lawsonite crystals and corresponding whole rocks from each location
479 reveals no viable isochron relationships (Fig. 9). To correct the isotopic ratios to the time of blueschist-
480 facies metamorphism, the data were corrected by assuming lawsonite growth ages of 60, 50 or 40 Ma
481 (Table 8). At first order, lawsonite separates display different signatures along the transect (Fig. 9a),
482 varying between 0.7113 near the Piemonte margin units (westward) and ratios around 0.7140 at the As-
483 sietta pass (eastward). Near the contact with the Piemonte margin units, LwsA at Desertes shows similar
484 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios to LwsB collected on the same outcrop (DS02A). The sample collected at Fraiteve (Ft-
485 D1) containing LwsD shows an $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.712635 (± 0.000013). At Triplex, where the lawsonite
486 content is maximum (see Fig. 1d), all four LwsA samples show similar $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (Table 8) around
487 0.7128, very close to the LwsC sample. The LwsB sample collected at Triplex shows higher $^{87}\text{Sr}/^{86}\text{Sr}$
488 ratios than LwsA and reaches 0.713708 (± 0.000015). LwsD contained in sample SL07B shows values
489 higher to LwsB and reaches up to 0.714352 (± 0.000008). LwsA at Bourget has higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio
490 than LwsA in Triplex. At the Assietta pass, LwsA and LwsB show comparable $^{87}\text{Sr}/^{86}\text{Sr}$ ratios around
491 0.7140.

492 Overall, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in lawsonite apparently increase eastward (Fig. 9a), though with
493 significant local variability best demonstrated at Triplex, from 0.7112 at Desertes to .7138 at Assietta.

494 The $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in lawsonite are not correlated with Sr and Rb concentrations (Fig. 9b-c) nor with
495 $^{87}\text{Rb}/^{86}\text{Sr}$ ratio. In the bulk rocks, Sr isotopic ratios are inversely correlated to Sr concentration for the
496 least radiogenic group, but not for the most radiogenic samples and the entire dataset appears scattered.
497 Whole rock and lawsonite values are relatively close to each other at Fraiteve and Triplex (to the west)
498 whereas whole rock and lawsonite values are markedly disparate at Bourget and Assietta (to the east).

499 **DISCUSSION AND GEOLOGICAL IMPLICATIONS**

500 **Composition of lawsonite in calcschists**

501 Lawsonite is increasingly suggested as a proxy for fluid/rock interactions during subduction
502 through its geochemistry, as reviewed by Whitney et al. (2020) and Kang et al. (2022). However,
503 metasedimentary lawsonite is less frequently observed in outcropping rocks and has not been much
504 studied compared to metamafic lawsonite. Comparing the Fe and Al + Cr + Ti contents to the
505 compilation of exhumed lawsonite worldwide (Fig. 10a; Kang et al., 2022) shows that calcschist
506 lawsonite follows a specific trend, different from those identified by Kang et al. (2022). Here, calcschist
507 lawsonite does not contain more than 0.04 Fe a.p.f.u. (regardless of the lawsonite type, Tables 3, 4, 5,
508 6) which may be linked to the incorporation of Fe in ankerite co-stable with lawsonite. The Pb and Sr
509 contents have been used by Whitney et al. (2020) to classify lawsonite compositions, where Sr/Pb ratio
510 below 40 are related to metasedimentary lawsonite (in Qz-rich metasediments, Kang et al., 2022); higher
511 Sr/Pb ratios were interpreted as reflecting metamafic lawsonite (characterized by moderate Sr/Pb below
512 130 and high Sr/Pb for greater values). Most analyses of the Alpine SL lawsonite types fall in the
513 moderate metamafic lawsonite field of Whitney et al. (2020; Fig. 10b) inconsistent with the lithology.
514 Abundance of lawsonite in type 2 and type 3 lithologies of the SL is linked to the presence of carbonates
515 involved in lawsonite crystallization. Here lawsonite compositional trends reflect local fluid-rock
516 interactions rather than larger-scale fluid migrations.

517 **Distribution of trace elements in lawsonite types**

518 Lawsonite easily incorporates trace elements into its structure owing to multiple potential
519 substitutions on Al and Ca sites, the latter being easily deformed and therefore prone to hosting large
520 cations (Ueno et al., 1999; Dubacq and Plunder, 2018). Itoigawaite has been recognized as the Sr
521 equivalent of lawsonite, where Sr occupies the Ca site (Miyajima et al., 1999), suggesting extensive
522 solid solution between the two.

523 *Hourglass sector zoning*

524 Hourglass zoning, observed in LwsA only, dramatically affects Ti (Fig. 6a, c, e). Ti-rich
525 domains also correspond to high LREE concentrations, as illustrated on Ce maps (Fig. 6a, c), to Zr and
526 Hf enrichment (Figs. 6b, d, 10c) but correspond to Sr depletion (Figs. 6e, 10d). Preferential incorporation
527 of Ti and LREE is commonly observed in metasedimentary lawsonite (Ueno et al., 1999; Martin et al.,
528 2011, 2014; Vitale Brovarone et al., 2014), while Ti and Fe hourglass zoning are reported for lawsonite
529 in mafic rocks (Fornash et al., 2019). Hourglass zoning is classically observed in magmatic
530 clinopyroxene and metamorphic staurolite (e.g., Hollister, 1970; Hollister and Gancarz, 1971; Dowty,
531 1976) and appears restricted to specific mineral groups, including tourmaline (Van Hinsberg, 2006).
532 Hourglass zoning is attributed to differential uptake of elements on crystal faces exposed to fluids (Van
533 Hinsberg, 2006). Dowty (1976) showed that this preferential incorporation is controlled by the crystal
534 structure: some elements show a higher affinity with some crystal faces due to electrostatic interactions,
535 increasing their adsorption rate during growth. Fornash et al. (2019) identified potential correlations
536 between the presence of subgrains and the shape of the hourglass zoning, potentially indicating that
537 deformation processes might also play a key role in trace element partitioning while lawsonite
538 crystallizes. Hourglass zoning has therefore been identified as an out-of-equilibrium feature, linked to
539 rapid crystallization (Vitale Brovarone et al., 2014). Practically, crystals showing hourglass zoning
540 provide support that equilibrium was not reached, at least for some elements, with the consequence that
541 thermodynamic modeling and/or partition coefficients based on formula unit properties will fail at
542 reproducing the incorporation of these trace elements in lawsonite.

543 *Concentric growth zoning*

544 In Ti-enriched domains, concentrations of Ti and LREE are not constant. LREE are observed to
545 decrease from core to rim, resulting in flattened REE patterns (Fig. 6b, d). Ti also uniformly decreases
546 from core to rim (Fig. 6c; patchy domains can be related to interface-coupled dissolution-precipitation,
547 see below). These core-to-rim depletions, also observed in tourmaline by Van Hinsberg et al. (2006),
548 are interpreted as reflecting a reservoir effect consistent with closed-system behavior, as for the classical
549 Mn bell-shaped zoning in prograde garnet sometimes referred to as "garnet fractionation" (Tropper and
550 Recheis, 2003; Kohn, 2005; Skora et al., 2006; Dubacq et al., 2019). Here concentric zoning is observed
551 for immobile elements (large trivalent and quadrivalent cations) in lawsonite from pelitic parts, with the
552 lowest permeability, after low-temperature crystallization (180 to 300°C). Assuming at least transient
553 closed-system behavior is therefore reasonable, and backed by previous studies (Henry et al., 1996;
554 Cook-Kollars et al., 2014; Lefeuvre et al., 2020; Herviou et al., 2021). The lack of systematics between
555 cores and rims reflects the transient and local character, with homogenization over small length-scales
556 in isolated parts of the schist. In other words, LwsA preferentially incorporates mobilized Ti and LREE
557 to its hourglass domains, but these elements are probably not distributed homogeneously at any given

558 time in the surrounding schist over more than tens of centimeters, linked to the thickness of
559 pelitic/carbonate alternations in the original sediment.

560 The combination of hourglass zoning and reservoir effects explains the dramatic variations in
561 La/Yb ratios in LwsA crystals (Fig. 10e). Most of the available LREE are incorporated into Ti-enriched
562 domains of LwsA while HREE contents remain homogeneous (Fig. 6b, d, g).

563 *Patchy zoning due to interface-coupled dissolution-precipitation*

564 A third feature observed on lawsonite chemical maps in the schistose lithologies (calcschists
565 types 1 to 3 containing LwsA and/or LwsA*) relates to patchy zoning, consisting of enriched or depleted
566 domains for some trace elements, and cross-cutting homogeneous or differently zoned domains for other
567 elements. This is best seen on the Ti maps of LwsA* in Sup. Mat. 4c, f, where thin Ti-depleted domains
568 are found together with hair-like Ti-enriched domains with a vague preferred orientation. Patchy Ti
569 zoning is also observed for LwsA, either in the hourglass domain (Fig. 6c) or in overgrowths (Fig. 6e,
570 f). Ce and Cr also show patchy or angular zoning in LwsA (Fig. 6a, c) and LwsA* (Sup. Mat. 4c, f).
571 This contrast with hourglass zoning and concentric growth zoning in that the domains appear blurry and
572 not linked to crystal axes. The domains cannot be attributed to contamination as depleted and enriched
573 domains alternate (Figs. 6a, c, e). Interpretation of these patchy domains involves faster crystal growth
574 than the homogenization rate of Ti (or Cr, Ce) in the fluid feeding the crystal at grain boundaries. This
575 has been recognized as similar to interface-coupled dissolution-precipitation, a mechanism leading to
576 spectacular zoning in many mineral groups (including carbonates, apatite, chloritoid, clinopyroxene and
577 garnet) over all metamorphic grades (Martin, 2009; Martin et al. 2011; Ruiz-Agudo et al., 2014; Ague
578 and Axler, 2016; Renard et al., 2019; Figowy et al., 2020). This mechanism is not observed in LwsB
579 (grown from fluids in veins), where element mobility is likely much higher than in schistose lithologies
580 and where crystals grew with much more open space, as the veins may represent incrementally opened
581 structures (Lefevre et al., 2020; Herviou et al., 2023). Interface-coupled dissolution-precipitation may
582 lead to zoning reflecting the distribution of immobile elements before crystallization of neoblasts, as
583 suggested by Carlson (2002) and Martin (2009). Combined with sensitivity on element mobility,
584 interface-coupled dissolution-precipitation will therefore lead to the decoupling of poorly mobile
585 elements from one another (variably affected) and from more mobile elements (unaffected). Here, this
586 translates as adding noise to correlations with Ti (Fig. 10c, d).

