

Trace and Rare Earth Element Compositions of Lawsonite as a Chemical Tracer of Metamorphic Processes in Subduction Zones

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Abstract

Lawsonite is a major host mineral of trace elements (TEs; e.g. REE, Sr, Pb, U, Th) and H₂O in various rock types (metabasite, metasediment, metasomatite) over a wide range of depths in subduction zones. Consequently, the composition of lawsonite is a useful archive to track chemical exchanges that occurred during subduction and/or exhumation, as recorded in high-pressure/low-temperature (HP/LT) terranes. This study provides an extensive dataset of major element and TE compositions of lawsonite in HP/LT rocks from two mélanges (Franciscan/USA; Rio San Juan/Dominican Republic), two structurally coherent terranes (Tavşanlı/Turkey; Alpine Corsica/France), and the eclogite blocks of the Pinchi Lake/Canada complex. Bulk major and TE compositions were also determined for lawsonite-bearing host rocks to understand petrogenesis and assess compositional evolution. Most analyzed mélange and coherent-terrané metabasalts have normal mid-ocean ridge/back-arc basin basalt signatures and they preserve compositional evidence supporting interactions with (meta)sediment ± metagabbro/serpentinite (e.g. LILE/LREE enrichments; Ni/Cr enrichments). Most lawsonite grains analyzed are compositionally zoned in transition-metal elements (Fe, Ti, Cr), other TEs (e.g. Sr, Pb), and/or REE, with some grains showing compositional variations that correlate with zoning patterns (e.g. Ti-sector zoning, core-to-rim zoning in Fe, Cr-oscillatory zoning). Our results suggest that compositional variations in lawsonite formed in response to crystallographic control (in Ti-sector zoning), fluid–host rock interactions, modal changes in minerals, and/or element fractionation with coexisting minerals that compete for TEs (e.g. epidote, titanite). The Cr/V and Sr/Pb ratios of lawsonite are useful to track the compositional influence of serpentinite/metagabbro (high Cr/V) and quartz-rich (meta)sediment (low Sr/Pb). Therefore, lawsonite trace and rare earth element compositions effectively record element redistribution driven by metamorphic reactions and fluid–rock interactions that occurred in subduction systems.

Keywords: lawsonite, metasomatism, metamorphism, subduction, fluid

INTRODUCTION

The slab–mantle interface in subduction zones is a locus of fluid–rock interactions, with considerable consequences for geochemical processes. Dehydration reactions occur over a range of pressure and temperature conditions and release fluids from subducted slabs (Schmidt & Poli, 1998; Hacker *et al.*, 2003; van Keken *et al.*, 2011; Muñoz-Montecinos *et al.*, 2021). The liberated fluids transfer incompatible elements (e.g. large ion lithophile elements (LILE)) into the overlying mantle (You *et al.*, 1996; Elliott, 2003), and at depths > 100 km in most subduction zones, induce flux melting in the mantle to generate arc magmas (Tatsumi *et al.*, 1986; Tatsumi & Eggin, 1995).

An abundant hydrous mineral in the crustal part of many subduction zones is lawsonite, a Ca–Al silicate [CaAl₂Si₂O₇(OH)·H₂O] that is a major host of transition metal elements (e.g. Cr, Fe, Ti), trace elements (TE; e.g. Sr,

Pb, U, Th, and rare earth elements (REE)) (Spandler *et al.*, 2003; Usui *et al.*, 2007; Martin *et al.*, 2011, 2014; Vitale Brovarone *et al.*, 2014; Hara *et al.*, 2018; Fornash *et al.*, 2019; Muñoz-Montecinos *et al.*, 2021), and H₂O (up to 11.5 wt%). Since lawsonite is stable from forearc to sub-arc depths (Pawley, 1994; Poli & Schmidt, 1995; Schmidt & Poli, 1998; Forneris & Holloway, 2003), it serves as a vessel for H₂O and TEs in subduction zones. TEs in lawsonite show diverse zoning patterns depending on (1) reaction histories between lawsonite and coexisting minerals that compete for elements (e.g. epidote/allanite, apatite) (Spandler *et al.*, 2003; Martin *et al.*, 2014; Fornash *et al.*, 2019), (2) the element selectivity that differs for each growth plane of lawsonite (Ueno, 1999; Vitale Brovarone *et al.*, 2014; Fornash *et al.*, 2019), and (3) interactions with internally/externally derived fluids (Martin *et al.*, 2014; Vitale Brovarone *et al.*, 2014; Fornash *et al.*, 2019). In addition, isotopic zoning preserved in lawsonite (e.g. Li, Sr, Pb) signals the timing of fluid–rock interactions and

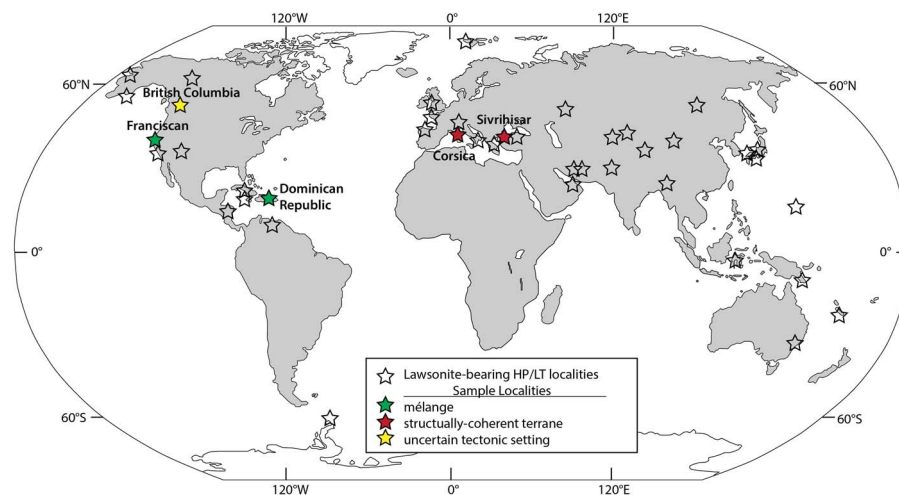


Fig. 1. Representative lawsonite-bearing locations, including sample locations for this study (modified from Whitney *et al.*, 2020).

indicates the origin of host-rock protolith and the source of metasomatic fluids (Simons *et al.*, 2010; Hara *et al.*, 2018). In this respect, lawsonite is a significant phase that can monitor changes in various aspects of subduction systems.

The occurrences of HP/LT rocks can be characterized as tectonic blocks within mélangé or as more structurally coherent complexes (Fig. 1). Examples of lawsonite-bearing HP/LT rocks in mélangé are Port Macquarie, Australia (Och *et al.*, 2003; Nutman *et al.*, 2013), the South Motagua Mélangé Zone, Guatemala (Harlow *et al.*, 2003; Endo *et al.*, 2012) and the northern Rio San Juan Complex of the Dominican Republic (Giaramita & Sorensen, 1994; Krebs *et al.*, 2008) (Fig. 1). Examples of mostly coherent complexes are in Alpine Corsica, France (Martin *et al.*, 2011; Vitale Brovarone *et al.*, 2014), the Tavşanlı Zone, Turkey (Davis & Whitney, 2006; Whitney *et al.*, 2014; Fornash *et al.*, 2019), the Zagros zone, Southern Iran (Agard *et al.*, 2006; Angiboust *et al.*, 2016), and the Sanbagawa belt, Japan (Tsuchiya & Hirajima, 2013) (Fig. 1). In some HP/LT terranes the field relations are uncertain; e.g. in the Pinchi Lake terrane, British Columbia, Canada, blueschist is structurally coherent but lawsonite eclogite occurs as blocks in glacial deposits (Ghent *et al.*, 1993, 1996, 2009; Fig. 1).

In this study, we report the compositional record of lawsonite in HP/LT metamafic and metasomatic rocks from two mélanges (Franciscan/USA; Rio San Juan/Dominican Republic), two structurally coherent terranes (Tavşanlı/Turkey; Alpine Corsica/France), and the Pinchi Lake/Canada complex. TE compositions were obtained using laser ablation inductively coupled plasma-mass spectrometry (LA-ICP-MS), and the distribution of some TEs (e.g. Cr, Ti, Fe) was determined by X-ray mapping with an electron microprobe. This microanalytical approach can detect compositional variations that developed at different stages of metamorphic evolution in individual lawsonite grains. Bulk-rock compositions were also determined for lawsonite-bearing host rocks to understand petrogenesis and track compositional evolution. This

allows the evaluation of lawsonite and bulk-rock compositions to investigate elemental redistribution that occurred at different stages of metamorphism and metasomatism. Therefore, this study is relevant to understanding chemical processes driven by metamorphic reactions and fluid-rock interactions in subduction systems.

GEOLOGICAL BACKGROUND

Here we briefly review the field relationships and P–T histories of the five subduction complexes analyzed.

Mélangé

Franciscan Complex, CA, USA

The Franciscan Complex of California developed by mid-Jurassic to Cretaceous subduction of the Farallon plate beneath the North American plate (Blake Jr., 1988; Wakabayashi, 1992; Ernst, 2011; Dumitru *et al.*, 2013, 2015). It mostly comprises coherent thrust sheets of metabasaltic and metasedimentary rocks (Coleman & Lanphere, 1971; Blake Jr., 1988) and mélangé units consisting of blocks within a serpentinite or siliciclastic matrix (Blake Jr., 1988; Wakabayashi, 1992; Wakabayashi *et al.*, 1999).

Lawsonite-bearing HP/LT rocks occur primarily as blueschists (Platt, 1975; Wakabayashi & Dumitru, 2007; Wakabayashi, 2015). High-grade blocks in mélangé evolved along an anticlockwise P–T path from epidote-amphibolite (600–700°C, 0.7–1 GPa) to epidote-eclogite (550–620°C, 2.2–2.5 GPa), through epidote-blueschist (300–400°C, 0.6–1 GPa), to lawsonite-blueschist facies conditions (150–250°C, 0.6–1 GPa) (Wakabayashi, 1990; Krogh *et al.*, 1994; Ernst & Liu, 1998; Tsujimori *et al.*, 2006; Page *et al.*, 2007) (Fig. 2).

Rio San Juan Complex, Dominican Republic

The Rio San Juan Complex of the Dominican Republic formed during Mesozoic to Cenozoic convergence between the Caribbean intra-oceanic arc system and the North American continent (Pindell & Kennan, 2009;

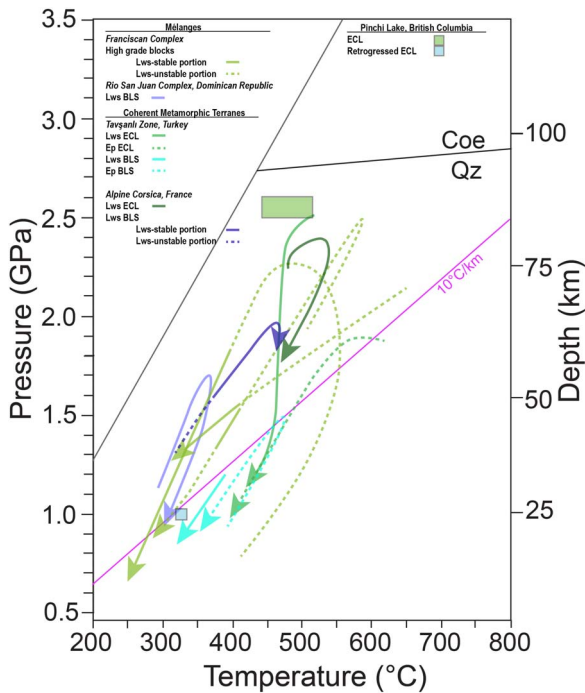


Fig. 2. P–T paths of lawsonite-bearing HP/LT rocks from Franciscan Complex/USA (Wakabayashi, 1990; Krogh *et al.*, 1994; Ernst & Liu, 1998; Tsujimori *et al.*, 2006; Page *et al.*, 2007), Rio San Juan Complex/Dominican Republic (Krebs *et al.*, 2011), Sivrihisar Massif/Turkey (Davis & Whitney, 2006; Pourteau *et al.*, 2019), and Alpine Corsica/France (Vitale Brovarone *et al.*, 2014). Blue and green rectangles indicate the prograde condition of retrogressed eclogite and the peak condition of eclogite from the Pinchi Lake/Canada, respectively (Ghent *et al.*, 1993, 2009).

Escuder-Virueze *et al.*, 2011, 2013). The northern part of the Rio San Juan complex consists of massive bodies of serpentinite, coherent bodies of greenschist-blueschist, and serpentinite-matrix mélanges. Mélange includes blocks of serpentinite, eclogite, blueschist, jadeitite, and marble, with the occurrence of lawsonite primarily confined to blueschist-facies rocks (Krebs *et al.*, 2008, 2011).

Lawsonite blueschist evolved along a clockwise P–T path (Krebs *et al.*, 2008, 2011) (Fig. 2). Peak conditions were 360–370°C and 1.6–1.7 GPa (Krebs *et al.*, 2008, 2011) (Fig. 2). Lawsonite eclogite has only been reported in one region (Samaná Peninsula; Zack *et al.*, 2004; Escuder-Virueze & Pérez-Estaún, 2006). This eclogite occurs as a boulder and has estimated peak conditions of 360°C and 1.6 GPa (Zack *et al.*, 2004).

Structurally coherent terranes

Sivrihisar Massif, Tavşanlı Zone, Turkey

The Tavşanlı Zone, Turkey, marks the subduction of passive continental margin and shallow marine sequences during Late Cretaceous convergence between the Anatolide-Tauride microcontinent and Eurasia (Okay & Kelley, 1994; Sherlock & Okay, 1999; Davis & Whitney, 2006; Okay & Whitney, 2010; Plunder *et al.*, 2013; Fornash *et al.*, 2016). The Sivrihisar Massif is located at the SE part of the Tavşanlı Zone and has excellent exposures of lawsonite eclogite (Davis & Whitney, 2006, 2008; Whitney

& Davis, 2006; Okay & Whitney, 2010; Teyssier *et al.*, 2010; Whitney *et al.*, 2014). Lawsonite eclogite occurs as meters-scale pods within blueschist-facies metabasaltic and metasedimentary (calcschist, quartzite) units (Davis & Whitney, 2008; Whitney *et al.*, 2014) or as mm- to dm-scale layers alternating with lawsonite blueschist (Davis & Whitney, 2006, 2008; Whitney & Davis, 2006; Teyssier *et al.*, 2010; Whitney *et al.*, 2014; Fornash *et al.*, 2016, 2019). Within these structurally coherent units, serpentinite and metagabbro occur as rare pods or lenses (Çetinkaplan *et al.*, 2008; Davis & Whitney, 2008; Whitney *et al.*, 2014). Some meter-scale lenses of antigorite serpentinite are in contact with lawsonite + chlorite-rich ($\pm$ garnet) metasomatic reaction zones (Plunder *et al.*, 2013; Zack, 2013; Whitney *et al.*, 2014; Fornash *et al.*, 2019).