587 *Lawsonite overgrowths and system opening*

588 In the schist, LwsA shows overgrowths with distinct chemical fingerprints (Sr-enriched, Ti-
589 depleted), as well as LwsB and LwsB* in HP veins and multiquartz aggregates (Figs. 6f, 7a, d, Sup.
590 Mat. 4a). Overgrowths are Ti-depleted (Figs. 6e, 7a, d, Sup. Mat. 4a) but enriched in Sr, Y and HREE

591 (Figs. 6e, g, 7d) whether found in veins or in the schist. LREE contents in lawsonite overgrowths are
592 variable but do not show clear systematic differences compared to cores (Figs. 6g, 7c, Sup. Mat. 4b).

593 Strontium-rich overgrowths (Fig. 7c) show that Sr is released in the fluid during lawsonite
594 crystallization. Contrary to out-of-equilibrium element incorporation described above for LwsA cores,
595 this likely reflects an increased length-scale for element transfer. Given that Sr is positively correlated
596 with CaO in the bulk rocks and more concentrated in type 4 (carbonate-rich) calcschists (Fig. 2b) and
597 considering that carbonates are the primary reservoir of Sr in these rock units, Sr-rich overgrowths may
598 be interpreted as evidence of opening of the system to carbonates via fluids. Quantifying this requires
599 modeling the origin and source of Sr in the rocks, for which bulk rock and lawsonite Sr isotopic
600 signatures can yield constraints.

601 Strontium isotopic ratios for all samples (Fig. 9) are more radiogenic than expected from
602 Cretaceous limestones (in the range 0.7073 – 0.7077; McArthur et al., 2001). The data from Triplex
603 perhaps hint at inheritance from a Mesozoic protolith (139 ± 65 Ma; MSWD=160; Sup. Mat. 5) but this
604 'errorchron' age is far too old for Alpine HP metamorphism and lawsonite growth. Clearly these rocks
605 failed to isotopically re-equilibrate at the local mineral scale during the Alpine metamorphism of these
606 rocks which occurred around 40-60 Ma (e.g., Agard et al., 2002; Ghignone et al., 2021; Herviou and
607 Bonnet, 2023; Gyomlai et al., 2023). Owing to the low temperatures of blueschist-facies metamorphism,
608 this is not surprising. In bulk samples, the high Sr isotopic ratios are thought to reflect incorporation of
609 terrestrial-derived sediments. This is motivated by the high Sr isotopic ratios of the ~300 to ~500 Ma
610 crystalline basement from Massif Central, Piemont micro-continent and Adria, being eroded at the time
611 of deposition of the SL: commonly in the range 0.703-0.715, ratios as high as 0.731 are reported (e.g.
612 Talbert and Duthou, 1983; Duthou, 1984; Downes et al., 1990, 1991; von Raumer et al., 1999) and high
613 Rb/Sr phases like micas deriving from these rocks could have even higher values. Non-metamorphic
614 Upper Cretaceous sediments of the “nappe des Gets” and Helminthoid flysch, taken as approximate
615 equivalents of the SL before subduction, contain variable and locally high amounts of terrigenous
616 sediments, mostly in the form of clays but including plagioclase feldspar (Caron and Weidmann, 1967;
617 Caron, 1972; Caron et al., 1981), a major host of Sr. Rock samples in the least radiogenic group (with
618 $^{87}\text{Sr}/^{86}\text{Sr}$ ratios around 0.709) show decreasing isotopic ratios with Sr and Rb concentrations (Fig. 9b, c)
619 possibly reflecting the rock pelitic/carbonate proportions. However, for the more radiogenic samples
620 and the lawsonite separates, the absence of correlation between Sr content and isotopic ratio (Fig. 9b) is
621 explained by the wide range of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the pelitic fraction of the sediments: a unique mixing
622 curve is not obtained because of the variability of the pelitic endmember. Incorporation of variable
623 fractions of Hercynian basement-derived, variably radiogenic Sr into the rock and/or the lawsonite
624 overgrowths leaves a blurry relationship between content and isotopic signature.

625 This small-scale variability of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios and the lack of data on isotopic signatures for
626 lawsonite cores vs rims precludes defining precisely the source of Sr feeding overgrowths. Therefore,
627 Sr-rich overgrowths are proof of increased Sr mobility and consistent with limited system opening, but
628 do not reflect increasing devolatilization of the limestones.

629 **Evolution of fluid circulation and element mobility length-scale in the Schistes** 630 **Lustrés metasediments**

631 Trace element distribution and zoning patterns in the different lawsonite types give insights into
632 the evolution of element mobility and fluid circulation length-scales in the calcschists during burial (see
633 Fig. 11).

634 LwsA (and LwsA*) in host calcschists appears first (Fig. 11a-b; Lefeuvre et al., 2020).
635 Hourglass zoning indicates out-of-equilibrium incorporation for some trace elements, including Ti and
636 LREE. Fine zoning in LREE and Ti shows small-scale migration of immobile elements via interface-
637 coupled dissolution-precipitation. LREE core-to-rim depletion suggests progressive depletion in the
638 surrounding fluid, explaining the high variability of trace element concentrations and the decreasing
639 La/Yb ratios over time, correlating positively with Ti but negatively with Sr (Figs. 6e, f, 7d, 10d). This
640 is consistent with a closed-system behavior and element transfer restricted to μm to mm scale. As LwsA
641 starts crystallizing early during burial (predicted crystallization at ~ 0.4 GPa and 180°C ; Lefeuvre et al.,
642 2020), element mobility was likely restricted by the small amount of fluid available at grain boundaries
643 and by sluggish diffusivity at such low temperature. The Sr-rich overgrowths in LwsA (Fig. 6e, f) are
644 interpreted as recording a slightly more open-system behavior, with length-scales of element mobility
645 reaching tens of centimeters (Fig. 11b). Increasing element mobility is likely due to the on-going
646 dehydration reactions (mostly from the pelitic fraction, Lefeuvre et al., 2020), occurring until peak
647 conditions (Fig. 11c, d). The similar Sr isotopic ratios of LwsA separates and bulk samples from Triplex
648 also show evidence of local-scale element redistribution during LwsA crystallization.

649 The significance of the apparent eastward increase of Sr isotopic ratios in LwsA across the
650 Strada dell'Assietta transect (Fig. 9a) remains unclear. However, metamorphic reactions occurring in
651 the pelitic fraction with increasing P-T conditions, including destabilization and dehydration of clay
652 minerals (see Vidal and Dubacq, 2009) and/or increasing dissolution of detrital plagioclase are likely to
653 account for Sr isotopic changes. A denser sampling and further analyses on other phases (calcite and
654 ankerite from the SL; plagioclase and clays from the Pre-Alps Gets unit, a non-metamorphic analog) are
655 required to clarify this, as the evolution of the average Sr isotopic ratio of the detrital sediments during
656 deposition is unknown. The large difference between lawsonite separates and whole rock $^{87}\text{Sr}/^{86}\text{Sr}$ for
657 all locations east of Triplex is also an unresolved first-order problem. There, lawsonite separates are far
658 more radiogenic than their host rocks, unlike locations further to the west. It is possible that the lawsonite

659 crystals grew from Rb-rich phases out-of-equilibrium with the rest of the whole rock (e.g., detrital micas
660 and mica-like clay minerals), inheriting their elevated $^{87}\text{Sr}/^{86}\text{Sr}$. Alternatively, this could reflect open
661 system influx of a radiogenic Sr rich fluid that was more significant in the east than in the west.

662 Although interpretations for LwsC and LwsD cannot be generalized due to a lack of data for
663 these scarce lawsonite types, it is noteworthy that LwsC veins in multiquartz aggregate present mineral
664 assemblages and trace element patterns similar to the neighboring LwsA* in the calcschists (Fig. 8g),
665 and LwsD in the host rocks is very similar to LwsA and LwsA* (Fig. 8b, e, f). Therefore, LwsC and
666 LwsD may have formed from the same local fluid, transferring elements only at the meter scale or
667 smaller, restricted to the meter.

668 The elemental homogeneity of LwsB and LwsB* cores (Fig. 8a, c, d, f, h) suggests that the vein
669 network collecting internally derived fluids allowed efficient homogenization and element
670 redistribution. As for LwsA, Sr-rich overgrowths (LwsB2, Fig. 7b, c) likely reflect an increasing
671 distance of element transfer during LwsB crystallization, in agreement with the incremental growth
672 patterns of LwsB fibers (Lefeuvre et al., 2020). LwsB $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are higher than those of LwsA at
673 Triplex but lower at Desertes and similar at Assietta, which is also consistent with opening of the LwsB
674 vein network to surrounding pelitic/carbonate layers with variable Sr isotopic ratios. LwsB and LwsB*
675 show similar trace element patterns all along the Strada dell'Assietta transect, suggesting that similar
676 small-scale processes acted synchronously along the transect, regardless of the varied pressure-
677 temperature conditions. However, the heterogeneity of their Sr isotopic signature (Fig. 9) suggests that
678 the scale of fluid homogenization in LwsB vein network did not reach the km-scale but was efficient on
679 the order of hundreds of meters (sub-unit scale; Fig. 11c) in a transiently connected LwsB vein network,
680 continually evolving with ductile deformation.

681 Overall, our results are in line with previous isotopic, trace elements and fluid inclusions studies
682 suggesting a closed-system behavior with a limited scale of fluid circulation inside the SL units during
683 subduction (Henry et al., 1996; Agard et al., 2000; Bebout et al., 2013; Cook-Kollars et al., 2014; Epstein
684 et al., 2020; Herviou et al., 2021). This small scale and pervasive fluid circulation is also supported by
685 the distribution of veins across outcrops (Herviou et al., 2023). However, a local and limited influx of
686 fluid containing radiogenic Sr cannot be ruled out at Triplex and locations further east, where lawsonite
687 separates show both the highest and most variable $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, and where multiquartz aggregates are
688 numerous.

689 During prograde and peak metamorphism, fluids are internally produced via dehydration
690 reactions in pelitic horizons and react with carbonate layers (see also Herviou et al., 2021) but mobilized
691 elements are rapidly trapped in lawsonite and newly formed carbonates. Growth of LwsA in the schist
692 leads to rapid, effective immobilization of poorly mobile elements like Ti, in addition to consuming

693 water. Crystallization of lawsonite follows a continuum (see Lefeuvre et al., 2020) starting early in the
694 subduction history and lasting until peak metamorphic conditions (Fig. 11d). Along this continuum,
695 opening of the system appears progressive, with increasing fluid-rock interaction scales (from mm-scale
696 to m-scale, Fig. 11a-c.), as shown by i) incremental growth of LwsA, ii) appearance of other lawsonite
697 types in veins, iii) reaction fronts along the carbonate/pelite interfaces, with rock mixing and reaction
698 rates enhanced by deformation (Lefeuvre et al., 2020), and iv) isotopic disequilibrium of Sr between
699 lawsonite veins and the surrounding schist. Despite evidence for increasing fluid circulation and system
700 opening over time, no evidence for large-scale mass transfer, even close to tectonic contacts, is found.

701 The limited fluid flux inside the SL units compared to the larger fluid circulation suggested in tectonic
702 contacts and in lithological boundaries (e.g., Angiboust et al., 2014; Locatelli et al., 2019; Herviou and
703 Bonnet, 2023) explains the important differences in the geochemical behavior of elements during
704 crystallization of lawsonite from lawsonitite found in metasomatic aureoles (e.g., Piccoli et al., 2018;
705 Vitale Brovarone et al., 2020).

706 CONCLUSIONS

707 The upper units of the Schistes Lustrés deep accretionary complex show an intricate record of
708 extensive fluid/rock interactions in sediments metamorphosed at blueschist-facies conditions. In these
709 units, lawsonite is ubiquitous in veins and in the surrounding schists, crystallized from early prograde
710 to peak burial conditions. This study based on major, trace elements and Sr isotopic ratios provides a
711 detailed framework for petrological and geochemical interpretation of the complex geochemical record
712 of lawsonite. White mica contamination has been found to be difficult to avoid and hinders
713 interpretations based on content of univalent cations. For other minor and trace elements, this study
714 shows that several processes affected the composition of lawsonite, decoupling elements from one
715 another, mostly based on their mobility. These processes take place at equilibrium and out-of-
716 equilibrium, resulting in zoning, apparently unrelated to the changing pressure-temperature conditions.
717 At crystal scale, two main types of zoning are observed: hourglass zoning in the calcschists and growth
718 zoning both in the schists and in the veins. Hourglass zoning preferentially affects Ti and LREE.
719 Concentric growth zoning is best observed for Ti, REE, Hf and Zr, and likely involves many other
720 elements less dramatically. Combination of these mechanisms results in large variability of trace element
721 contents, especially REE that vary over four orders of magnitude. Over time during prograde
722 metamorphism, the La/Yb ratio decreases strongly (from ~100 to ~1) as La appears much more sensitive
723 than Yb to crystal surface effects leading to hourglass zoning.

724 Interface-coupled dissolution-precipitation is a third mechanism decoupling elements in the
725 calcschists and involving poorly mobile elements. It is again best observed for Ti, but affects REE and
726 Cr too, resulting in poor correlations between elements.

727 Lastly, overgrowths, best observed in varying Sr content, show system opening in the veins.
728 System opening remains limited in the host calcschists. Sr isotopic ratios that are consistently more
729 radiogenic than the marine carbonate protolith rule out limestone devolatilization as the main driver of
730 fluid flow and source of the Sr. The overall mineral compositional evolution records progressive and
731 incremental system opening, first limited to tens of centimeters, then to the subunit scale, at the order of
732 hundreds of meters at most. Large-scale mass transfer and fluid influx from external sources are not
733 supported even though the vein network extends over time until peak conditions. This vein network
734 appears fed by internal fluids and enhanced by deformation. In the schists, transfer via fluid advection
735 was limited to mobile elements like Sr during the last growth increments. Comparison between the
736 composition of lawsonite in veins and in the schist has been found critical for interpretation of varying
737 fluid composition over space and time.