Lawsonite-bearing blueschist, eclogite, and interlayered blueschist/eclogite record a range of peak P–T conditions (380–570°C, 1.2–2.4 GPa) (Davis & Whitney, 2006, 2008; Kang *et al.*, 2020) and there are indications of an anticlockwise P–T path (Davis & Whitney, 2006, 2008) (Fig. 2). Retrograde lawsonite blueschist records metamorphic conditions of ~450–500°C and ~1.5 GPa (Davis & Whitney, 2006, 2008).

Alpine Corsica, France

Alpine Corsica consists of a stack of nappes, including the Schistes Lustrés complex derived from oceanic successions with remnants of the ocean–continent transition (Waters, 1990; Molli *et al.*, 2006; Vitale Brovarone *et al.*, 2011a). Lawsonite-bearing HP/LT units are exposed in the Schistes Lustrés and show an increase in metamorphic grade structurally downward from blueschist (350–460°C, 1.5–1.8 GPa) to eclogite-facies units (490–550°C, 2.2–2.4 GPa) (Ravna *et al.*, 2010; Martin *et al.*, 2011; Vitale Brovarone *et al.*, 2011b) (Fig. 2). Lawsonite-blueschist units differ from lawsonite-eclogite units in their higher proportions of metasedimentary vs. metaophiolitic sequences (Vitale Brovarone *et al.*, 2011a; Meresse *et al.*, 2012). In both units, lawsonite metasomatite formed along the lithological boundaries between metaophiolite and metasediment (Martin *et al.*, 2011; Vitale Brovarone *et al.*, 2011a, 2014).

Other field setting

Pinchi Lake, British Columbia, Canada

The Pinchi Fault Zone in central British Columbia marks a Permian–Triassic oceanic subduction zone (Paterson & Harakal, 1974; Paterson, 1977; Ghent *et al.*, 1996). In the fault zone, a series of elongate units vary in lithology and metamorphic grade, including blueschist facies metasedimentary and metamafic rocks and serpentinized ultramafic rocks (Paterson, 1973, 1977).

Coherent blueschist units contain lawsonite and record a range of P–T (230–330°C, 0.8–1.2 GPa) (Paterson, 1973). Lawsonite-bearing eclogites are exposed at two different locations in the Pinchi Lake area (Paterson, 1977; Ghent *et al.*, 1993, 2009). They occur as isolated

boulders without showing any clear geological context (Paterson & Harakal, 1974; Ghent *et al.*, 1993, 2009). Despite the uncertain field relations, lawsonite eclogite boulders have been interpreted to be part of a *mélange* that is genetically associated with coherent blueschist units given their similarities in muscovite and phengite cooling ages (218 ± 7 Ma) (Paterson & Harakal, 1974; Ghent *et al.*, 1996) and close proximity (Ghent *et al.*, 1993, 2009). The least retrogressed eclogite has peak P–T conditions of 450°C and 2.5 GPa (Ghent *et al.*, 2009), whereas more retrogressed eclogite records 310°C as a prograde T at a lower P limit of 1.0 GPa (Ghent *et al.*, 1993) (Fig. 2).

PETROLOGY AND PETROGRAPHY

A summary of sample locations and modal mineralogy is provided in Table 1.

Franciscan Complex, CA, USA (*mélange*)

We analyzed four samples from two different serpentinite-matrix *mélanges* (Ring Mountain and North Berkeley Hills) and one siliciclastic-matrix *mélange* (Blind Beach, Sonoma County).

Ring Mountain, Tiburon Peninsula, Marin County

The Ring Mountain *mélange* is known for the abundance of high-grade metabasaltic blocks (blueschist, eclogite, and amphibolite) within a serpentinite matrix (Coleman & Lanphere, 1971; Wakabayashi, 1992; Tsujimori *et al.*, 2006). The *mélange* is structurally overlain by an ultramafic unit (Coleman & Lanphere, 1971; Wakabayashi, 1992; Tsujimori *et al.*, 2006), and some high-grade blocks are partially encased by metasomatic actinolite-chlorite-talc-phengite rinds (Coleman & Lanphere, 1971; Catlos, 2003). The two lawsonite-bearing samples analyzed from this locality were provided by S.R. Mulcahy: RR-1 (Mulcahy *et al.*, 2014) and LVT-1.

Sample LVT-1 is a metabasalt comprised of omphacite-rich and glaucophane-rich layers alternating at a mm-to cm-scale (Fig. 3a). Both layers have the same mineral assemblage (garnet + omphacite + lawsonite + epidote + glaucophane + phengite + rutile, partially replaced by titanite + apatite with later chlorite + Fe-oxide) but differ in mode, and they contain a lawsonite-rich layer (vein) (Fig. 3a). In both blueschist and eclogite layers, lawsonite, phengite, glaucophane, and omphacite define the foliation (Fig. 3a), and garnet contains inclusions of epidote, omphacite, quartz $\pm$ rutile $\pm$ apatite $\pm$ zircon. The lawsonite-rich layer (vein) is up to 4 mm thick, and it is concordant with the foliation.

Lawsonite in the lawsonite-rich layer is euhedral and significantly coarser than euhedral, matrix lawsonite (up to 2.8 mm vs. $90\text{--}520\text{ }\mu\text{m}$ in length), thereby interpreted to have crystallized *in situ* in the vein (Fig. 3a and b). Partially chloritized garnet + omphacite + glaucophane + phengite + epidote + rutile (partially replaced by titanite) either crosscut or occur inside the vein (Fig. 3a and b).

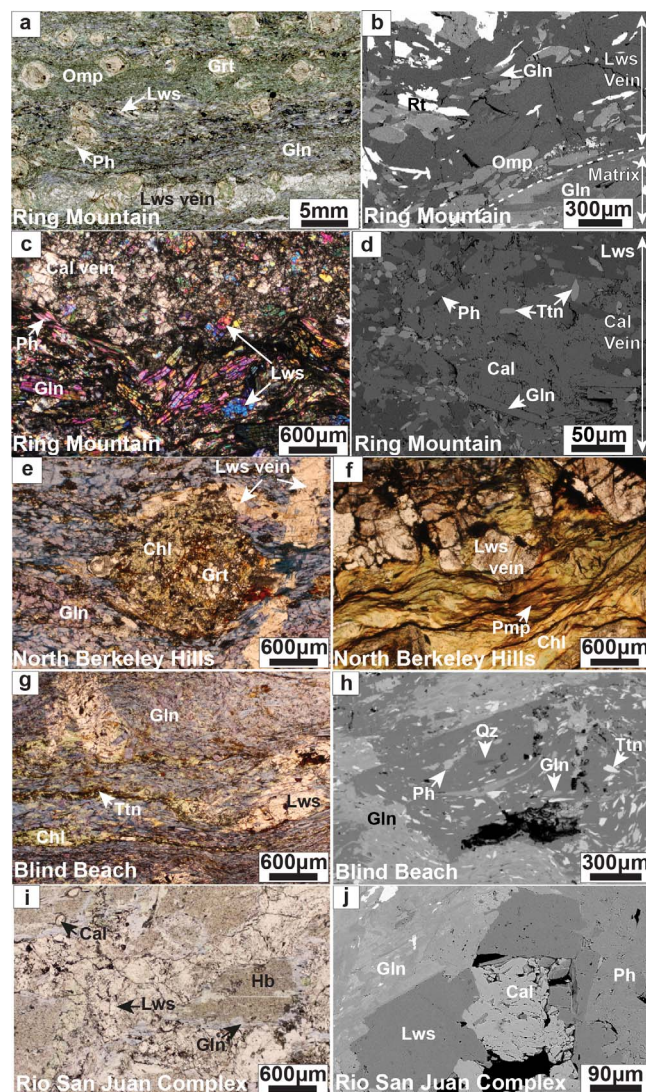


Fig. 3. Lawsonite-bearing samples from *mélanges*. (a, b) Images of interlayered metabasalt within the Ring Mt serpentinite-matrix *mélange* (LVT-1), showing alternating omphacite-rich and glaucophane-rich layers (a; PPL), and inclusions in lawsonite-rich layer (b; BSE). Dashed line indicates the boundary between the glaucophane-rich matrix and the lawsonite-rich layer (b; BSE). (c, d) Images of blueschist within the Ring Mt serpentinite-matrix *mélange* (RR-1), showing calcite vein (c; XPL) and inclusions in the vein (d; BSE). (e, f) Photomicrographs of blueschist from the North Berkeley Hills (EC-1B). Two different generations of lawsonite veins (e; PPL) show a partial-to-extensive replacement by chlorite and pumpellyite (f; PPL). (g, h) Images of blueschist within the siliciclastic-matrix *mélange* at Blind Beach (SM-8), showing matrix phases (g; PPL) and inclusions in lawsonite (h; BSE). (i, j) Images of blueschist-facies metagabbro exposed in the northern Rio San Juan Complex (IEC15-3.5), showing matrix minerals (i; PPL), and lawsonite partially replaced by calcite (j; BSE). Mineral abbreviations follow Whitney & Evans (2010).

Sample RR-1 is a lawsonite-blueschist, consisting of glaucophane, lawsonite, and phengite, with minor amounts of titanite and apatite (Fig. 3c). A calcite vein in the sample developed parallel to the foliation defined by the major matrix minerals (Fig. 3c). All matrix minerals are included in or crosscut the calcite vein (Fig. 3d). Lawsonite in the matrix and the vein are subhedral to euhedral (Fig. 3c and d) and commonly contain titanite.

Table 1: Sample locations and mineral modal abundances

Sample	Location	Mineral modal asbundance (%)														
		Lws	Ep	Gln	Amp	Omp	Aug	Ph	Grt	Rt	Ttn	Qz	Ap	Chl	Pmp	Cal
LVT-1	37°54'18.59"N 122°29'32.46"W	9	5 ^G	30		13 ^G		20	9	8* ^G	L	G	G	3		3
RR-1	37°54'19.67"N	10	6 ^G	14		28 ^G		18	10	6* ^G	5 ^L	G	<1 ^G	5		2
	122°29'29.16"W	14		46				26			3 ^L		1		7	
EC-1B	37°55'55.08"N	33		19		Cal-Vein		1			4		<1		43	
	122°18'32.34"W	22		41 ^L		Matrix		8	<1	<1*	2+		<1 ^L	24		
SM-8	38°25'34.75"N 123°6'58.00"W	26		8		Chl + Pmp-rich area		12					<1+	50		
IEC15-3.5														(Chl + Pmp)		
SV08-283C	39°35'42.85"N 31°16'17.36"E	39		38 ^L		Matrix		1 ^L			7 ^L	L	1 ^L	15 ^L	L	<1
SV01-50A	39°36'0.72"N 31°15'20.58"E	15	1	8	62 (Hb)			5		2* ^L					L	<1
		14 ^G	19 ^{L,G}	5 ^{L,G}	8 (Act)			19 ^G	12	1 ^{L,G}	2 ^G	2 ^{L,G}	<1 ^G	3		
SV01-75A	39°43'22.28"N 29°48'42.49"E	41		32 ^L		Matrix (BLS)		6		14*	L		L	1	L	1
		23		13 ^L		Cal-Vein (BLS)		1		7*	L		L	16		39
TZ10-2.2C	39°55'55.55"N 31°15'56.40"E	36	5	13		Matrix (ECL)		<1		13*	L		L			
C13	54°30'N 124°W	22	6	<1		Cal-Vein (ECL)		<1		10*	3 ^{L,G}	3 ^L	L	4	44	
BLR4	54°30'N 124°W	17 ^G	1 ^L	34 ^{L,G}		Matrix		32	<1	L ^G	5 ^L	4 ^L	1 ^{L,G}	6	L	L ^G
BLR5	54°30'N 124°W	47	2 ^L			Matrix		15			1 ^L	L	<1 ^L	51 ^L	L	<1 ^L
		18 ^G		52		Matrix		16 ^G	6	1		8 ^G		4		
				41		Matrix		10	6	2* ^G		3 ^G	2	1		

* Some rutile grains are partially replaced by titanite. ^G Minerals that occur as garnet inclusions. ^L Minerals that occur as lawsonite inclusions.

Lawsonite in the vein is fine-grained (60–240 μm) relative to lawsonite in the matrix (250 μm –1.6 mm).

North Berkeley Hills

Blocks of blueschist outcrop either on top of or within a serpentinite body that occupies the crestline of the North Berkeley Hills (Brothers, 1954; de Roever & Brothers, 1955). A blueschist block (sample EC-1B) exposed along Arlington Avenue in North Berkeley, CA, USA, has been considered as one such block, and it consists of lawsonite + glaucophane + phengite + titanite, with minor amounts of garnet (partially replaced by chlorite) + rutile (partially replaced by titanite) + apatite + pyrite (Fig. 3e). Lawsonite in the matrix is euhedral and fine-grained (260–300 μm), and it contains apatite and glaucophane. Lawsonite veins are either concordant or discordant to the foliation defined by lawsonite, glaucophane, and phengite (Fig. 3e). Chlorite and pumpellyite are texturally later than other matrix phases and have partially to completely replaced vein-lawsonite (Fig. 3f).

Blind Beach, Sonoma County

At Blind Beach, prehnite-pumpellyite facies siliciclastic-matrix mélangé hosts clasts and blocks of diverse lithologies, including serpentinite, clinopyroxenite, blueschist, and amphibolite, as well as prehnite-pumpellyite facies sandstone, chert, and basalt (Wakabayashi, 2015). One lawsonite-blueschist sample (SM-8) was collected from a block, and it consists of lawsonite, glaucophane, phengite, apatite, and titanite with texturally late chlorite and Fe-oxide (Fig. 3g). Lawsonite has a variable grain size (210 μm –1.7 mm), with some coarse grains impinged by trails of glaucophane and titanite (Fig. 3g). Lawsonite is subhedral to euhedral and contains most matrix minerals as inclusions (Fig. 3h).

Rio San Juan Complex, Dominican Republic (mélangé)

Sample IEC15-3.5 is from a blueschist facies metamafic boulder in the northern Rio San Juan Complex. Amphibole occurs as a major matrix phase (Table 1) and is very coarse-grained (2.6–5.8 mm). The sample contains lawsonite, phengite, and hornblende rimmed by glaucophane, with rare epidote, rutile (partially replaced by titanite), and calcite (Fig. 3i). Lawsonite has a grain size of 120–410 μm and commonly contains omphacite, rutile, pumpellyite, and albite. Some lawsonite grains are partially replaced by calcite and are therefore subhedral in shape (Fig. 3j). Owing to the coarse-grained size of the matrix phases, we described this sample as a metagabbro. This sample was provided by S.M. Gordon from a field trip stop on the 2015 International Eclogite Conference (sample IEC15).

Tavşanlı Zone, Turkey (structurally coherent)

Lawsonite-bearing samples were collected from an eclogite pod (SV08-283C), an interlayered metabasalt (SV01-50A), a blueschist layer (SV01-75A), and a lawsonite +

chlorite-rich metasomatic block (TZ10-2.2C). The SV samples are from the Halilbağı area of the Sivrihisar Massif and the TZ sample is from further west in the Tavşanlı Zone.