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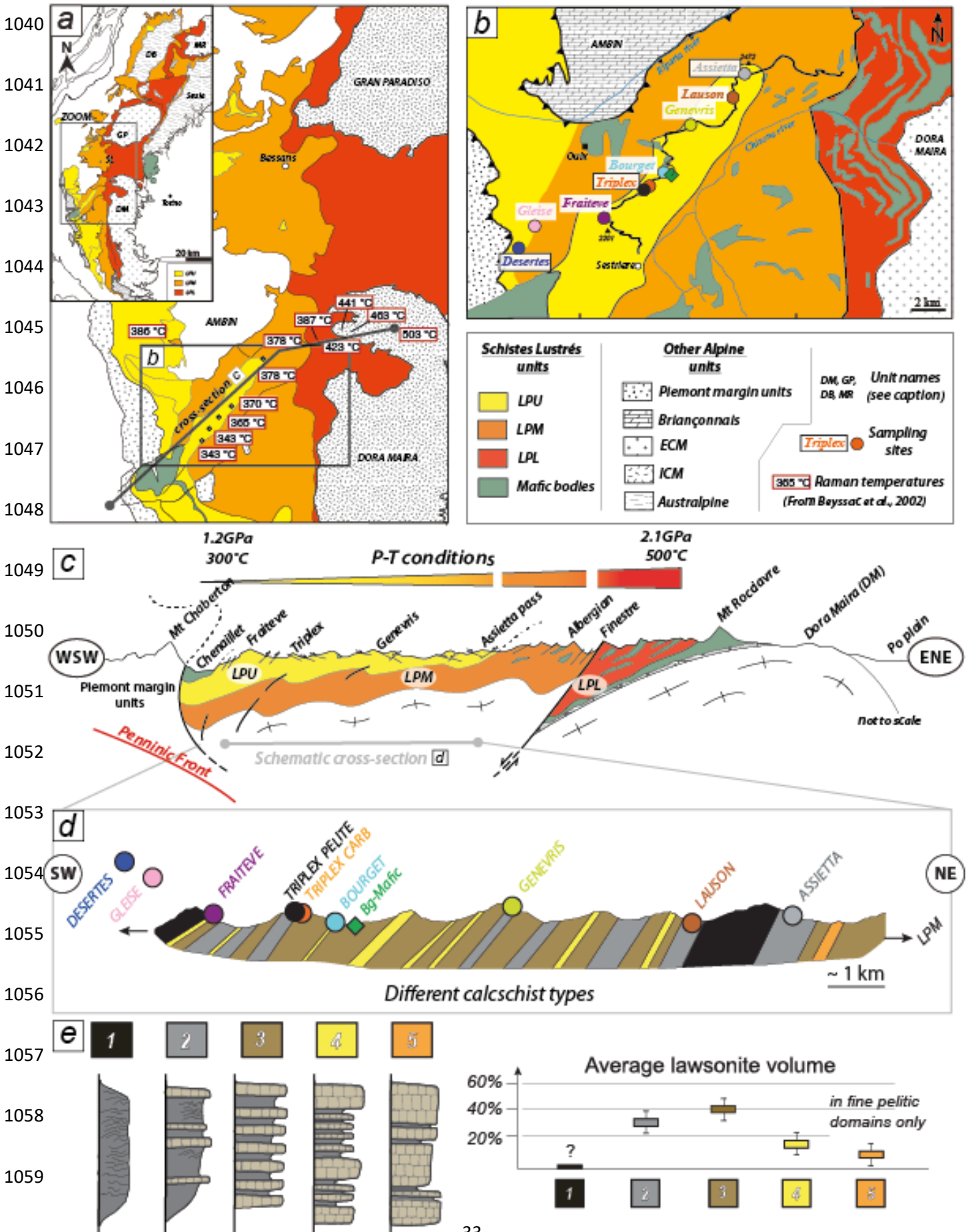
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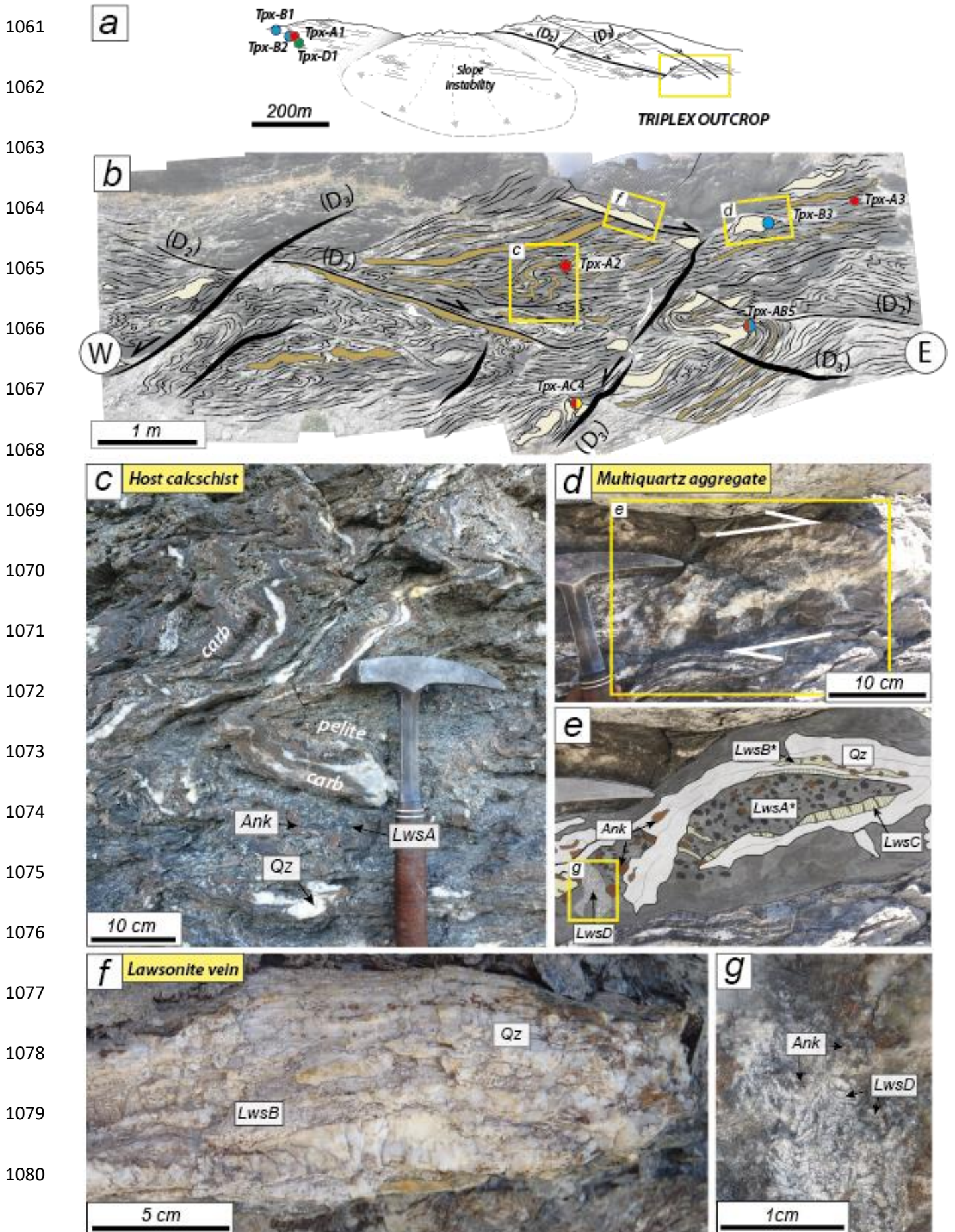
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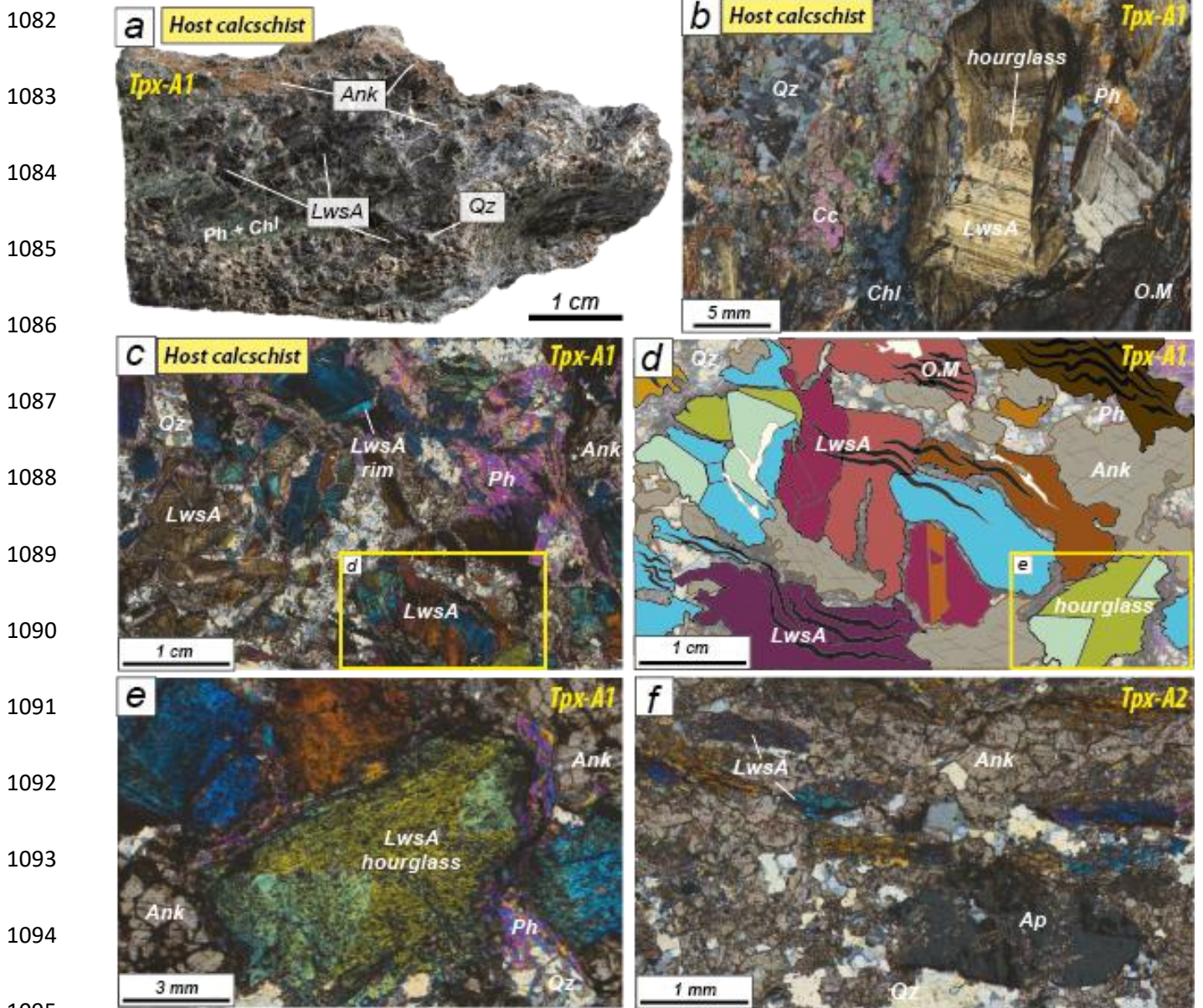
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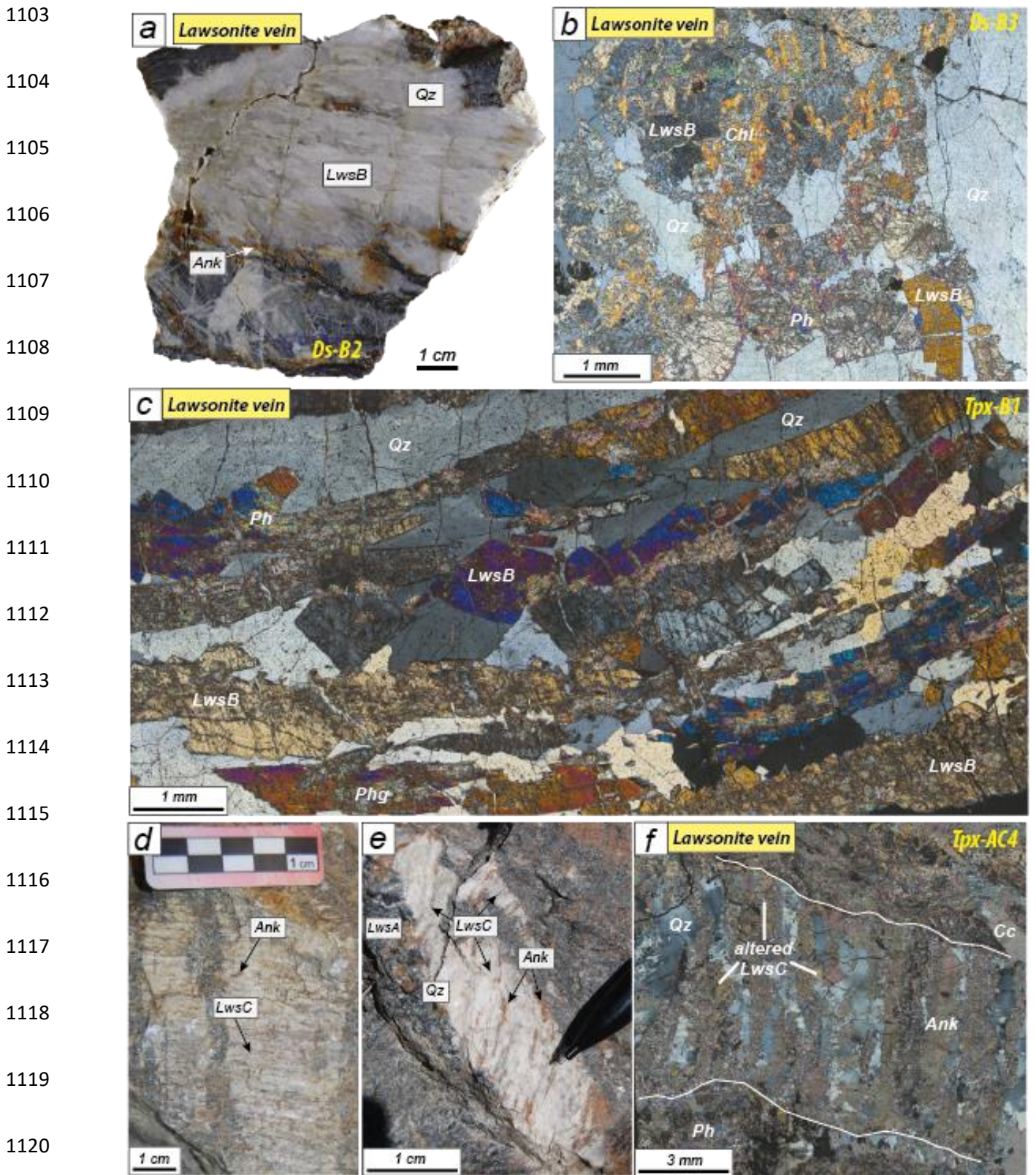
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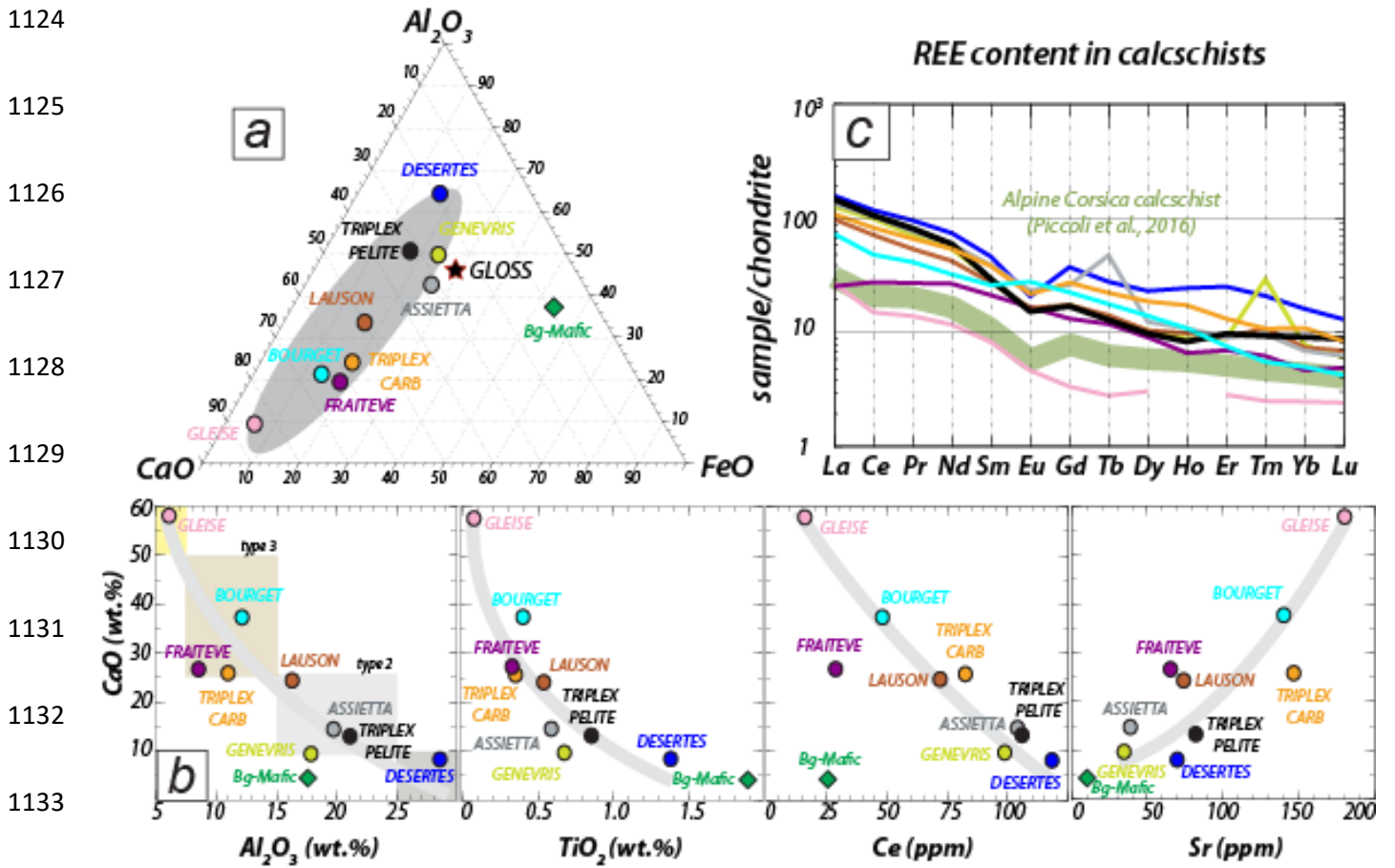
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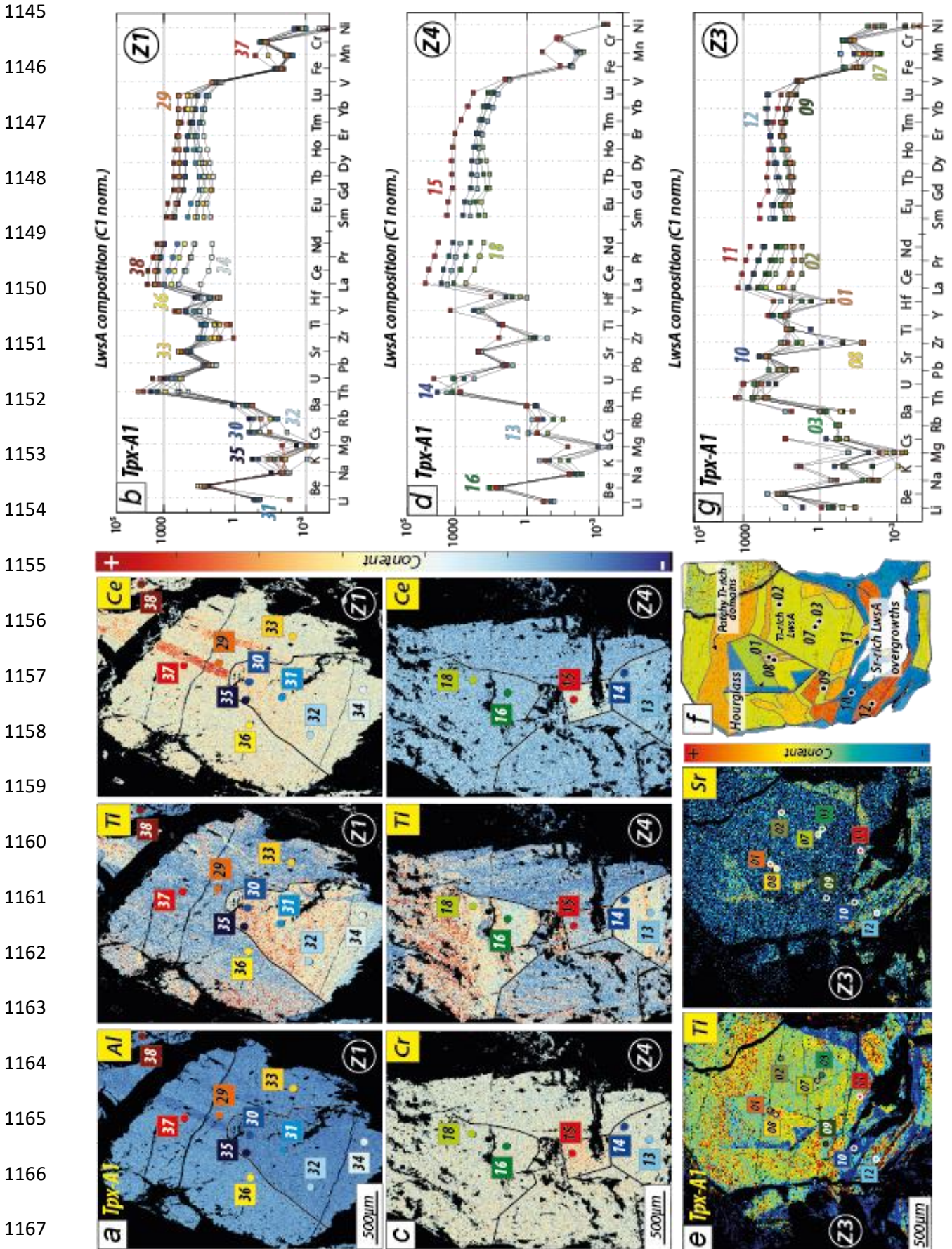
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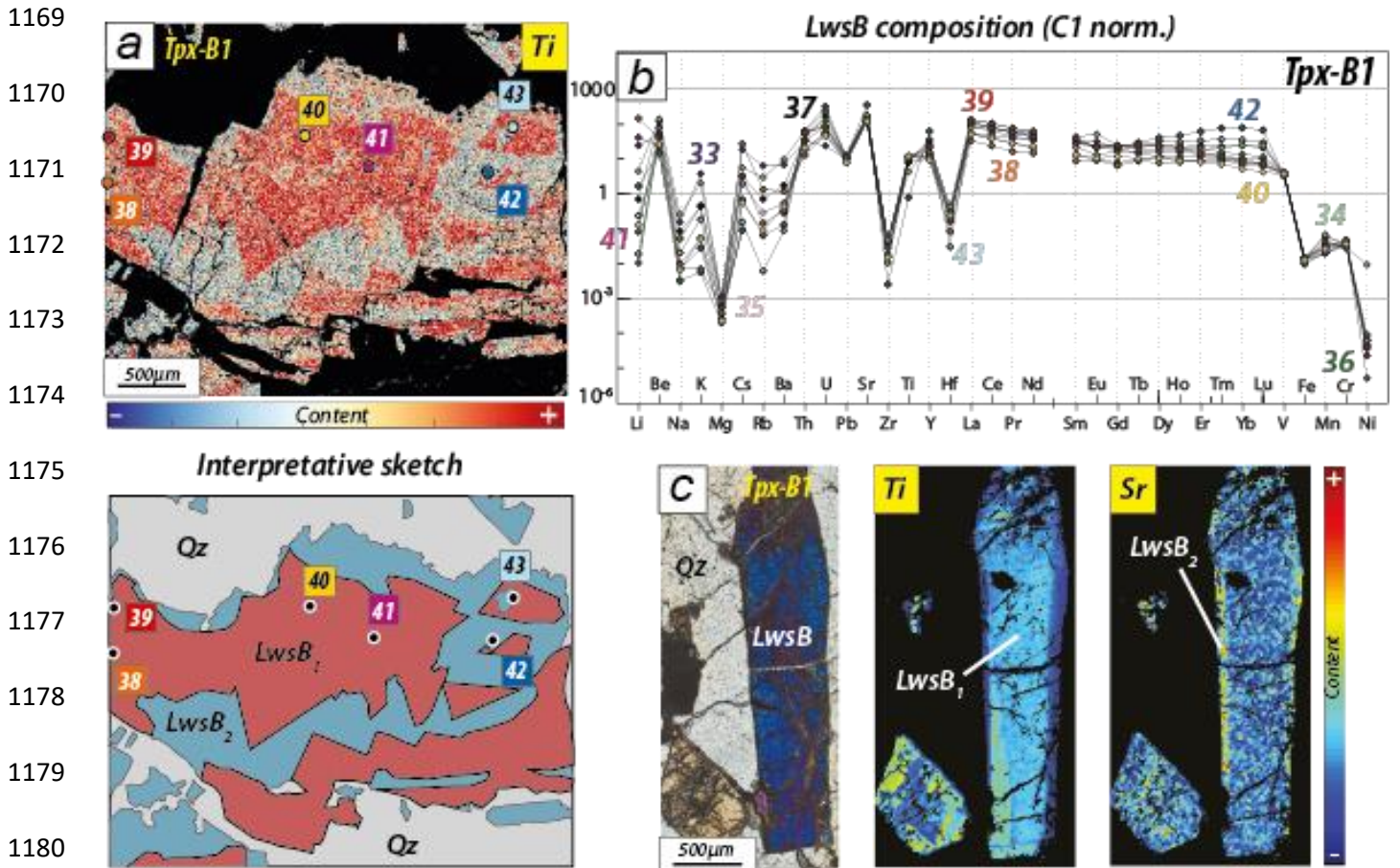
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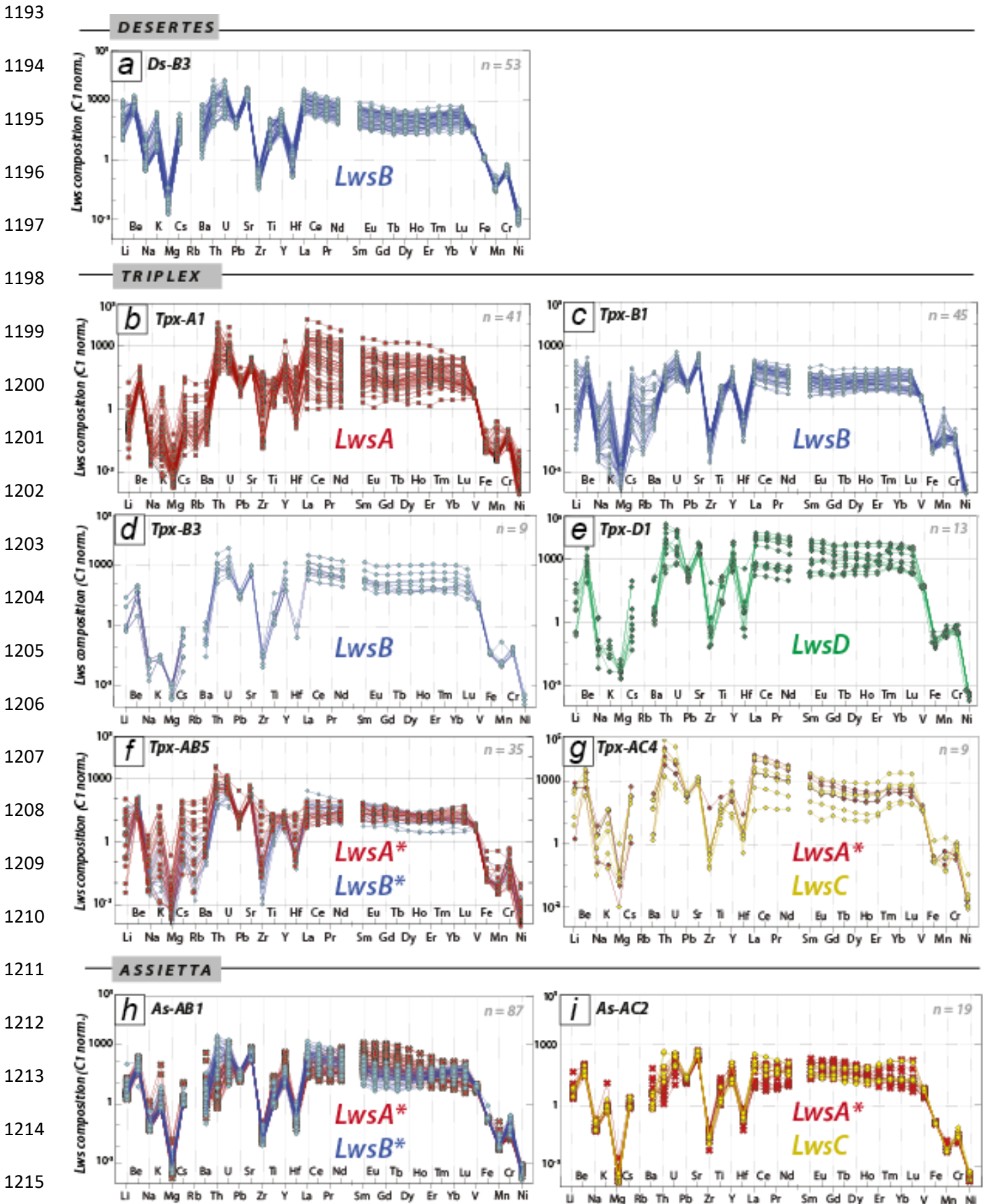