Sample SV08-283C is an eclogite, consisting of omphacite + garnet + lawsonite + epidote + phengite + glaucophane and minor amounts of quartz + apatite + rutile + titanite + zircon, with actinolite (partially replacing glaucophane) + chlorite as secondary phases (Fig. 4a). Garnet contains most matrix phases as inclusions. Matrix lawsonite is mostly euhedral (Fig. 4a and b) and has a grain size of 470–530 μm . Some lawsonite grains include epidote, glaucophane, quartz, and rutile (Fig. 4b).

Sample SV01-50A comprises glaucophane-rich (blueschist) and omphacite-rich (eclogite) layers alternating at a mm- to cm-scale (Fig. 4c). Both layers consist of lawsonite + glaucophane + omphacite + phengite + rutile (partially replaced by titanite) + texturally late chlorite but differ in mode (Fig. 4c). Epidote only occurs in eclogite layers (Fig. 4d). Calcite and quartz veins are in both layers and they either crosscut or are parallel to the foliation defined by lawsonite, glaucophane, and omphacite (Fig. 4c). Most matrix minerals are included in the calcite veins (Fig. 4d). Lawsonite is fine-grained (50–270 μm) and euhedral in both the matrix and the vein. Lawsonite commonly includes glaucophane, omphacite, quartz, apatite, titanite, and pumpellyite.

Sample SV01-75A is from an extensive (tens of meters in outcrop) blueschist layer and consists of lawsonite + phengite + glaucophane + garnet and minor amounts of quartz + epidote + titanite + apatite (Fig. 4e), with texturally late chlorite and quartz and calcite veins. Lawsonite and garnet contain glaucophane, epidote, apatite, rutile, titanite, and zircon as inclusions (Fig. 4f). In addition, garnet includes lawsonite. Lawsonite is euhedral in shape and variable in grain size (190 μm to 1.5 mm) (Fig. 4e and f).

Sample TZ10-2.2C is from a metasomatic zone located adjacent to a serpentinite lens (Plunder et al., 2013; Zack, 2013; Whitney et al., 2014; Fornash et al., 2019). It consists of subhedral to euhedral coarse-grained lawsonite (>0.5 mm) in a chlorite-rich matrix with minor amounts of omphacite, epidote, titanite, apatite, and zircon (Fig. 4g). Lawsonite contains epidote, aegirine-augite, titanite, apatite, zircon, pumpellyite, and Fe-oxide as inclusions. The sample site is near stop 2.2 in the field guide of Okay & Whitney (2010).

Alpine Corsica, France (structurally coherent)

In Alpine Corsica, lawsonite-bearing omphacitite has been found as mm- to cm-scale veinlets in blueschist-facies metagabbro (Vitale Brovarone, 2013). Omphacitite-hosting metagabbro typically occurs as blocks in meters-scale metasedimentary sequences (e.g. quartzite, calc-schist, marble) (Vitale Brovarone, 2013) and they are in proximity to metaophiolitic blocks (Lagabriele & Lemoine, 1997).

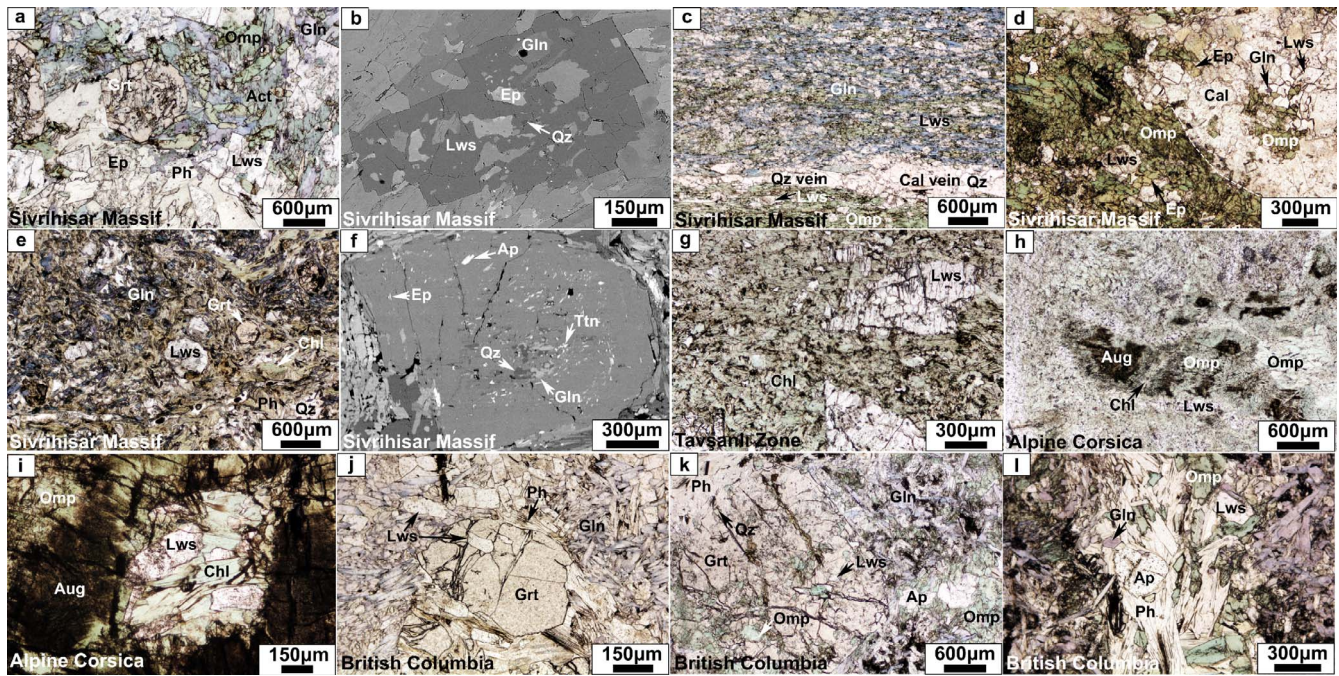


Fig. 4. Lawsonite-bearing samples from structurally coherent terranes. (a, b) Images of eclogite in the Sivrihisar Massif (SV08-283C), showing matrix phases (a; PPL) and inclusions in lawsonite (b; BSE). (c, d) Photomicrographs of Sivrihisar metabasalt, consisting of alternating blueschist and eclogite layers. Both blueschist and eclogite layers contain lawsonite in the matrix (c; PPL) and in the calcite vein (d; PPL). White dashed line indicates the boundary between the vein and the eclogite matrix (d). (e, f) Images of blueschist in the Sivrihisar Massif (PPL; SV01-75A), showing matrix phases (e; PPL) and inclusions in lawsonite (f; BSE). (g) Image of lawsonite + chlorite-rich metasomatite in the Tavşanlı Zone (PPL; TZ10-2.2C). (h, i) Photomicrographs of omphacite-hosting metagabbro in Alpine Corsica (C13). Coarse-grained omphacite preserves relict augite (h; PPL), with partially dissolved areas filled with lawsonite + chlorite (i; PPL). (j) Photomicrograph of retrogressed eclogite from the Pinchi Lake, Canada (PPL; BLR4). (k, l) Photomicrographs of eclogite from the Pinchi Lake (BLR5), showing matrix phases (k; PPL) and lawsonite included in a phengite-rich area (l; PPL).

Sample C13 is an omphacite-hosting metagabbro block exposed in a landslide. It mainly consists of omphacite and lawsonite with lesser amounts of epidote, titanite, and apatite (Fig. 4h). Coarse-grained omphacite (up to ~0.6 cm) has been boudinaged and preserves relict augite in the grain (Fig. 4h). Some omphacite grains are partially dissolved and filled with chlorite ± omphacite ± lawsonite aggregates (Fig. 4i). Lawsonite is euhedral and has a grain size of 190–540 μm. Omphacite, epidote, apatite, titanite, quartz, and pumpellyite occur as inclusions in lawsonite. The absence of key minerals (e.g. garnet, glaucophane) and unclear field relations (a landslide sample) make it hard to assign metamorphic facies for the sample. Therefore, the sample is referred to as a lawsonite-omphacite metagabbro (Lws-Omp metagabbro) in this study. The sample was provided by A. Vitale Brovarone.

Pinchi Lake, British Columbia, Canada (eclogite blocks)

Both samples (BLR4, 5) are from the Beaver Lake Road area, where eclogite blocks occur with glacial deposits. Sample BLR4 is a retrogressed eclogite, consisting of garnet, glaucophane, phengite, and lawsonite, with lesser amounts of rutile and quartz (Fig. 4j). Talc was reported in this sample by Ghent *et al.* (1993) but was not observed in the thin section analyzed.

Sample BLR5 is an eclogite, composed of garnet + omphacite + glaucophane + phengite + lawsonite +

titanite + rutile (partially rimmed by titanite) + quartz + apatite with secondary chlorite (Fig. 4k). Some lawsonite grains in BLR5 are included in texturally later phengite-rich areas (Fig. 4l). Garnet in BLR4 has an inclusion assemblage (omphacite + lawsonite + phengite) slightly different from that in BLR5 (omphacite + lawsonite + quartz ± rutile; Fig. 4k). In both samples, matrix lawsonite is euhedral and has a grain size similar to or coarser than lawsonite inclusion in garnet (50–560 μm vs. 51–280 μm). The samples were collected by E.D. Ghent.

ANALYTICAL METHODS

Bulk-rock compositions of vein-excluding matrix areas were determined for the samples from mélange areas in the Franciscan (three samples: LVT-1, RR-1, EC-1B) and Rio San Juan Complex (one sample: IEC15-3.5), and from structurally coherent terranes in the Tavşanlı Zone (three samples: SV08-283C, SV01-75A, TZ10-2.2C) and Alpine Corsica (one sample: C13). We additionally analyzed the bulk-rock compositions of vein areas in two Franciscan blueschists (RR-1, EC-1B). Bulk major-element compositions were determined by X-Ray Fluorescence and selected TEs were measured by ICP-MS at the GeoAnalytical Lab at Washington State University (Table 2). Bulk-rock major element compositions were calculated for the Pinchi Lake/British Columbia eclogites (BLR4, 5), using mineral modes and compositions because we only have thin sections of these two samples. To determine mineral

modes, Energy-dispersive X-ray spectroscopy mapping analyses were performed for selected areas within thin sections.

Major- and minor-element compositions of minerals as well as X-ray element maps of lawsonite and garnet were acquired with a JEOL JXA-8530F Plus Electron Probe Microanalyzer in the Department of Earth and Environmental Sciences at the University of Minnesota. The *in situ* TE compositions of lawsonite, garnet, and apatite were measured using a Resonetics M-50 193 nm ArF excimer laser ablation system coupled to an Agilent 7700s quadrupole ICP-MS at the GeoHistory Facility, Curtin University. Details of analytical methods are presented in the Supplementary Material 1.

RESULTS

We present bulk-rock compositions (Table 2) as well as lawsonite, garnet, and apatite compositions (Tables 3 and 4; Supplementary Table S1). The compositions of other matrix minerals are presented in Supplementary Table S1.

Bulk-rock compositions

Metabasites from mélange and coherent-metamorphic terranes

Mafic samples analyzed are considered as metabasalt and metagabbro (inferred to have a metavolcanic and plutonic protolith, respectively). In most cases this designation is based on grain size: coarse-grained samples are inferred metagabbro, although grain size in a metamorphic rock in some cases may not relate to grain size in the protolith owing to the effects of deformation and metamorphic reactions (e.g. Kang *et al.*, 2020). Nevertheless, in cases in which a volcanic or plutonic protolith has been interpreted by others who have collected and/or studied the samples, we use these previously determined designations.

Analyzed metabasalt shows variable enrichment and/or depletion relative to normal mid-ocean ridge basalt (N-MORB; Gale *et al.*, 2013) in terms of Al_2O_3 (12.1–18.7 wt%), MgO (3.38–9.26 wt%), CaO (5.41–15.5 wt%), and P_2O_5 (0.11–1.62 wt%) with higher MnO (0.17–0.29 wt%) and K_2O (0.35–2.4 wt%) (Fig. 5; Table 2). As compared with the metabasalts from mélange areas, those from coherent-metamorphic terranes are lower in TiO_2 , FeO_{tot} , and Na_2O (Fig. 5; Table 2). Metagabbro samples have bulk-rock compositions mostly within the range of these metabasalts, except for a few oxides; lower MnO in the Rio San Juan blueschist-facies metagabbro (IEC15-3.5; mélange), and higher Na_2O as well as lower TiO_2 , FeO_{tot} , K_2O , and P_2O_5 in the Corsica Lws-Omp metagabbro (C13; coherent-terrane) (Fig. 5; Table 2).

Analyzed metabasalt samples are variably enriched and/or depleted in bulk V, Cr, and Ni with respect to N-MORB (174–366 $\mu\text{g/g}$ V, 82.5–443 $\mu\text{g/g}$ Cr, 57.7–323 $\mu\text{g/g}$ Ni; Fig. 5; Table 2). The highest Ni and Cr concentrations

were measured from the Sivrihisar blueschist (SV01-75A; coherent-terrane). Metagabbro samples have V, Cr, and Ni concentrations mostly within the range of the metabasalts, except the Corsica Lws-Omp metagabbro (C13; coherent-terrane) that has lower Cr and Ni contents.

Chondrite-normalized REE patterns of metabasalt samples are nearly flat-to-positively sloped ($\text{La/Yb}_\text{N} = 0.85\text{--}12.1$; $\text{N} = \text{Chondrite-normalized}$) (Fig. 6a). Primitive mantle (PM)-normalized TE patterns of these metabasalts display enrichments in LILE (e.g. Cs, Rb, and Ba) (Fig. 6b). Pb and Sr behave nonuniformly relative to the associated LREE; the coherent metabasalts show a positive Pb anomaly and a flat-to-negative Sr anomaly, whereas most mélange metabasalts show slight-to-significant depletions in Pb and Sr, except for the Ring Mt blueschist/eclogite (LVT-1; mélange) with a positive Pb anomaly and the Ring Mt blueschist (RR-1; mélange) with a positive Sr anomaly (Fig. 6b).

The Rio San Juan blueschist-facies metagabbro (IEC15-3.5; mélange) shows a TE pattern similar to the metabasalts, and it is characterized by LREE, LILE and Sr enrichments with a negative Pb anomaly (Fig. 6). The Corsica Lws-Omp metagabbro (C13) has a distinct TE pattern with positive Pb and Sr anomalies and significant depletions in LREE ($\text{La/Yb}_\text{N} = 0.58$) and LILE as compared with the other metabasite samples (Fig. 6).

Metabasalt from Pinchi Lake, British Columbia

Retrogressed eclogite (BLR4) is depleted in TiO_2 , FeO_{tot} , CaO, and Na_2O , but enriched in SiO_2 , MgO, and K_2O relative to eclogite (BLR5) (Fig. 5; Table 2). The Cr content of BLR4, calculated from bulk Cr_2O_3 , is higher than that of BLR5 (Fig. 5; Table 2). Despite the different degree of retrogression, both eclogite samples show significant enrichments in SiO_2 (54.5–58.3 vs. 42.2–52.0 wt%) and Na_2O (3.79–4.48 vs. 1.62–3.96 wt%) as compared with the other metabasite samples (Fig. 5; Table 2).