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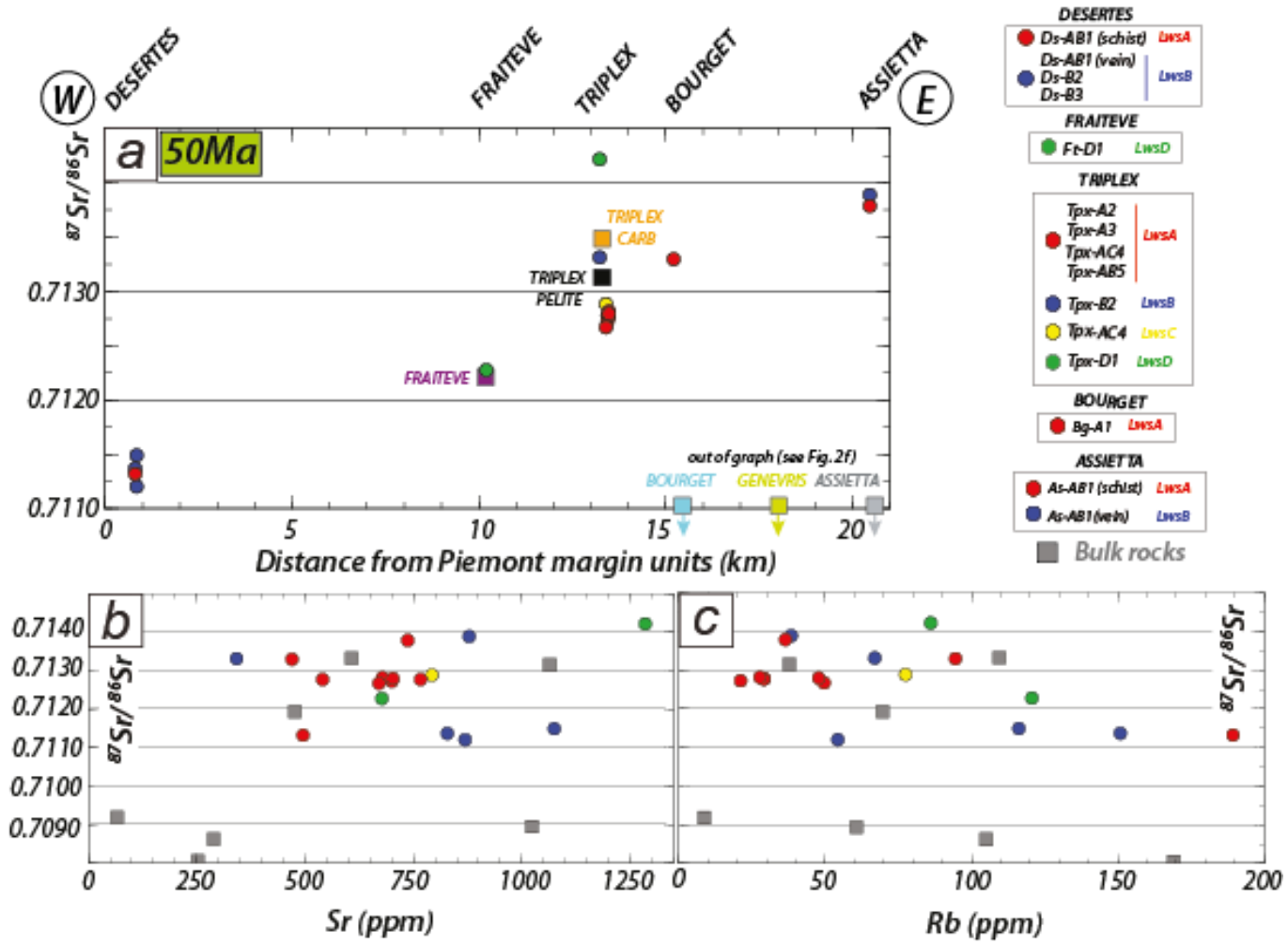
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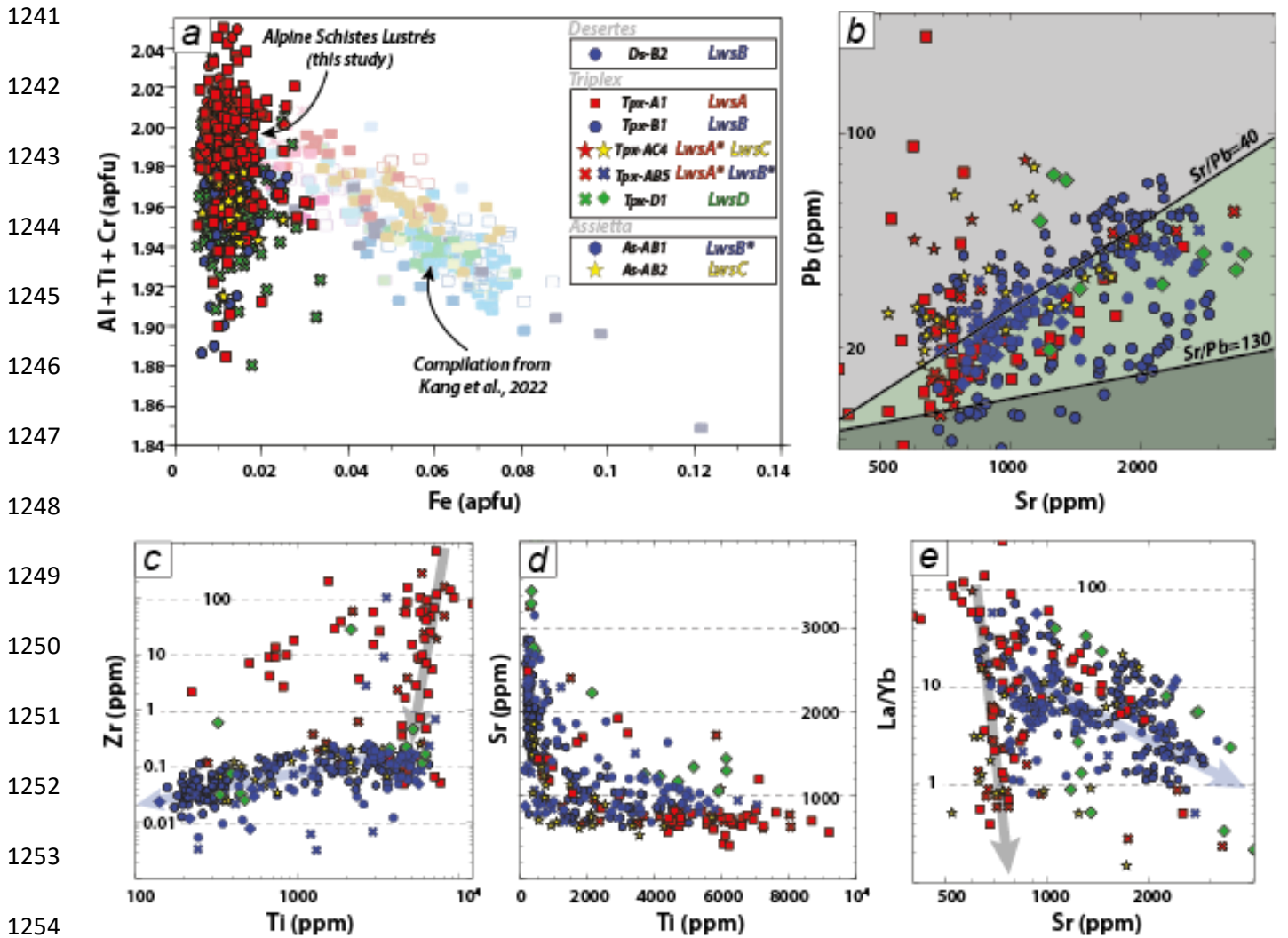
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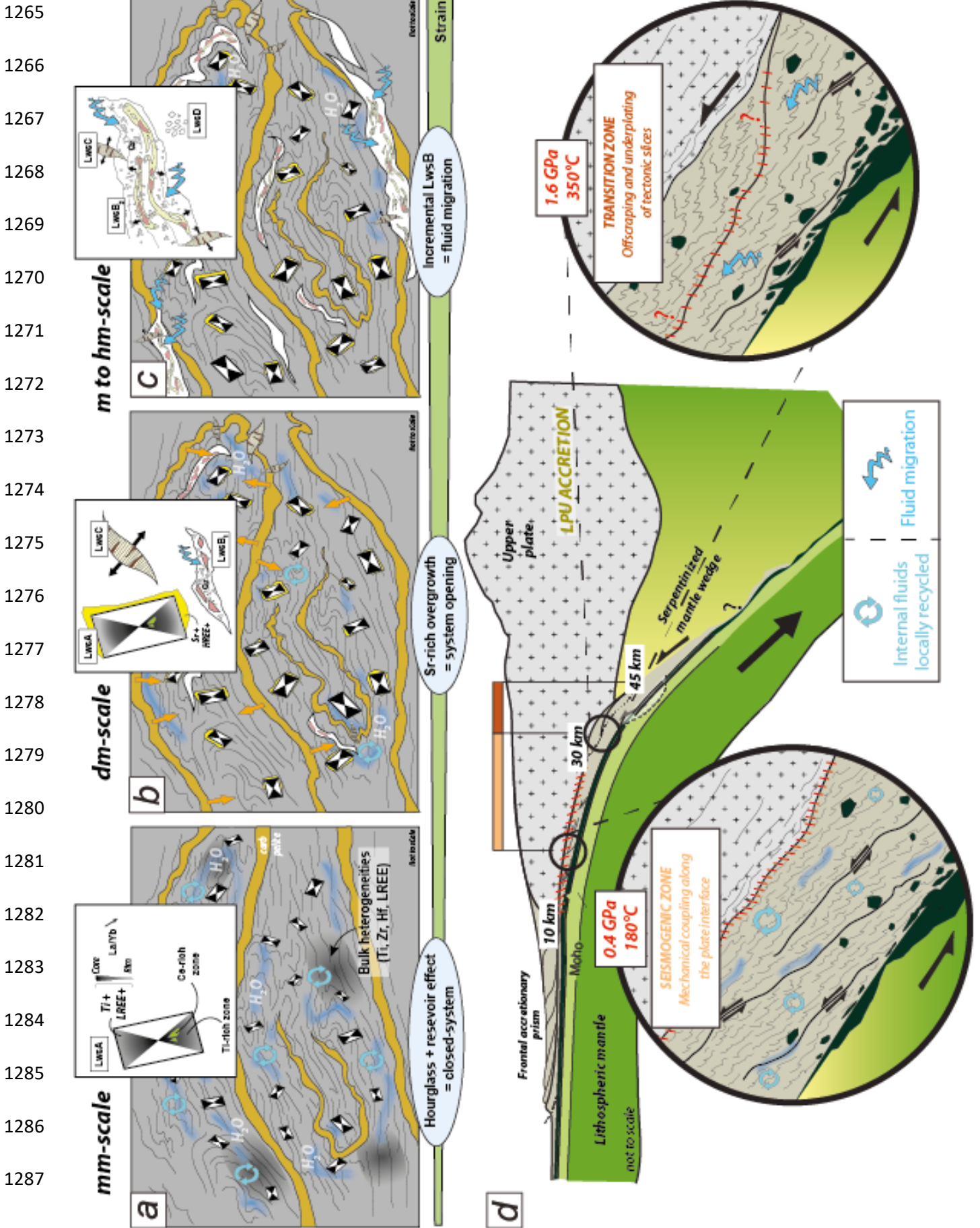
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1240 Fig. 10