Metasomatite and lawsonite-bearing veins

Tavşanlı metasomatite (TZ10-2.2C; coherent-terrane) is significantly low in SiO_2 , Na_2O and K_2O , but high in Al_2O_3 , FeO_{tot} , MnO, MgO, and V relative to the metabasite samples (Fig. 5; Table 2). The TiO_2 , CaO, P_2O_5 , Cr, and Ni contents of metasomatite are within the compositional ranges of the metabasites (Fig. 5; Table 2). The REE and TE patterns of Tavşanlı metasomatite are largely similar to those of the Sivrihisar metabasalts (SV01-75A, SV08-283C), showing LREE and Pb enrichments, with a negative Sr anomaly (Fig. 6). However, LILE content is significantly depleted in the metasomatite (Fig. 6b).

Lawsonite-bearing veins in two Franciscan mélange blueschists have compositions different from host blueschist matrix. In the North Berkeley Hills blueschist (EC-1B), the lawsonite vein is lower in SiO_2 , TiO_2 , Na_2O and K_2O , but higher in Al_2O_3 , MgO and CaO relative to the host matrix (Fig. 5; Table 2). The bulk Ni and Cr concentrations of this vein is markedly higher than those

Table 2: Bulk-rock major and TE compositions

Locality	Mélange areas					Coherent metamorphic terranes							
	North Berkeley Hills/Franciscan Complex		Ring Mountain/Franciscan Complex			Blind Beach/Franciscan Complex	Rio San Juan Complex	Tavşanlı Zone		Alpine Corsica	Pinchi Lake/British Columbia		
Rock type	BLS (Matrix)	BLS (Vein)	BLS (Matrix)	BLS (Vein)	BLS/ECL	BLS	BLS-facies Metagabbro	BLS	ECL	Metasomatite	Lws-Omp Metagabbro	Retrogressed ECL*	ECL*
Sample	EC-1B-M	EC-1B-R	RR-1-M	RR-1-V	LVT-1	SM-8	IEC15-3.5	SV01-75A	SV08-283C	TZ10-2.2C	C13	BLR4	BLR5
SiO ₂	49.3	44.3	42.2	39.0	49.2	48.4	48.5	52.0	43.7	30.8	48.6	58.3	54.5
TiO ₂	2.20	1.48	2.32	2.02	1.44	2.01	1.06	1.02	1.22	2.16	0.62	1.02	2.15
Al ₂ O ₃	14.8	16.1	16.4	14.4	14.4	15.4	15.4	12.1	18.7	21.0	15.4	16.2	16.1
FeO _{tot}	12.4	12.5	6.62	6.22	9.38	11.9	8.14	8.09	7.70	14.0	6.10	7.71	8.61
MnO	0.19	0.26	0.19	0.15	0.22	0.23	0.16	0.17	0.29	0.30	0.18	0.15	0.14
MgO	5.96	8.98	3.38	3.12	8.04	5.96	7.77	9.26	4.26	13.8	6.16	7.18	6.40
CaO	5.41	6.63	13.5	16.5	8.96	6.73	10.2	5.67	15.5	7.51	14.5	4.07	6.52
Na ₂ O	3.68	2.31	2.51	2.33	3.47	3.94	2.91	2.35	1.62	b.d.l.	3.96	3.79	4.48
K ₂ O	1.39	0.36	2.40	3.02	1.47	0.35	0.92	1.91	1.50	<0.01	<0.01	1.54	1.10
P ₂ O ₅	0.25	0.12	0.59	0.52	0.11	0.18	0.08	0.14	1.62	0.53	0.02		
LOI	4.07	6.48	9.28	12.3	2.75	4.66	4.33	6.99	3.61	9.73	4.20		
Sum	95.5	93.0	90.2	87.3	96.7	95.1	95.2	92.7	96.1	90.0	95.6	100	100
μg/g													
La	8.30	4.87	34.6	39.0	8.90	5.27	4.85	17.6	50.7	56.6	1.52		
Ce	23.5	12.4	64.6	77.5	20.9	14.0	10.2	34.0	93.7	117	3.70		
Pr	3.80	2.10	7.50	9.28	2.87	2.47	1.60	4.24	11.9	14.0	0.72		
Nd	19.3	10.5	28.7	35.6	13.6	13.2	8.25	17.4	46.1	55.7	4.10		
Sm	6.21	3.38	5.73	7.42	4.10	4.51	2.90	4.32	9.92	13.3	1.69		
Eu	2.05	1.30	1.85	2.39	1.68	1.71	1.11	1.20	2.45	4.59	0.91		
Gd	7.78	4.50	5.43	6.74	5.27	6.38	3.85	4.57	9.52	14.1	2.53		
Tb	1.42	0.82	0.84	1.07	0.98	1.20	0.73	0.83	1.64	2.38	0.51		
Dy	8.81	5.27	4.75	5.90	6.29	8.03	4.76	5.32	10.3	14.7	3.39		
Ho	1.81	1.11	0.95	1.12	1.26	1.74	1.02	1.05	2.12	3.01	0.75		
Er	4.84	3.09	2.39	2.89	3.31	4.83	2.85	2.80	5.75	8.27	2.08		
Tm	0.70	0.46	0.33	0.40	0.46	0.70	0.42	0.41	0.82	1.21	0.30		
Yb	4.50	2.79	2.05	2.46	2.89	4.47	2.60	2.44	5.11	7.79	1.88		
Lu	0.69	0.43	0.31	0.39	0.42	0.71	0.40	0.36	0.80	1.31	0.30		
Ba	196	91.1	636	445	898	97.0	222	150	232	4.39	8.79		
Th	0.54	0.48	5.24	5.98	0.90	0.29	1.02	3.29	14.4	21.2	0.08		
Nb	6.75	3.92	48.7	55.6	7.60	3.34	1.08	20.7	24.3	18.6	0.32		
Y	44.5	28.5	23.9	28.6	30.7	43.4	24.9	26.1	59.8	79.0	18.7		
Hf	5.03	2.68	5.12	5.91	2.55	3.43	1.84	3.33	6.23	9.95	0.86		
Ta	0.48	0.28	3.14	3.60	0.55	0.25	0.09	1.44	2.07	1.92	0.04		
U	0.22	0.21	0.84	1.07	0.72	0.37	0.18	0.71	3.94	3.20	0.04		
Pb	2.09	1.05	4.29	3.66	8.98	0.41	0.47	6.37	32.3	23.0	0.97		
Rb	30.3	7.72	59.6	48.0	37.4	8.24	15.9	50.4	38.1	0.22	0.39		
Cs	0.88	0.21	1.23	0.92	2.05	0.33	0.19	1.88	1.72	0.01	<0.01		
Sr	52.3	58.7	648	480	188	38.5	164	128	817	755	126		
Sc	45.0	33.6	18.2	20.4	38.4	46.4	35.9	24.3	30.0	86.6	41.3		
V	291	270	174	147	237	366	231	174	209	451	229		
Cr	227	550	82.5	74.0	399	235	317	443	177	115	66.8	300	105
Ni	84.1	331	57.7	52.3	194	77.1	71.5	323	71.1	58.3	45.4		
Zr	191	99.6	224	252	98.1	125	66.0	133	245	356	24.4		

FeO_{tot} = All iron is reported as FeO wt%; LOI = H₂O determined from loss of ignition. *Bulk-rock compositions calculated from mineral modes and compositions.

of the matrix with a minor difference in V content (Fig. 5; Table 2). In contrast, a lawsonite-bearing calcite vein in the Ring Mt blueschist (RR-1) is lower in concentrations for most major element oxides (except CaO and K₂O), V, Cr, and Ni as compared with the matrix (Fig. 5; Table 2).

These lawsonite-bearing veins generally follow the REE and TE patterns of the corresponding matrices but differ in REE abundances from the matrices; veins in EC-1B and RR-1 contain lower and higher REE contents, respectively (Fig. 6a).

Zoning Patterns															Normalized to chondrite				
	Fe	Ti	Cr	Fe (wt%)	Ti (wt%)	Cr	Sr	Pb	V	Sr/Pb	Cr/V	La/Dy _N	Dy/Yb _N	La/Yb _N					
Mélanges Ring Mountain, Franciscan Complex LVT-1 (BLS/ECL)	Matrix Lws (BLS; Zone 2)	Core to rim	N.D.	0.57	0.06	319	243	2.60	347	93.6	0.92	0.40	0.26	0.10					
				Max	409	422	4.16	444	101	0.92	0.65	0.82	0.54						
				Avg	364	333	3.38	396	97.5	0.92	0.53	0.54	0.32						
	Matrix Lws (BLS; Zone 3)			0.29	0.01	403	326	2.09	369	127	0.91	0.18	0.33	0.09					
				Max	483	813	4.49	445	181	1.22	0.29	1.10	0.29						
	Avg	441	582	3.60	401	160	1.11	0.26	0.76	0.19									
	Lws Vein (BLS; Zone 2)	Hourglass- sector	Irregular	0.49	0.11	1397	843	10.0	346	78.1	3.40	0.45	2.78	1.25					
				Max	1727	887	11.4	450	85.7	4.53	0.68	3.47	2.37						
	Lws Vein (BLS; Zone 3)			0.49	0.13	1530	866	10.5	397	82.5	3.88	0.59	3.19	1.89					
				Min	473	251	3.41	262	73.6	1.81	0.43	0.70	0.31						
North Berkeley Hills, Franciscan Complex EC-1B (BLS)	Matrix Lws	Oscillatory	N.D.	0.70	0.10	745	483	4.41	356	110	2.30	0.86	0.75	0.65					
				Avg	587	336	4.03	333	84.1	1.78	0.67	0.45	0.31						
				Min	559	658	7.09	498	92.8	1.06	0.25	0.19	0.06						
	Lws in Cal-Vein	Core to rim	N.D.	0.66	0.05	572	916	8.13	527	113	1.15	0.49	0.24	0.09					
				Avg	566	787	7.61	513	103	1.10	0.37	0.22	0.08						
	Matrix Lws	Oscillatory	N.D.	0.27	0.05	37.5	296	0.98	138	57.9	0.20	4.58	2.39	16.2					
				Max	194	593	6.40	290	453	0.77	24.7	5.34	97.3						
	Lws in Cal-Vein	Core to rim	N.D.	0.38	0.13	106	445	3.58	231	181	0.45	10.5	3.98	41.5					
				Mantle*	89.9	201	4.58	227	44.6	0.38	3.79	0.75	2.86						
	North Berkeley Hills, Franciscan Complex EC-1B (BLS)	Matrix Lws	Core to rim	N.D.	0.41	0.21	158	305	4.91	272	66.0	0.58	10.6	1.81	19.4				
Avg					117	243	4.71	245	53.2	0.46	6.52	1.18	9.47						
Mantle*					303	191	1.84	541	104	0.56	6.10	2.48	12.2						
Matrix Lws		Core to rim	N.D.	1.17	0.03	87.7	385	3.66	774	109	0.11	4.70	0.48	2.24					
				Avg	333	235	2.05	621	141	0.56	4.86	2.59	9.57						
Lws Vein		Core to rim	Irregular	Core to rim	1.32	0.19	876	58.1	0.74	515	78.5	1.70	1.59	0.45	0.71				
	Rim*			1.13	0.11	1179	83.3	0.70	466	117	2.57	1.71	0.78	1.35					
	Avg			1.14	0.12	905	68.8	0.75	472	93.7	1.93	1.60	0.96	1.46					

Continued

Table 3: Continued

	Fe	Ti	Cr	Fe (wt%)	Ti (wt%)	Cr	Sr	Pb	V	Str/Pb	Cr/V	La/Dyn	Dy/Yb _N	La/Yb _N
Zoning Patterns														
Normalized to chondrite														
Blind Beach, Franciscan Complex														
SM-8 (BLS)	Matrix Lws	Core to rim, Irregular	Core to rim	Min	0.45	0.03	99.3	71.1	0.45	508	118	0.16	1.06	0.23
				Max	2.09	0.36	284	135	1.14	611	158	0.56	1.44	0.32
				Avg	0.76	0.08	192	103	0.80	560	138	0.36	1.25	0.28
Rio San Juan Complex, Dominican Republic														
IEC15-3.5	Matrix Lws	Irregular	N.D.	Min	0.12	0.02	7.70	323	0.63	46.9	184	0.14	0.86	0.45
(Metagabbro)				Max	0.70	0.24	92.0	682	2.05	241	1083	0.60	3.73	25.7
				Avg	0.30	0.08	33.2	546	1.28	121	518	0.28	1.99	11.6
Coherent Metamorphic Terranes														
Sivrihisar Massif, Turkey														
SV08-283C	Matrix Lws	Irregular	N.D.	Min	0.90	0.03	190	977	25.9	302	30.6	0.53	4.92	5.76
(ECL)				Max	1.74	0.28	424	1391	43.5	361	38.5	1.33	11.3	23.9
				Avg	1.15	0.11	300	1185	36.5	334	32.8	0.91	7.38	13.8
SV01-75A	Matrix Lws	Hourglass-sector, Oscillatory	Oscillatory	Min	1.02	0.03	338	514	11.0	332	46.7	1.02	1.92	2.00
(BLS)	(Zone 1)			Max	2.04	0.67	354	879	15.1	348	58.1	1.02	2.82	2.52
				Avg	1.65	0.41	346	697	13.1	340	52.4	1.02	2.37	2.26
				Min	0.96	0.11	663	630	13.3	337	47.4	1.89	2.28	2.17
Matrix Lws				Max	1.34	0.70	1003	702	13.7	350	51.3	2.98	2.32	2.22
(Zone 2)				Avg	1.22	0.44	833	666	13.5	343	49.4	2.44	2.30	2.19
				Min	0.90	0.03	1134	1139	18.2	260	1.4	2.98	0.03	0.01
Matrix Lws				Max	1.34	0.17	2020	2056	22.4	439	91.8	7.77	1.22	5.61
(Zone 3)				Avg	0.96	0.03	1577	1598	20.3	349	46.6	5.37	0.63	2.81
				Min	0.90	0.02	1007	630	12.7	291	31.1	2.57	1.88	1.23
Matrix Lws				Max	1.51	1.80	2456	1363	20.6	460	85.3	8.45	19.9	32.2
(Zone 4)				Avg	1.24	0.62	1717	868	17.4	377	52.3	4.88	6.92	12.8
				Core*	0.57	0.22	373	102	6.19	402	16.5	0.93	2.75	17.3
SV01-50A	Matrix Lws	Core to rim	Core to rim	Rim*	0.94	0.04	568	294	8.50	511	34.8	1.12	1.07	2.75
(BLS/ECL)	(BLS)			Avg	0.81	0.12	435	206	7.45	450	26.6	0.95	2.07	5.12
				Core*	0.80	0.19	188	116	6.75	291	17.2	0.65	4.64	11.7
				Rim*	1.09	0.05	60.6	401	8.05	328	49.8	0.18	1.52	8.26
				Avg	1.00	0.09	152	337	6.79	320	50.3	0.48	2.17	6.85

Continued

Table 3: Continued

	Fe	Ti	Cr	Fe (wt%)	Ti (wt%)	Cr	Sr	Pb	V	Sr/Pb	Cr/V	La/DyN	Dy/YbN	La/YbN	Normalized to chondrite	
Zoning Patterns																
Lws in Cal-vein (BLS)	Core to rim	Core to rim	Core to rim	Core*	0.73	0.36	124	114	7.51	405	15.1	0.31	2.78	1.74	4.84	
Lws in Cal-vein (ECL)	Irregular	Irregular	Irregular	Rim*	2.23	0.20	1055	215	4.99	331	44.6	3.39	5.41	1.27	7.14	
TZ10-2.2C (Metasomatite)	Matrix Lws	Core to rim, Oscillatory, Irregular	N.D.	Max	1.61	0.14	284	776	10.6	373	73.5	0.76	6.16	2.46	15.2	
Alpine Corsica, France C13 (Metagabbro)	Matrix Lws	Core to rim	N.D.	Core*	0.87	0.02	b.d.l.	596	1.72	1047	347	b.d.l.	0.77	1.39	1.07	
Lws in Cpx (Zone 1)	Core to rim	Hourglass-sector	N.D.	Rim*	0.59	0.03	3.80	610	0.94	1120	750	b.d.l.	0.88	1.16	1.00	
Lws in Cpx (Zone 2)	Core to rim	Hourglass-sector	N.D.	Max	0.71	0.03	1.90	546	1.10	1085	577	b.d.l.	0.83	1.39	1.13	
Lws in Cpx (Zone 3)	Core to rim	Hourglass-sector	N.D.	Max	0.69	0.02	b.d.l.	184	0.00	122	Pb b.d.l.	Cr b.d.l.	0.33	0.92	0.34	
Lws in Cpx (Zone 2)	Core to rim	Hourglass-sector	N.D.	Max	0.74	0.03	b.d.l.	283	2.20	123	Pb b.d.l.	Cr b.d.l.	0.44	1.03	0.41	
Lws in Cpx (Zone 2)	Core to rim	Hourglass-sector	N.D.	Max	0.71	0.02	b.d.l.	234	1.10	122	Pb b.d.l.	Cr b.d.l.	0.39	0.98	0.37	
Lws in Cpx (Zone 3)	Core to rim	Hourglass-sector	N.D.	Max	0.63	0.04	b.d.l.	160	b.d.l.	113	Pb b.d.l.	Cr b.d.l.	0.30	0.45	0.28	
Lws in Cpx (Zone 3)	Core to rim	Hourglass-sector	N.D.	Max	0.76	0.05	b.d.l.	165	b.d.l.	127	Pb b.d.l.	Cr b.d.l.	0.67	0.93	0.30	
Lws in Cpx (Zone 3)	Core to rim	Hourglass-sector	N.D.	Max	0.69	0.05	b.d.l.	162	b.d.l.	120	Pb b.d.l.	Cr b.d.l.	0.49	0.69	0.29	
Lws in Cpx (Zone 3)	Core to rim	Hourglass-sector	N.D.	Max	0.77	0.00	b.d.l.	300	b.d.l.	131	Pb b.d.l.	Cr b.d.l.	0.49	0.15	0.08	
Pinchi Lake, British Columbia BLP4 (Retrogressed ECL)	Lws in Grt	Core to rim, Irregular	Irregular	Min	0.87	0.02	b.d.l.	426	0.48	155	Pb b.d.l.	Cr b.d.l.	0.70	0.23	0.12	
Lws in Grt	Core to rim	Core to rim	Irregular	Max	0.82	0.01	b.d.l.	373	0.18	145	Pb b.d.l.	Cr b.d.l.	0.56	0.18	0.10	
Lws in Grt	Core to rim	Core to rim	Irregular	Max	0.16	0.02	616	202	1.10	74.0	184	7.25	2.76	0.69	1.89	
Lws in Grt	Core to rim	Core to rim	Irregular	Max	0.52	0.32	649	394	1.40	85.0	281	8.77	4.08	1.37	5.57	
Lws in Grt	Core to rim	Core to rim	Irregular	Max	0.17	0.30	633	298	1.25	79.5	233	8.01	3.42	1.03	3.73	

Continued

Table 3: Continued

	Fe	Ti	Cr	Fe (wt%)	Ti (wt%)	Cr	Sr	Pb	V	Sr/Pb	Cr/V	La/Dyn	Dy/Yb _N	La/Yb _N
Zoning Patterns														
Matrix Lws	Core to rim, Oscillatory, Irregular	Irregular	Core to rim, Oscillatory, Irregular	Core*	0.09	193	689	3.80	62.7	181	3.08	2.83	0.24	0.68
				Mantle*	0.06	871	982	58.0	359	16.9	2.43	17.4	10.9	240
				Rim*	0.07	437	452	11.2	210	57.1	2.14	5.89	3.12	16.5
				Avg	0.30	629	740	33.9	270	49.4	2.38	11.3	6.71	127
BLR5 (ECL)	Core to rim, Irregular	Core to rim, Irregular	Irregular	Min	0.02	123	369	2.49	264	76.4	0.46	2.13	0.59	2.25
	Irregular	Irregular	Irregular	Max	0.09	823	796	9.99	477	148	1.87	6.17	1.06	3.01
Matrix Lws	Core to rim, Irregular	Irregular	Core to rim	Avg	0.19	522	582	5.80	400	111	1.21	3.68	0.91	2.57
	Irregular	Irregular	Core to rim	Mantle*	0.08	491	708	5.68	426	147	1.31	4.20	0.86	2.91
Lws in Ph-vein	Core to rim	Core to rim	Core to rim	Rim*	0.04	198	483	9.36	568	63.0	0.32	18.3	3.62	70.5
	Oscillatory	Core to rim	Core to rim	Avg	0.07	403	641	6.78	469	121	1.01	8.43	1.69	23.2
				Mantle*	0.37	838	379	6.99	579	54.2	1.45	3.39	4.73	16.0
				Rim*	0.08	119	606	18.5	433	33.9	0.29	19.3	4.88	92.1
Sr	9184	445	499	Avg	0.17	343	514	13.3	458	50.3	0.72	13.0	4.64	61.5
	8.50	8.08	2.10	78.1	939	4.38	1185	0.33	1784	546	740	4.49	641	1.03
V	b.d.l.	231	399	14.7	2.65	48.4	3.49	54.3	25.3	b.d.l.	0.94	62.2	1.35	73.4
	b.d.l.	106	415	475	357	81.8	334	48.7	1210	1085	270	47.8	469	53.4
Cr	b.d.l.	7.88	0.03	206	1238	154	300	143	176	1.90	629	348	403	74.3
	2.04			10.2	3.42	1.65	0.06	2.85	23.0	0.08	2.21	0.54	0.07	0.79

Concentrations are reported in g/g unless otherwise indicated; b.d.l. = below than detection limit (i.e. analyzed but not detected). *The average compositions of lawsonite with Fe-concentric zoning at core, mantle, and rim.

Table 4: Average TE concentrations of lawsonite, garnet, and apatite in the matrix

Locality	Coherent metamorphic terranes									
	Mélange areas					Tavşanlı Zone				
	North Berkeley Hills/Franciscan Complex	Ring Mountain/Franciscan Complex	Blind Beach/Franciscan Complex	Rio San Juan Complex	BLS	BLS	BLS-facies Metagabbro	SV01-75A	SV08-283C	TTZ10-2.2C
Rock type	BLS	BLS	BLS/ECL	BLS	BLS	BLS	ECL	ECL	Metaso-matite	Lws-Omp Metagabbro
Sample	EC-1B	RR-1	LVT-1	SM-8	IEC15-3.5	SV01-75A	SV08-283C	TTZ10-2.2C	C13	BLR4
Mineral	Lws	Ap	Lws	Lws	Lws	Lws	Grt	Lws	Lws	Grt
La	20.4	5.20	69.4	1.00	3.24	7.78	0.09	9.07	5.20	54.9
Ce	62.6	17.0	146	3.04	11.0	19.4	0.23	21.3	13.3	124.2
Pr	10.7	5.40	16.9	0.50	2.22	2.63	0.02	2.88	1.86	15.3
Nd	60.3	42.8	67.5	2.98	14.0	32.2	0.27	13.5	9.58	68.3
Sm	18.6	31.6	14.2	1.18	6.19	7.86	b.d.l.	3.62	3.56	16.2
Eu	3.79	33.0	6.13	0.41	2.40	3.02	0.06	0.84	1.44	1.36
Gd	19.5	194	11.7	2.15	9.23	7.75	0.66	4.82	6.41	3.22
Tb	2.50	50.4	1.61	0.43	1.67	0.69	0.45	0.79	1.03	0.50
Dy	12.5	453	8.69	3.79	11.8	4.33	9.25	5.89	7.34	3.54
Ho	1.80	124	1.36	1.02	2.56	0.83	4.04	1.12	1.73	0.77
Er	4.04	486	3.17	3.93	7.50	1.91	18.7	2.17	5.53	1.99
Tm	0.49	90.8	0.30	0.64	1.09	0.21	3.46	0.15	0.83	0.26
Yb	2.78	540	1.59	4.60	7.23	1.44	23.8	0.49	6.57	1.74
Lu	0.36	91.4	0.19	0.81	1.25	0.12	3.45	0.04	1.36	0.29
Ba	5.66	0.00	28.1	0.24	2.35	2.51	0.79	0.34	3.00	0.27
Th	0.88	0.00	3.85	0.04	0.03	0.50	0.10	4.00	3.13	0.01
Y	40.1	3678	38.5	29.0	65.9	23.1	116	33.2	51.4	19.4
Hf	0.04	b.d.l.	0.27	b.d.l.	0.23	0.09	0.02	b.d.l.	0.70	0.04
U	0.49	b.d.l.	1.13	b.d.l.	0.22	0.26	b.d.l.	0.44	0.58	0.89
Pb	2.05	28.0	3.58	3.53	0.60	1.28	0.38	36.5	36.8	1.10
Rb	0.61	b.d.l.	0.01	0.02	b.d.l.	1.84	0.28	b.d.l.	0.01	0.09
						0.69	0.69			0.08
						0.26	0.26			0.03
						0.74	0.74			0.02
						0.02	0.02			0.03
						0.03	0.03			0.03
						0.26	0.26			0.03
						0.08	0.08			0.05

Concentrations are reported in $\mu\text{g/g}$; b.d.l. = below than detection limit (i.e. analyzed but not detected).

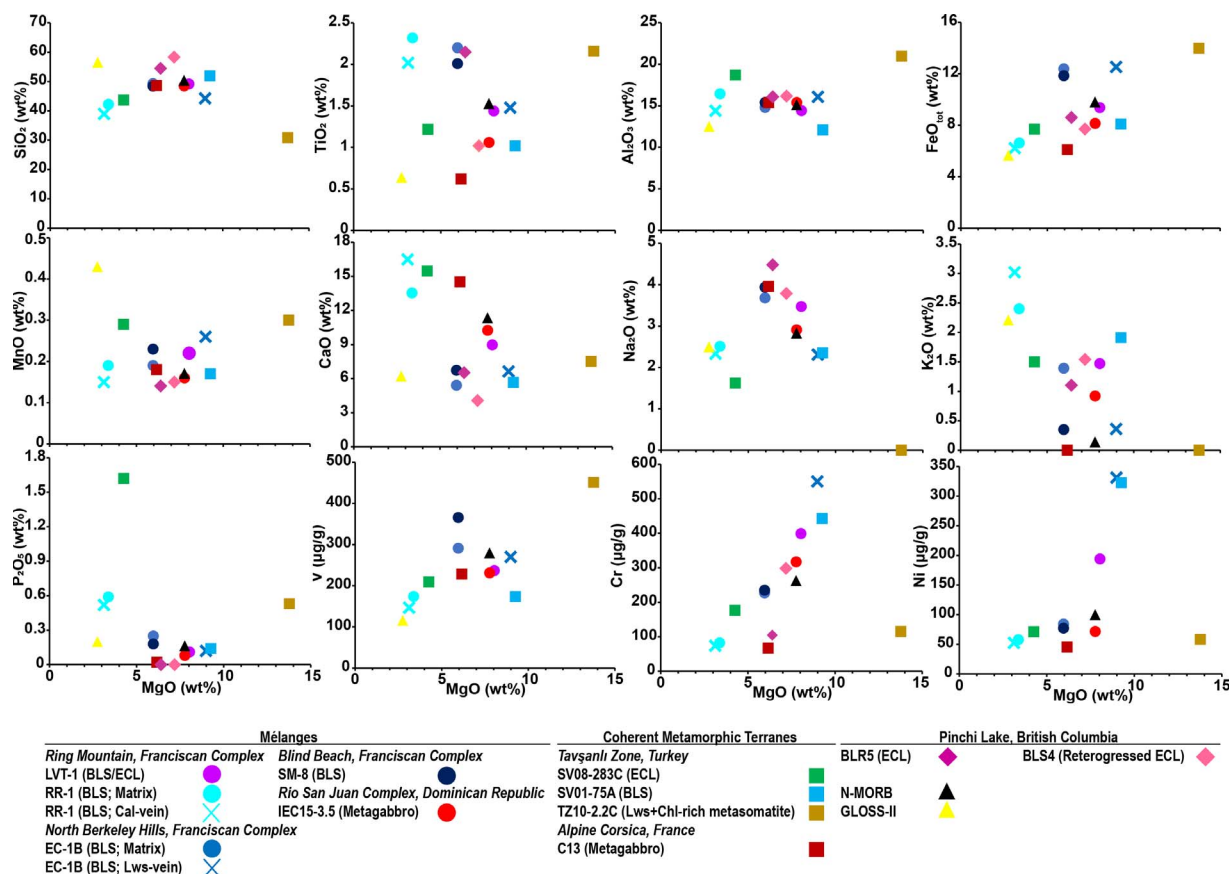


Fig. 5. Variation diagrams between MgO and other major and TEs (V, Cr, Ni). Also shown are N-MORB (Gale et al., 2013) and GLOSS-II (Plank, 2014) compositions.

Lawsonite composition

Lawsonite grains in various textural sites (i.e. garnet, matrix, and vein) with diverse textural features (e.g. included in the dissolved domains of omphacite) were analyzed in this study. Analyzed matrix grains are free of textural replacement, except a few grains replaced by calcite in the Rio San Juan blueschist-facies metagabbro (IEC15-3.5).

Transition-metal element compositions

Analyzed lawsonite contains detectable amounts of Cr and/or Ti, with Fe as the most common substituent (up to 2.9 wt% FeO_{tot}) (Table 3; Supplementary Table S1). Fe content is negatively correlated with the sum of Al, Ti, and Cr contents when these elements are expressed in atoms per formula unit (Fig. 7). This negative correlation is lost when Ti or Cr is plotted against Al + Cr + Fe and Al + Ti + Fe, respectively. This confirms that Fe is the most common substituent for Al relative to Cr and Ti, and it is likely present as Fe^{3+} . Most lawsonite grains have Cr and Ti contents <1 wt%, but in the Sivrihisar blueschist SV01-75A, Cr and Ti are incorporated at the weight percent level (~1 wt% Cr_2O_3 , ~3 wt% TiO_2) (Table 3; Supplementary Table S1).