1264 Fig. 11



1288 **Figures captions**

1289 **Fig1. Geological settings**

1290 **(a)** Simplified geological map of the Western Alps modified after Herviou et al. (2022). GP: Gran
1291 Paradiso; DB: Dent Blanche; MR: Monte Rosa; DM: Dora Maira. SL units are in yellow for upper units
1292 (LPU), orange for middle units (LPM) and red for lower units (LPL). Raman spectra on carbonaceous
1293 material temperatures of Beysac et al. (2002) are reported. **(b)** Close-up of simplified geological map
1294 of the zone of interest. Mafic bodies are shown in green. Bulk rock samples collected along the Strada
1295 dell'Assietta transect are shown as colored circles and diamonds. **(c)** WSW-ENE simplified cross section
1296 of the SL illustrating the geometry and distribution of the stacked units of the SL. Peak pressure-
1297 temperature conditions from west to east are reported according to Agard et al. (2001). **(d)** Simplified
1298 cross-section of the transect between Fraiteve and the Assietta pass (see Fig. 1). The color code
1299 (schematized in (e)) indicates the rock type, from 1 (pelite) to 5 (carbonate with very few pelite layers).
1300 Samples are located and labeled along the transect (colored circles). **(e)** Average volumetric content of
1301 lawsonite (regardless of lawsonite type) for each lithology.

1302 **Fig2. Several types of lawsonite observed at Triplex**

1303 **(a)** Schematized Triplex outcrop where main deformation structures (D2, D3) are highlighted. Samples
1304 Tpx-A1, Tpx-B1, Tpx-B2 and Tpx-D1 are located with colored dots. **(b)** Triplex outcrop photograph
1305 overlaid by a sketch of the structural framework with locations of samples Tpx-A2, Tpx-A3, Tpx-B3,
1306 Tpx-AC4 and Tpx-AB5. Carbonate horizons are in light brown and lawsonite-bearing veins in white.
1307 Yellow rectangles locate following subfigures. **(c)** Field photograph of intensely folded metasediments
1308 composed of centimeter-scale alternations of pelite-rich and carbonate-rich domains (carb). LwsA black
1309 prisms are abundant in pelite-rich layers and associated with ankerite (Ank). Quartz (Qz) and thin calcite
1310 veins are numerous. **(d)** Centimeter-scale multiquartz aggregate in which all lawsonite types are
1311 observed, schematically interpreted in **(e)**. In these aggregates, clasts of pelitic schists are often
1312 embedded in LwsB bearing veins ductilely deformed. **(f)** Isolated quartz-rich LwsB vein in the SL
1313 metasediments. **(g)** LwsD in the host calcschist, next to LwsB-bearing veins.

1314 **Fig3. Mineralogy of LwsA-bearing host calcschists**

1315 **(a)** Hand specimen of a LwsA-bearing calcschist (Tpx-A1) with LwsA as black prisms, associated with
1316 ankerite and quartz. Note retrograde chlorite (Chl) and phengite (Ph replacing lawsonite). **(b), (c)**
1317 Photographs of the associated thin section (Tpx-A1) underlining the high abundance of LwsA in the
1318 calcschist. Lawsonite crystal cores contain abundant organic matter (OM) inclusions. **(d)** Interpretative
1319 sketch of the area in yellow rectangle in (c) highlighting hourglass zonations commonly observed in

1320 LwsA. **(e)** Close-up view of a hourglass-zoned LwsA associated with quartz and ankerite and partly
1321 retrogressed in phengite. **(f)** LwsA located in the carbonate horizon of sample Tpx-A2 where ankerite
1322 is predominant and where apatite crystals have been observed, a rare accessory mineral.

1323 **Fig4. Mineralogy of LwsB and LwsC-bearing veins**

1324 **(a)** Hand specimen of a LwsB-bearing vein (Ds-B2). LwsB cream-colored crystals is associated with
1325 quartz and grows as fibers elongated parallel to the vein selvages. **(b), (c)** Photographs of Qz and LwsB-
1326 bearing vein thin sections (Tpx-B3, Tpx-B1, cross-polarized). These lawsonite crystals never contain
1327 OM inclusions but are often retrogressed into Ph + Chl mixture. **(d), (e)** LwsC vein in the calcschists,
1328 with ankerite and quartz. **(f)** Photograph of the sample Tpx-AC4 thin section centered on a LwsC-
1329 bearing vein. LwsC are often pseudomorphed into a mixture of phengite, chlorite and calcite,
1330 complicating geochemical analysis.

1331 **Fig5. Representative geology and whole rock geochemistry**

1332 **(a)** Al₂O₃, FeO, CaO relative weight fractions in bulk rock samples. Average pelite composition is
1333 represented in gray and the GLOSS composition is shown for reference. **(b)** Major and trace element
1334 contents for bulk rocks. From left to right: Al₂O₃ (wt. %), TiO₂ (wt. %), Ce (ppm) and Sr (ppm) contents
1335 against the CaO (wt. %) content, the latter reflecting carbonate proportion. **(c)** REE concentrations of
1336 the calcschists, normalized to chondritic values from McDonough and Sun (1995). Alpine Corsican
1337 calcschists from Piccoli et al. (2016) are reported for comparison.

1338 **Fig6. Incremental growth recorded in host calcschist (Tpx-A1)**

1339 **(a)** Al, Ti and Ce chemical maps (SEM) of a LwsA crystal from Tpx-A1 (map C1). These maps highlight
1340 hourglass zoning. **(b)** Spider diagram showing element contents normalized to C1 chondrite, for ablation
1341 spots 29 to 38 located on maps from Fig. 7a. **(c)** Cr, Ti and Ce chemical maps (SEM) of a LwsA crystal
1342 from Tpx-A1 (map C4). Hourglass zoning is best seen on the Ti map. **(d)** Spider diagrams showing
1343 element contents normalized to C1 chondrite, for ablation spots 13 to 18 located on maps from Fig. 7c.
1344 **(e)** Ti and Sr chemical maps (EPMA) of a LwsA crystal from Tpx-A1 (map C3). These maps highlight
1345 a complex history of incremental growth interpreted in sketch **(f)**. The crystal core shows hourglass
1346 zoning entrapped in a Ti-rich domain presenting a patchy distribution overgrown by a Sr-rich, Ti-
1347 depleted rim. **(g)** Spider diagram showing element contents normalized to C1 chondrite (ablation spots
1348 01 to 12 located in Fig.7e).

1349 **Fig7. Incremental growth recorded in quartz-rich LwsB-bearing vein (Tpx-B1)**

1350 (a) Ti chemical map (SEM) of a LwsB crystal from Tpx-B1 (map C3). This map highlights angular Ti-
1351 rich domains in LwsB cores, rimmed by Ti-poor lawsonite, interpreted as incremental growth during
1352 the vein formation. Below: interpretative sketch of LwsB incremental growth in the quartz-rich vein.
1353 LwsB₁ cores are Ti-enriched whereas LwsB₂ rims are Ti depleted. (b) Spider diagram with element
1354 contents normalized to C1 chondrite (ablation spots 38 to 43 located in Fig.8a). (c) Optical microscopy,
1355 Ti and Sr chemical maps (SEM) of a LwsB crystal from Tpx-B1.

1356 **Fig8. Trace element patterns of lawsonite**

1357 Spider diagrams of 35 elements analyzed by laser-ablation ICP-MS in the lawsonite types encountered
1358 in the field. All 311 spectra are normalized to C1 chondrite from McDonough and Sun (1995) and are
1359 presented by sampling sites and lawsonite types to ease reading. The color code refers to the lawsonite
1360 type even when encountered in the same sample (in multiquartz aggregates: Fig. 10f-i).