Fe, Ti, and Cr zoning in lawsonite

Analyzed lawsonite grains show characteristic zoning patterns in Ti, Fe, and Cr, as has been documented in previous studies (Mevel & Kienast, 1980; Ueno, 1999; Vitale Brovarone et al., 2014; Fornash et al., 2019; Whitney et al., 2020). The types of zoning patterns mostly differ by element and vary from sample to sample. Fe is commonly concentrically zoned (Fig. 8a); some grains have an Fe-richer rim and others have an Fe-richer core. Some grains exhibit irregular zoning patterns in Fe (Fig. 8b; Table 3); in particular, grains that have been partially replaced by other phases, such as calcite (i.e. Rio San Juan blueschist-facies metagabbro IEC15-3.5). In the Tavşanlı metasomatite (TZ10-2.2C), Fe zoning is patchy in the core, and between the core and rim regions there are domains of higher or lower Fe (Fig. 8c).

Ti in lawsonite typically displays either concentric or hourglass sector zoning (Table 3). Ti-hourglass zoning is characterized by a locally preserved Ti-intermediate core (Zone 1), Ti-rich mantle (Zone 2), and Ti-poor rim (Zone 3) (Fig. 8d and e); we adopt here the zone definitions used by Vitale Brovarone et al. (2014) for lawsonite in Corsica metasomatites. An additional outermost rim domain (Zone 4) with variable Ti-content and prominent Cr-oscillatory zoning (Fig. 8e) was recognized in lawsonite from the Sivrihisar blueschist (SV01-75A), which has the

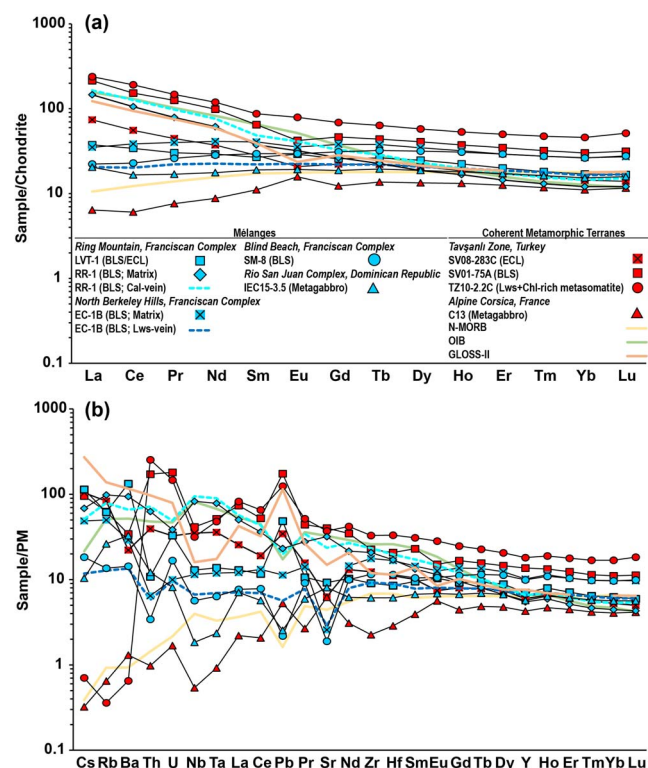


Fig. 6. REE and TE abundances of lawsonite-bearing metabasites, metasomatite, and veins. (a) Chondrite-normalized REE patterns. (b) PM-normalized TE patterns. Normalization values are from [Palme & O'Neill \(2007\)](#) (PM), [Sun & McDonough \(1989\)](#) (Chondrite, N-MORB, OIB), and [Plank \(2014\)](#) (GLOSS-II).

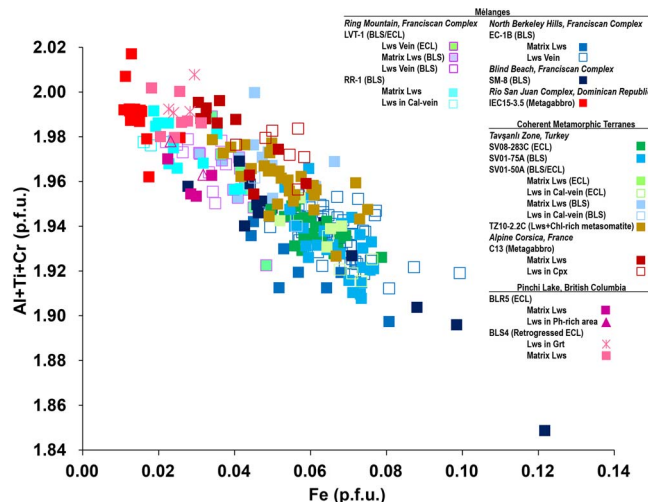


Fig. 7. Al + Ti + Cr vs. Fe in lawsonite.

highest Cr concentration (up to 1 wt% Cr_2O_3). Ti zoning in some grains correlates with the same type of zoning in Fe; e.g. in the Franciscan (Ring Mt) blueschist RR-1, matrix lawsonite locally shows Fe-oscillatory zoning that negatively correlates with Ti-oscillatory zoning (Fig. 8f).

Cr zoning is rare in the lawsonite analyzed, but where present, it generally follows Fe zoning (Table 3). A few exceptions include matrix lawsonite in the Sivrihisar blueschist (SV01-75A) (Fig. 8e). The Pinchi Lake grains

in the phengite-rich region (BLR4) display Fe-oscillatory zoning, with Cr-rich domains in the core (Fig. 8g).

REE compositions

REE in lawsonite vary in concentrations and patterns at both sample and grain scales. In general, matrix lawsonite from metabasite samples displays LREE enrichments relative to middle REE (MREE) and/or heavy REE (HREE) ($\text{La}/\text{Dy}_\text{N} = 1\text{--}24.7$, $\text{La}/\text{Yb}_\text{N} = 1.03\text{--}97.3$) with a slight-to-significant negative Eu anomaly (Fig. 9a and b; Table 3). Matrix lawsonite from the Rio San Juan blueschist-facies metagabbro (IEC15-3.5), one Franciscan blueschist (Ring Mt RR-1), and the Corsica Lws-Omp metagabbro (C13) show a positive Eu anomaly (Fig. 9a and b).

In the Pinchi Lake retrogressed eclogite (BLR4), lawsonite in garnet is lower in REE ($1 \times \sim 10 \times$ chondrite) than matrix lawsonite ($\sim 1 \times \sim 1000 \times$ chondrite) and it displays a positive REE pattern ($\text{La}/\text{Yb}_\text{N} = 1.89\text{--}5.57$) with a positive Eu anomaly (Fig. 9c). Matrix lawsonite is generally enriched in LREE relative to M-HREE ($\text{La}/\text{Yb}_\text{N} = 1.45\text{--}856$) with no Eu anomaly (Fig. 9c; Table 3). Lawsonite in garnet and in the matrix from the Pinchi Lake eclogite (BLR5) mostly show nearly flat REE patterns with a few matrix grains displaying positive-sloped patterns ($\text{La}/\text{Yb}_\text{N} = 1.29\text{--}129$) (Fig. 9d). Lawsonite in phengite-rich areas of eclogite BLR5 exhibits a positive-sloped REE pattern ($\text{La}/\text{Yb}_\text{N} = 15.1\text{--}116$) and has higher REE contents ($\sim 1 \times \sim 1000 \times$ chondrite) than matrix lawsonite and lawsonite inclusions in garnet ($\sim 1 \times \sim 100 \times$ chondrite) (Fig. 9d).

Lawsonite in the Tavşanlı metasomatite (TZ10-2.2C) contains higher REE concentrations than lawsonite in Sivrihisar metabasites (SV01-75A, SV08-283C) ($\sim 1 \times \sim 1000 \times$ chondrite vs. $1 \times \sim 100 \times$ chondrite) with great variations in REE patterns ($\text{La}/\text{Yb}_\text{N} = 0.09\text{--}1.95$) (Fig. 9b and e) and a slight negative-to-positive Eu anomaly (Fig. 9e). In the Sivrihisar blueschist/eclogite (SV01-50A), no significant differences were observed in REE patterns and concentrations between lawsonite in the matrix and in the calcite-vein (Fig. 9b and e). In contrast, vein-lawsonite in two Franciscan blueschists (N Berkeley Hills EC-1B, Ring Mt RR-1) differs in REE patterns and concentrations from matrix lawsonite in the same samples; vein-lawsonite mostly shows a negative-sloped REE pattern ($\text{La}/\text{Yb}_\text{N} = 0.11\text{--}0.94$) with lower LREE concentrations ($\sim 1 \times \sim 100 \times$ chondrite vs. $\sim 1 \times \sim 1000 \times$ chondrite) (Fig. 9a and f). In the Corsica Lws-Omp metagabbro (C13), lawsonite that occurs in the partially replaced areas of omphacite exhibits great variations in REE patterns ($\text{La}/\text{Yb}_\text{N} = 0.08\text{--}0.41$) with higher REE concentrations ($\sim 1 \times \sim 100 \times$ chondrite vs. $\sim 1 \times \sim 10 \times$ chondrite) compared with matrix lawsonite (Fig. 9b). Vein-lawsonite consisting of lawsonite-rich layers (veins) in Ring Mt blueschist/eclogite (LVT-1) has L-MREE concentrations higher than matrix lawsonite in the blueschist layers of LVT-1 ($\sim 1 \times \sim 100 \times$ chondrite vs.

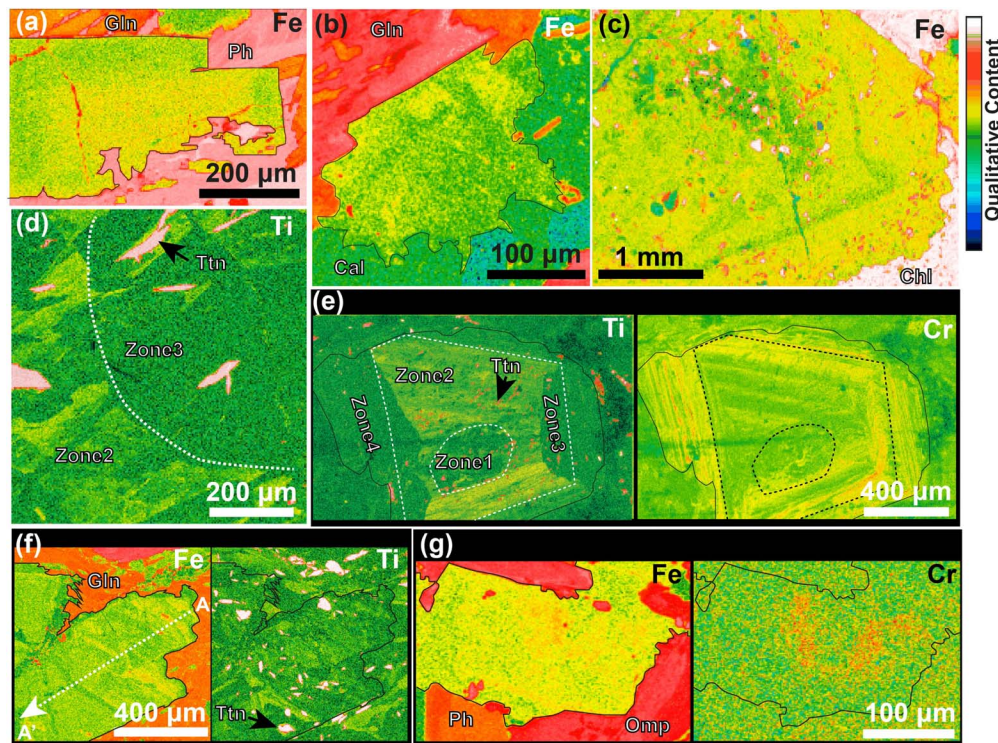


Fig. 8. Fe, Ti, and Cr zoning patterns in lawsonite. (a) Fe concentric zoning in matrix lawsonite (Ring Mountain blueschist/eclogite LVT-1). (b) Irregular Fe zoning in matrix lawsonite, partially replaced by calcite (Rio San Juan metagabbro IEC15-3.5). (c) Irregular Fe zoning in matrix lawsonite with oscillatory pattern toward the rims (Tavşanlı metasomatite TZ10-2.2C). (d) Ti hourglass zoning in lawsonite-vein with Ti-rich (Zone 2) and Ti-poor (Zone 3) domains (Ring Mountain blueschist/eclogite LVT-1). White dotted line indicates the boundary between different zones. (e) Ti and Cr zoning in one lawsonite crystal in the matrix (Sivrihisar blueschist SV01-75A). Ti-hourglass zoning shows four distinct domains, including Ti-intermediate (Zone 1), Ti-rich (Zone 2) and Ti-poor (Zone 3) domains. Zone 4 is the outermost domain with prominent Cr-oscillatory zoning and a range of Ti content. White and black dotted lines indicate the boundaries between different zones. (f) Fe and Ti oscillatory zoning in one lawsonite crystal in the matrix (Ring Mountain blueschist RR-1). (g) Fe and Cr zoning in one lawsonite crystal in a phengite-rich area (Pinchi Lake eclogite BLR5). Black solid lines: grain boundaries of lawsonite grains.

$1 \times -10 \times$ chondrite; Fig. 9a and f) and it shows variable REE patterns; vein-lawsonite in garnet blueschist layers show LREE enrichments ($\text{La/Yb}_N = 0.31-2.37$), whereas that in eclogite layers show HREE enrichments ($\text{La/Yb}_N = 0.06-0.65$) (Fig. 9f; Table 3).

Other TE compositions

Pb, Sr, V, and Cr are present at trace levels in most analyzed lawsonite grains (Tables 3 and 4; Supplementary Table S1), with concentrations varying at the intragrain and intergrain scales. A compilation of published data and our analyses suggests that Pb and Sr contents in lawsonite are positively correlated, with distinct trends in Sr/Pb (Martin et al., 2014; Fornash et al., 2019; Whitney et al., 2020); lawsonite in quartz-rich metasediment tends to have lower Sr/Pb than lawsonite in metabasite (4–25 vs. >30; Fig. 10a). Lawsonite in calcschist generally has Sr/Pb within the range of lawsonite in metabasite (>30; Supplementary Table S1).

Most analyzed matrix grains have Sr/Pb comparable with lawsonite from metabasalt with moderate Sr/Pb (30–50; Fig. 10a), with a few exceptions: the Pinchi Lake eclogites (BLR4, 5) and Sivrihisar blueschist/eclogite (SV01-50A). In the Pinchi Lake retrogressed eclogite (BLR4), some matrix grains have lower Sr/Pb in the region

between core and rim than in the core/rim regions (16–18 vs. 30–181; Table 3). In contrast, matrix grains in the Sivrihisar blueschist/eclogite (SV01-50A) have lower Sr/Pb in the core than in the mantle/rim regions (17–20 vs. 29–71; Table 3). These low Sr/Pb ratios are comparable with those of lawsonite from quartz-rich metasediment (4–25; Whitney et al., 2020).

Intergrain-scale variations were observed in the Rio San Juan blueschist-facies metagabbro (IEC15-3.5); matrix grains replaced by calcite have a lower range of Sr/Pb than those with no replacement (184–542 vs. 185–1083; Fig. 10b and c). Notwithstanding the different field settings of the host subduction complex, matrix grains from metagabbro (Corsica C13, Rio San Juan IEC15-3.5) have a higher range of Sr/Pb than those from metabasalt (129–1775 vs. 16–453; Fig. 10b and c). The extent of Sr/Pb variations is significantly high in lawsonite from metagabbro as compared with lawsonite from metabasalt (Fig. 10b and c).

Lawsonite in veins and in domains such as the partially replaced areas of omphacite in Corsica C13 and the phengite-rich area in the Pinchi Lake eclogite BLR5 mostly show a positive correlation between Pb and Sr (Fig. 10d). These grains have a range of Sr/Pb that differs from matrix grains and/or garnet inclusions in the

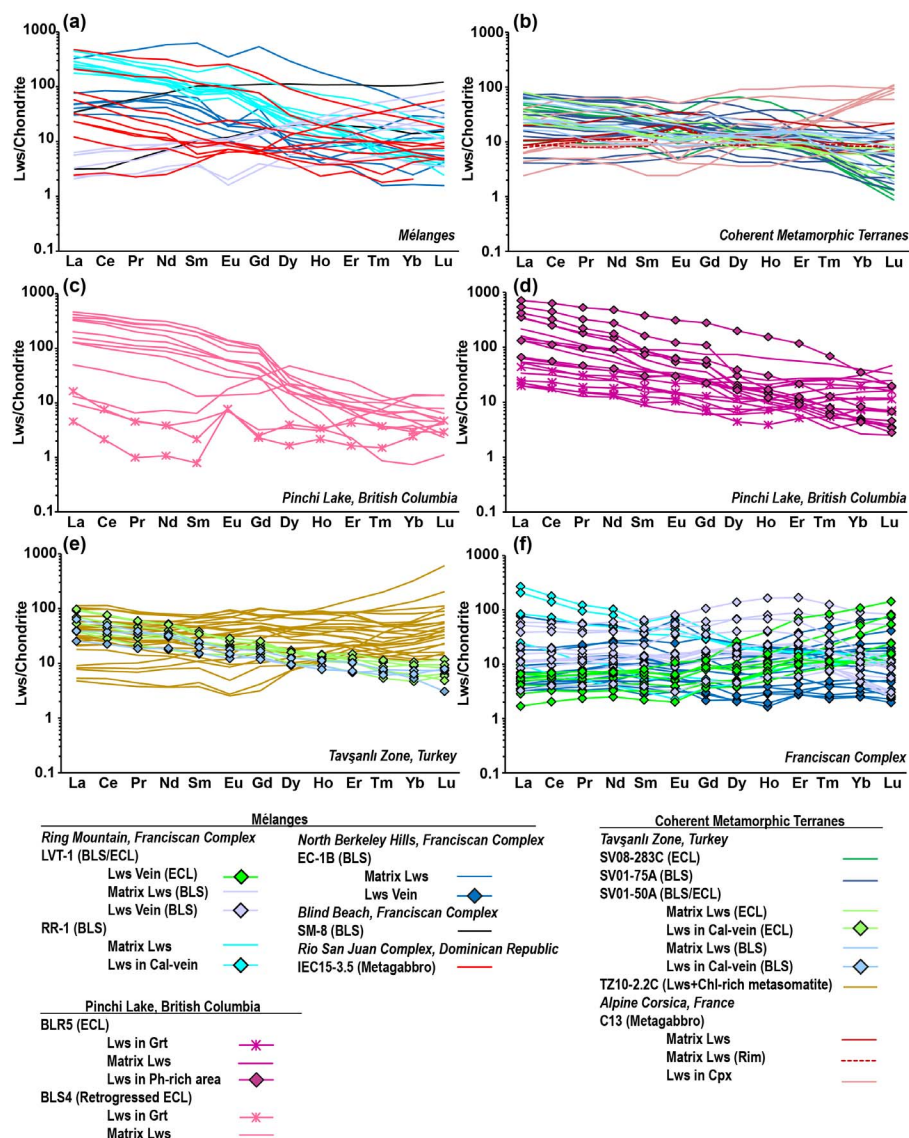


Fig. 9. REE patterns of lawsonite. (a, b) Matrix lawsonite in metabasite from mélange (a) and coherent-metamorphic terranes (b). (c, d) Lawsonite in Pinchi Lake retrogressed eclogite (c) and eclogite (d). (e) Lawsonite in the matrix from Tavşanlı metasomatism and in the vein from Sivrihisar metabasalt. (f) Lawsonite in the vein from Franciscan metabasalts.

same samples. Matrix lawsonite in the Ring Mt blueschist (RR-1) has a higher range of Sr/Pb than lawsonite in the calcite-rich vein (Fig. 10b-d; Table 3). In the Pinchi Lake eclogites (BLR4, 5), lawsonite in garnet has Sr/Pb similar to or higher than matrix lawsonite, whereas lawsonite in the phengite-rich areas of the eclogite (BLR5) has Sr/Pb similar to matrix lawsonite (Fig. 10b-d).

V concentration in lawsonite ranges from ~39–2866 ppm (Table 3; Supplementary Table S1). In general, lawsonite in quartz-rich metasediment has a similar range of V but a lower range of Cr compared with lawsonite in calcschist and metabasite (Supplementary Table S1). The V and Cr abundances of most matrix grains are positively correlated (Fig. 10e), but in some cases display intragrain variations (Fig. 10b, c and e). Matrix lawsonite from the Sivrihisar blueschist (SV01-75A) shows the most significant variations in Cr/V, with Cr/V values increasing from Zone 1 to Zone 4

(Fig. 10b; Table 3). In contrast to matrix grains, some vein-associated lawsonite exhibit no obvious correlation between Cr and V (Fig. 10f). Vein-lawsonite in Franciscan metabasalts (EC-1B, LVT-1) has higher Cr/V than matrix lawsonite (Fig. 10b and c; Table 3).

Garnet composition

Matrix garnets in the Sivrihisar blueschist and eclogite (SV01-75, SV08-283C) and the Pinchi Lake eclogites (BLR4, 5) were analyzed. Matrix garnet in the Sivrihisar blueschist (SV01-75A) and eclogite (SV08-283C) is Fe- and Ca-rich, and it is compositionally zoned with the core depleted in Fe and enriched in Mn relative to the rim; core: alm₅₀₋₅₈sps₆₋₁₆prp₄₋₁₀grs₂₅₋₃₀; rim: alm₅₅₋₆₂sps₄₋₈prp₇₋₁₀grs₂₄₋₃₁ (Fig. 11a). An oscillatory zoned outer-rim is locally preserved on garnet from the Sivrihisar eclogite, with higher Mn correlated with lower Fe; Mn-rich region: alm₅₁₋₅₂sps₁₄₋₁₅prp₆grs₂₇₋₂₈;

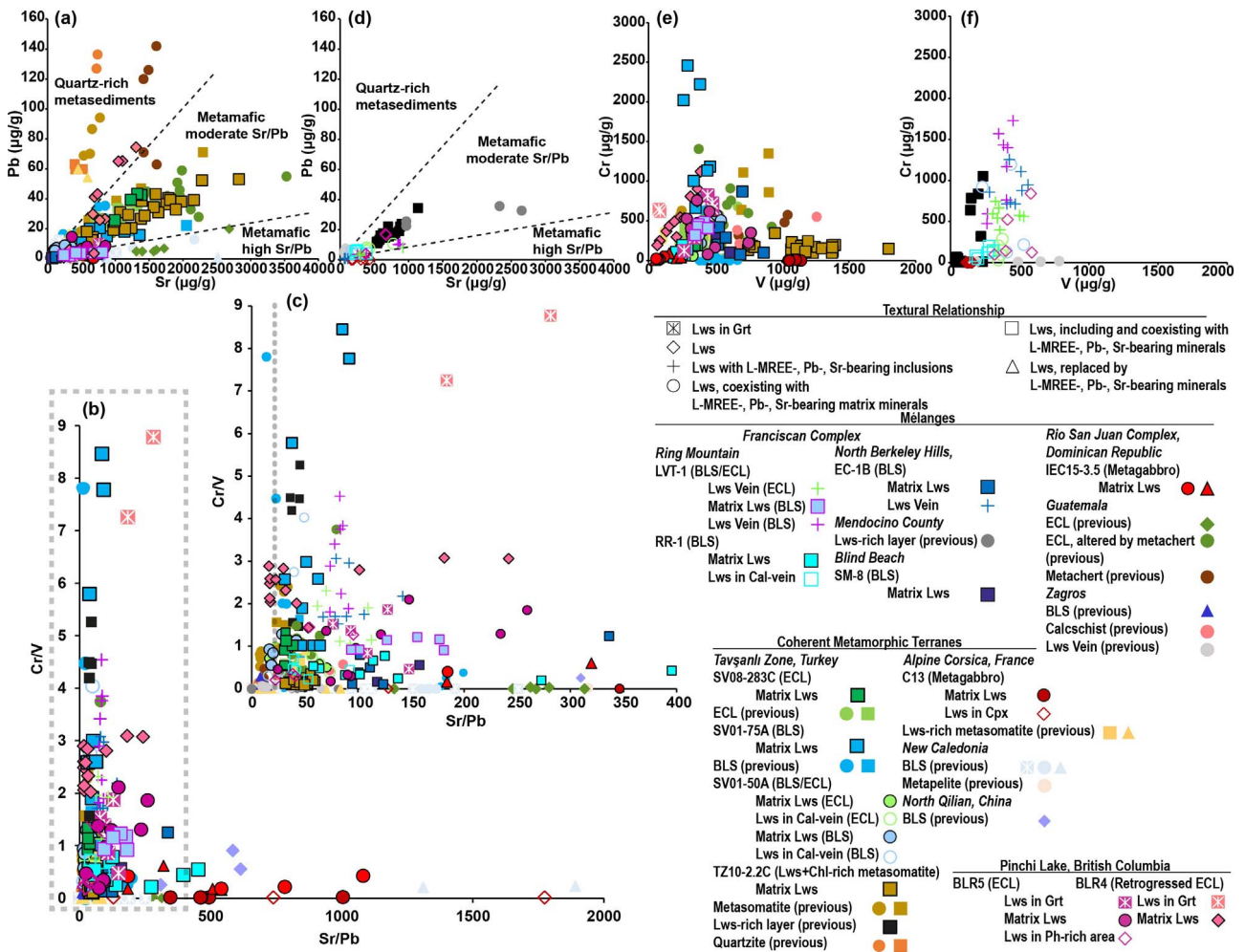


Fig. 10. (a) Pb vs. Sr in lawsonite in garnet and matrix (modified from Whitney *et al.*, 2020). (b, c) Plots of Cr/V vs. Sr/Pb in lawsonite. Inset of Fig. 10(c) is a magnified image, showing a selected area of the original plot (marked with dashed lines). Gray dotted line delineates the range of Sr/Pb in lawsonite from quartz-rich metasediment (<25) vs. lawsonite from metabasite (>25). (d) Pb vs. Sr in lawsonite that occurs in texturally distinct domains (e.g. veins, partially dissolved areas of omphacite, phengite-rich area). (e, f) Cr vs. V in lawsonite that occurs in various textural sites (e: matrix, garnet; f: texturally distinct domains). Also shown are previous analyses of lawsonite from Franciscan Complex (Martin *et al.*, 2014), South Motagua Mélange Zone/Guatemala (Hara *et al.*, 2018), Sivrihisar Massif/Turkey (Fornash *et al.*, 2019; Fornash & Whitney, 2020), Alpine Corsica/France (Martin *et al.*, 2014; Vitale Brovarone *et al.*, 2014), Western Alps/France (Lefevre *et al.*, 2020), New Caledonia (Spandler *et al.*, 2003), North Qilian/China (Xiao *et al.*, 2013), and Zagros Orogen/Iran (Muñoz-Montecinos *et al.*, 2021).

Mn-poor region: $\text{alm}_{57-59}\text{sps}_{7}\text{prp}_{7}\text{grs}_{27-28}$ (Fig. 11a). In the Sivrihisar blueschist, matrix garnet is locally surrounded by a Mn-rich outer-rim with the composition of $\text{alm}_{50-54}\text{sps}_{12-19}\text{prp}_{4}\text{grs}_{27-30}$. From core to oscillatory zoned outer-rim, matrix garnet in the eclogite shows noticeable increases in REE abundances ($\sim 1 \times - \sim 100 \times$ vs. $\sim 1 \times - \sim 1000 \times$ chondrite; Fig. 11b). This contrasts to the Mn-rich outer-rim on matrix garnet in the blueschist, which has M-HREE abundances similar to core-to-rim ($\sim 1 \times - \sim 100 \times$ chondrite; Fig. 11b). Matrix garnet in both eclogite and blueschist is characterized by a negative-sloped pattern ($\text{Dy}/\text{Yb}_N = 0.02-0.37$).

Matrix garnet in the Pinchi Lake eclogites (BLR4, 5) displays a gradual decrease in Mn, but increases in Fe and Mg from core to rim (Fig. 11a); core: $\text{alm}_{44-68}\text{sps}_{2-33}\text{prp}_{3-10}\text{grs}_{20-23}$; rim: $\text{alm}_{64-68}\text{sps}_{1-5}\text{prp}_{9-12}\text{grs}_{20-23}$. This garnet is characterized by the enrichment of MREE relative to LREE and HREE ($\text{La}/\text{Dy}_N \leq 0.02$; $\text{Dy}/\text{Yb}_N = 1.60-4.91$), with

lower M-HREE concentrations in the rim than the core ($\sim 10 \times - \sim 100$ vs. $\sim 10 \times - \sim 1000 \times$ chondrite; Fig. 11c).

Apatite composition

The REE composition of one apatite grain in the matrix of the N Berkeley Hills blueschist (EC-1B) was analyzed. This matrix grain has a negative-sloped REE pattern ($\text{La}/\text{Yb}_N \leq 0.02$) and its REE contents increase from core to rim (Fig. 11d).