1361 **Fig9. Strontium isotopic signal of lawsonites**

1362 (a) ⁸⁷Sr/⁸⁶Sr ratios in lawsonite separates, plotted against the sample distance to the Piemont margin
1363 units westward. Sampling locations are reported in Fig. 1b. The different types of lawsonite are
1364 highlighted by specific colors (LwsA: red, LwsB: dark blue; LwsC: yellow; LwsD: green). TIMS
1365 ⁸⁷Sr/⁸⁶Sr data have been corrected considering an age of crystallization of 50 Ma for lawsonite. Estimated
1366 errors are smaller than point size. All the data and corrections are available in Table 8. (b, c) ⁸⁷Sr/⁸⁶Sr
1367 ratios plotted against Sr (b) and Rb (c) concentrations (ICP-MS).

1368 **Fig10. Geochemical correlations**

1369 (a) Al + Ti + Cr content (a.p.f.u.) in function of the Fe (a.p.f.u.) content of the Alpine Schistes Lustrés
1370 lawsonites compared to the compilation of worldwide lawsonites collected in different geodynamical
1371 contexts after Kang et al. (2022). The samples are detailed in the legend and color-coded to identify the
1372 different lawsonite types. (b) Sr content (in ppm) plotted against Pb content (in ppm). The three
1373 highlighted domains (gray, light and dark green) are plotted according to Whitney et al. (2020) where
1374 these Pb/Sr ratios have been inferred to discriminate sedimentary lawsonite from mafic rocks lawsonite.
1375 (c) Ti content (in ppm) plotted against Zr content (in ppm). (d) Ti content (in ppm) plotted against Sr
1376 content (in ppm). (e) La/Yb ratio plotted against Sr content (in ppm). (f) Yb content (in ppm) plotted
1377 against Sr content (in ppm).

1378 **Fig11. Scenario of fluid circulation and element mobility length-scale**

1379 (a) Early crystallization of LwsA in the pelitic schists results from restricted, mm-scale element
1380 migration through locally released fluid. LwsA zoning patterns reveal out-of-equilibrium growth
1381 processes occurring at low temperature (i.e., sluggish diffusivity) in a rather closed system: hourglass
1382 zoning, growth zoning (i.e., reservoir effect) and interface-coupled dissolution-precipitation processes
1383 (i.e., patchy zoning) affecting poorly mobile elements (Ti, LREE, Cr). (b) Along burial, increasing
1384 temperature, deformation and fluid release contribute to progressively increase the scale of fluid-rock
1385 interactions. Sr-rich LwsA overgrowths and local homogeneity of Sr isotopic ratios suggest that fluid
1386 homogenization between pelitic and carbonate-rich layers remained restricted to the dm-scale. LwsC
1387 tension gashes were likely fed by local element transfer. (c) Close to peak burial conditions, the
1388 transiently connected vein network allowed fluid homogenization at the m- to hm-scale (sub-unit scale).
1389 Incremental growth of LwsB veins with Sr-rich rims records the progressive increase of element
1390 mobility length-scale. LwsB incremental veins and multiquartz aggregates acted as drains for fluids.
1391 However, the lack of homogeneity of Sr isotopic signature suggests that the SL unit remained a closed-
1392 system and did not record massive infiltration of external fluids. (d) Integration of this petrological
1393 scenario at subduction-zone scale. The P-T conditions correspond to the LwsA massive formation
1394 (around 0.4 GPa, 180°C; Lefeuvre et al., 2020). At this stage, fluids are locally produced through
1395 metamorphic dehydration reactions and are internally recycled forming large amounts of lawsonite.
1396 Incremental formation of lawsonite veins induces progressive opening of the system, mass transfer likely
1397 reaching the hm-scale. Reaching peak burial (1.6 GPa, 350°C; Agard et al., 2001), the SL units were
1398 scrapped off and progressively stacked to the upper plate allowing their later exhumation. This implies
1399 a shift in the mechanical behavior of the sedimentary pile.

1400

1401 **Table captions**

1402 **(tables and supplementary materials are available contacting the**
1403 **corresponding author)**

1404 **Table 1. Sample characterization**

1405 Samples investigated here, sorted by locations (Desertes, Fraiteve, Bourget, Triplex, Assietta).
1406 Lawsonite types are provided, together with the mineralogy, performed analyses and GPS coordinates.
1407 LwsA* and LwsB* relate to multiquartz aggregates. Mineral proportions are given with a qualitative
1408 scale ('-': not seen; '+': rarely seen; '++': present in the entire thin section in proportions < 50 %; '+++':
1409 ubiquitous).

1410 **Table 2. Bulk rock geochemistry (ICP-OES and ICP-MS)**

1411 Bulk rock analyses. Major elements are expressed in weight %, trace elements in ppm. Bulk rocks are
1412 calcschists, except for sample Bg-Mafic which is a pillow breccia embedded in the Bourget calcschists.

1413 **Table 3. Major elements of LwsA from microprobe analysis**

1414 Weight percentages of investigated oxides for each analysis made on LwsA crystals. TiO₂, SrO and
1415 Cr₂O₃ are given with a greater precision because of special analytical conditions for these elements (see
1416 methods). B.d.l.: below detection limits. The formula units are calculated on 8 oxygen atoms.

1417 **Table 4. Major elements of LwsB from microprobe analysis**

1418 Weight percentages of investigated oxides for each analysis made on LwsB crystals. TiO₂, SrO and
1419 Cr₂O₃ are given with a greater precision because of special analytical conditions for these elements (see
1420 methods). B.d.l.: below detection limits. The formula units are calculated on 8 oxygen atoms.

1421 **Table 5. Major elements of LwsC from microprobe analysis**

1422 Weight percentages of investigated oxides for each analysis made on LwsC crystals. TiO₂, SrO and
1423 Cr₂O₃ are given with a greater precision because of special analytical conditions for these elements (see
1424 methods). B.d.l.: below detection limits. The formula units are calculated on 8 oxygen atoms.

1425 **Table 6. Major elements of LwsD from microprobe analysis**

1426 Weight percentages of investigated oxides for each analysis made on LwsD crystals. TiO₂, SrO and
1427 Cr₂O₃ are given with a greater precision because of special analytical conditions for these elements (see
1428 methods). B.d.l.: below detection limits. The formula units are calculated on 8 oxygen atoms.

1429 **Table 7. Lawsonite geochemistry (LA-ICP-MS/MS)**

1430 Trace element concentrations (in ppm) of different types of lawsonite obtained by laser ablation ICP-
1431 MS/MS.

1432 **Table 8. Strontium isotopic ratios (TIMS)**

1433 Strontium isotopic ratios of bulk rocks and lawsonite separates. The Rb and Sr concentrations (in ppm)
1434 and ⁸⁷Rb/⁸⁶Sr ratios measured with ICP-MS/MS have been added to the dataset. The ⁸⁷Sr/⁸⁶Sr have been
1435 corrected assuming three different crystallization ages for the lawsonite: 40 Ma, 50 Ma and 60 Ma.

1436 **Supplementary material captions**

1437 **Sup. Mat. 1. Mineralogy of LwsA* and LwsB*-bearing samples in multiquartz aggregates**

1438 (a) Multiquartz aggregate observed in the Triplex area with coalescent LwsB-bearing veins. Fragments
1439 of the host calcschists are included in the veins, as schematized on (b). (c) Hand specimen of a
1440 multiquartz aggregate (Tpx-AC4) where schistose clasts containing LwsA* are embedded in a LwsB*-
1441 bearing quartz-rich vein. LwsB* crystals are sometimes associated with ankerite where located next to
1442 the pelitic fragments. (d) LwsB* crystals thin section illustrating elongation of lawsonite fibers
1443 (polarized light). In this sample (Assietta, As-AB1), LwsB* crystals are essentially associated with
1444 quartz and retrogressed to phengite and chlorite. (e) Photograph of the Tpx-AC4 centered on a calcschist
1445 clast where LwsA* are ubiquitous, presenting the same microstructures as LwsA: association with
1446 ankerite and quartz, inclusions of OM in the cores and OM-free rims. (f) Field photograph of a
1447 multiquartz aggregate presenting LwsA* in a host rock fragment and a LwsC-bearing vein. (g) Close-
1448 up view of the sharp interface between LwsA* bearing clasts and the LwsC bearing vein (As-AC2).
1449 LwsC crystals grow perpendicularly to the vein selvages (polarized light).

1450 **Sup. Mat. 2. Lawsonite separation protocol**

1451 Schematized protocol followed in this study to isolate lawsonite crystals from the bulk rocks. Each step
1452 is specified from crushing to the hand picking. SEM imaging was used to ensure purity after heavy
1453 liquids and Frantz magnetic separation.

1454 **Sup. Mat. 3. Univalent contamination of lawsonite by white micas**

1455 Mica contamination evidenced by positive correlations between K (ppm) and several other elements
1456 (Na, Ba, Rb, Li and Cs). Symbols code for lawsonite types.

1457 **Sup. Mat. 4. Distribution of trace elements in multiquartz aggregates (LwsA* & LwsB*)**

1458 (a) Ti chemical map (EPMA) of a LwsB* crystal from Tpx-AB5 (zone C1, see location on the thick
1459 section in Sup.Mat. 4). Ti-rich cores rimmed by Ti-poor borders suggest incremental growth during vein
1460 formation. Spots show locations of the laser ablation analyses (Fig.9b). (b) Spider diagram of C1
1461 chondrite-normalized element contents for spots 29 to 35 (localized in (a)). (c) Ti, Cr and Ce chemical
1462 maps (EPMA) of a LwsA* crystal from Tpx-AB5 (zone C3, see location on the thick section in Sup.Mat.
1463 4). (d) Plane polarized photography of LwsA* highlighting OM-enriched core surrounded by a clear
1464 outer rim. (e) Spider diagram of C1 chondrite-normalized element contents for spots 02 to 09 localized
1465 in (c) and (d). (f) Optical microscopy, Ti and Sr maps (SEM) centered on a LwsC-bearing vein and the

1466 LwsA*-bearing clast. Note that the OM and the Ti are restricted to the LwsA* crystal, illustrating
1467 hourglass zonation.

1468 **Sup. Mat. 5. Strontium isotopic “errorchron”**

1469 $^{87}\text{Sr}/^{86}\text{Sr}$ versus $^{87}\text{Rb}/^{86}\text{Sr}$ for both bulk rocks and lawsonite separates collected at Triplex. The respective
1470 errors are reported on the graphic.

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