DISCUSSION

Lawsonite composition can vary in response to (1) the compositional evolution of the host rock, (2) changes in relative mineral abundances, (3) element-fractionation with minerals, and/or (4) element selectivity that differs for each growth plane of lawsonite. We evaluate the influence of each factor on lawsonite composition

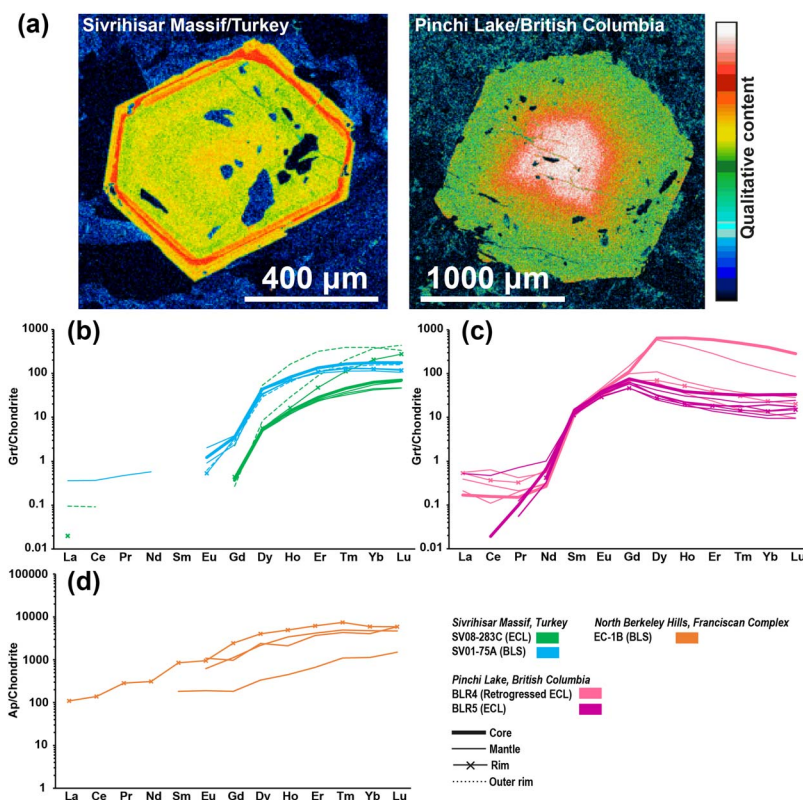


Fig. 11. (a) Mn zoning patterns in matrix garnet from Sivrihisar eclogite (SV08-283C) and Pinchi Lake eclogite (BLR5). (b, c) REE patterns of matrix garnet in Sivrihisar metabasalts (SV08-283C, SV01-75A) (b) and Pinchi Lake eclogites (BLR4, 5) (c). (d) REE patterns of matrix apatite in North Berkeley Hills blueschist (EC-1B).

to investigate metamorphic reaction history, fluid–rock interactions, and the potential sources of fluids in subduction systems.

Compositional evolution of lawsonite-bearing host rocks

The lawsonite-bearing samples have a range of bulk-rock compositions (e.g. metabasite, metasomatite; Table 2), which may have deviated from their protolith compositions owing to seafloor alteration, metamorphic reactions, metasomatism, and/or mechanical mixing. Any changes in host rock compositions during lawsonite growth likely induced compositional variations in lawsonite. To evaluate the controls of host rock signatures on lawsonite composition, it is critical to understand the compositional evolution of host rocks.

Based on a V vs. Ti/1000 diagram, most metabasalt samples from Franciscan mélange areas and the Sivrihisar coherent terrane plot in the MORB/back-arc basin basalt (BABB) field (Fig. 12a), except a Ring Mt blueschist (RR-1) that has an oceanic island basalt (OIB) signature (Fig. 12a). This is partially inconsistent with the results of an Hf–Th–Ta diagram, in which the Sivrihisar eclogite (SV08-283C) and blueschist (SV01-75A) are classified as having a calc-alkaline basalt (CAB) and a within-plate basalt signature, respectively (Fig. 12b). This inconsistency is not surprising given the high bulk Th contents of the Sivrihisar metabasalts (Table 2), which result in high Th/La, causing the Sivrihisar metabasalts

plot outside the MORB field in a Th/La vs. Sm/La diagram (Fig. 12c). Th/La has been interpreted to be less fractionated during subduction and its high value (i.e. >0.35 in terrigenous sediment) is likely derived from interactions with (meta)sediment (Plank, 2005) and/or (meta)sediment-sourced fluids before or during subduction metamorphism (Fig. 12c). The high K₂O content and the LREE and LILE enrichments of the Sivrihisar metabasalts further support the possible influence of (meta)sediment (Figs 5 and 6). This interpretation is consistent with high Nb/Cr ratio in matrix rutile observed in Sivrihisar metabasalt, which is comparable with rutile in metapelite (Fornash & Whitney, 2020). In the case of Sivrihisar blueschist (SV01-75A), Cr and Ni enrichments (Fig. 5) indicate the additional influence of ultramafic rocks (e.g. serpentinite, metagabbro). Serpentinite can also act as a source for LILE (e.g. Cs and Ba) (Tenthorey & Hermann, 2004; Deschamps et al., 2011, 2013).

The Franciscan metabasalts, classified as MORB/BABB, are enriched in LILE and LREE, with higher K₂O content relative to N-MORB (Figs 5 and 6). This suggests some extent of interaction with (meta)sediment or (meta)sediment-sourced fluid, possibly via mechanical mixing, which is common in mélange. Alternative LILE sources include serpentinite (Scambelluri et al., 2001, 2004; Tenthorey & Hermann, 2004), altered basaltic crust (Ryan et al., 1995; Becker et al., 2000; John et al., 2008), and/or fluids sourced from these lithologies. The high Cr and Ni contents of the Ring Mt blueschist/eclogite

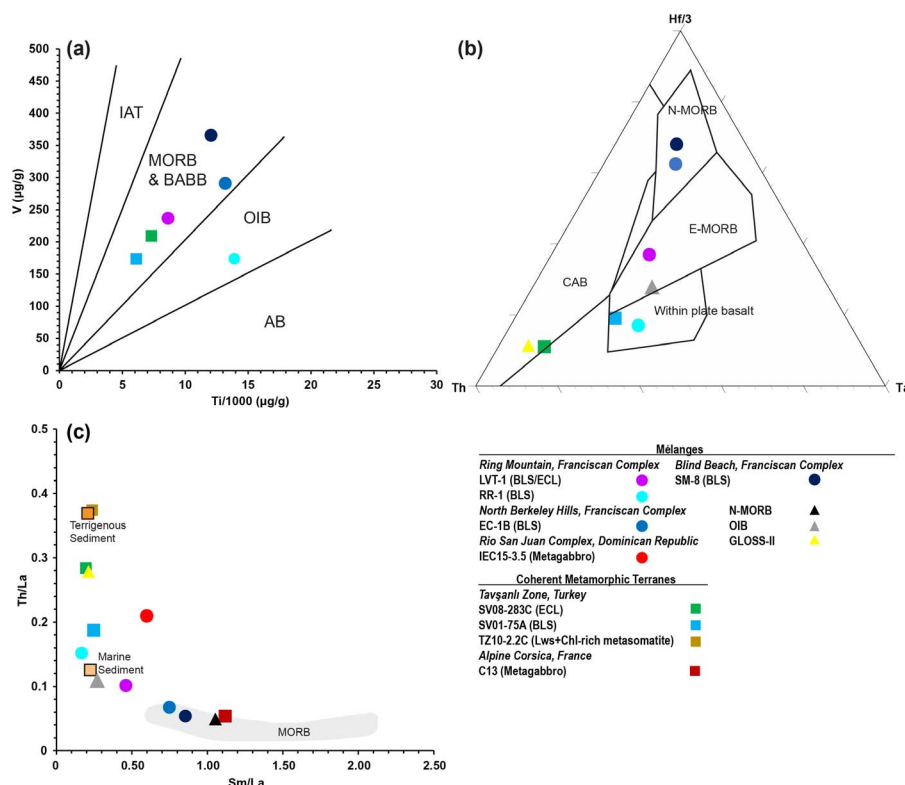


Fig. 12. Discrimination diagrams for metabasite and metasomatite. (a) V vs. Ti/1000 (Shervais, 1982). (b) Hf/3 vs. Th vs. Ta (Wood, 1980). (c) Th/La vs. Sm/La (Plank, 2005, 2014). Abbreviations: N-MORB, normal mid-ocean ridge basalt; E-MORB, enriched mid-ocean ridge basalt; BABB, back-arc basin basalt; IAT, island arc tholeiite; OIB, oceanic island basalt; AB, alkali basalt; CAB, calc-alkaline basalt.

(LVT-1) indicate additional interactions with ultramafic rocks (Fig. 5). The LILE and LREE enrichments of the Ring Mt blueschist (RR-1) could have been inherited from the composition of an OIB protolith (Figs 6 and 12a,b).

The Tavşanlı metasomatite (TZ10-2.2C) records high Th/La—comparable with the ratio of terrigenous sediment (Fig. 12c)—but it shows significant K₂O and LILE depletions (Figs 5 and 6b). The high MgO and significantly low SiO₂ of this metasomatite indicate an ultramafic protolith (Fig. 5; Table 2), consistent with low K₂O and LILE abundances. The high Th/La and the LREE enrichment, however, suggest interactions with terrigenous sediment during metasomatism.

The Corsica Lws-Omp metagabbro (C13) has no clear compositional indication of alteration by (meta)sediment, whereas the Rio San Juan blueschist-facies metagabbro (IEC15-3.5) preserves evidence for interactions with (meta)sediment (e.g. high Th/La, bulk enrichments in LILE and LREE relative to N-MORB; Figs 6 and 12c). Except for the Corsica metagabbro, all samples preserve compositional signatures supporting interactions with (meta)sediment ± metagabbro/serpentinite, although it is unclear when and how the samples obtained these signatures.

Bulk-rock-normalized element concentrations and element budget of lawsonite

Lawsonite composition can be closely related to the host rock composition, but this may be simultaneously

influenced by other factors (e.g. changes in mineral modes). To interpret controls of host rock signatures on lawsonite composition, the TE composition of matrix lawsonite was averaged and normalized to the corresponding bulk composition of its host rock (Fig. 13a). If lawsonite composition was continuously equilibrated with evolving host rock composition, lawsonite would have inherited its elemental signatures entirely from the host rock. In this case, the value of normalization would be close to unity. In contrast, any deviation from unity would be expected with the increasing influence of factors other than the host rock signature. The normalized TE concentrations (referred to as 'Bulk' in the subscript of element concentrations) were then compared with the TE budgets of matrix lawsonite.

In most cases, the average element concentrations of matrix lawsonite are lower than the corresponding bulk concentrations of the host matrix (<1 in Fig. 13a), and matrix lawsonite hosts less than 100% of bulk REE and TEs (Fig. 13b and c). Such matrix grains host variable proportions of bulk Sr, Pb, REE, Th, U, Cr, and V (up to 100%), and low proportions (<30%) of bulk LILE (i.e. Rb, Ba) and high field strength elements (HFSE; Zr, Hf) (Fig. 13b and c). The calculated element budget is consistent with the previous mass balance calculations of lawsonite-bearing HP/LT rocks (Spandler *et al.*, 2003; Martin *et al.*, 2014; Hara *et al.*, 2018; Whitney *et al.*, 2020). LILE- (e.g. phengite, glaucophane), L-MREE- (e.g. apatite, titanite, epidote), and U/Th-bearing phases (e.g. apatite,

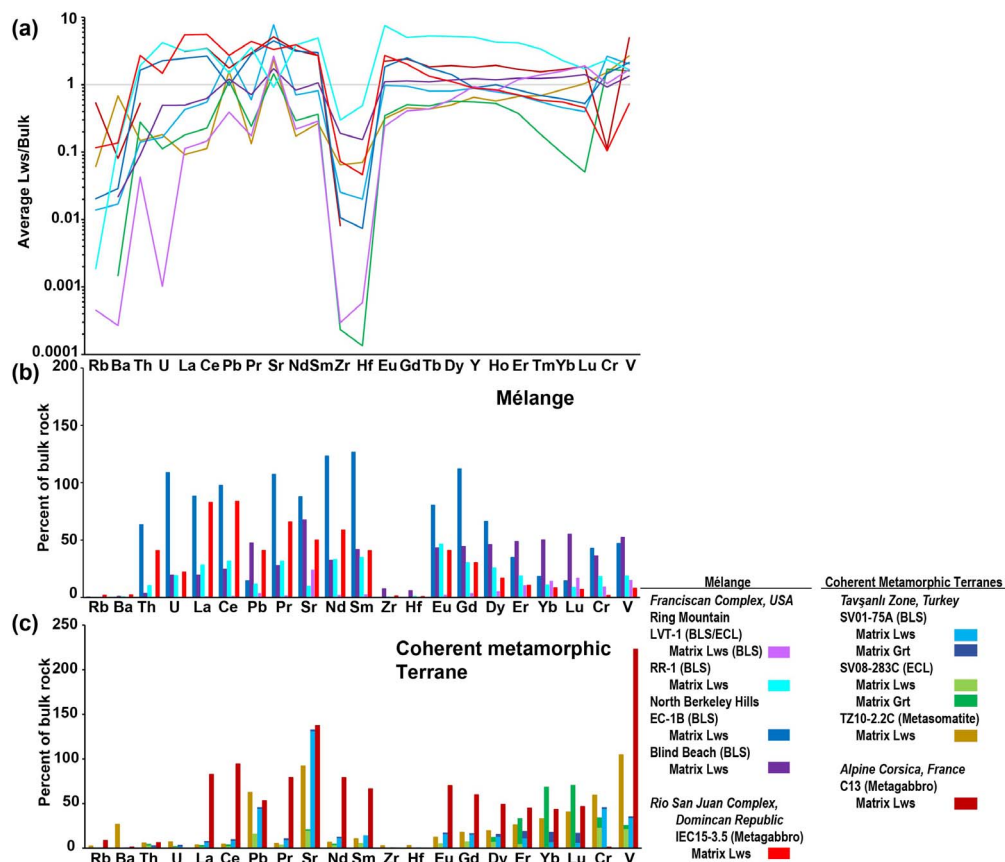


Fig. 13. (a) Bulk-rock-normalized average element concentrations of matrix lawsonite. (b, c) Element budgets of matrix lawsonite and matrix garnet.

titanite) occur in the matrix of most samples analyzed (Table 1). Therefore, the concurrent growth of these minerals can explain the low abundances of these elements in lawsonite.

In contrast, some matrix lawsonite grains apparently host more than 100% of bulk REE and TEs (e.g. Sr, V) and the average concentrations of these elements are higher than the bulk-rock concentrations (>1) (e.g. Sivrihisar blueschist SV01-75A; Fig. 13). Such overestimated element budgets have also been reported (Spandler *et al.*, 2003; Martin *et al.*, 2014) and interpreted as the influence of μm -sized inclusions in lawsonite. Possible Sr-, V- and/or REE-bearing inclusions in lawsonite are epidote, apatite, and titanite (Table 1).

In contrast, matrix lawsonite in the Rio San Juan blueschist-facies metagabbro (IEC15-3.5) and the Ring Mt blueschist (RR-1) accounts for less than 90% of bulk L-MREE and 40% of bulk Th and U, although the average concentrations of these elements are higher than bulk-rock concentrations (Fig. 13a and b). This can be explained by (1) the absence or low modal abundance of minerals that compete for these elements and/or (2) low lawsonite mode. Lawsonite in IEC15-3.5 best exemplifies the former case, given the absence of U, Th, L-MREE-bearing competitors except for minor epidote (Table 1) and texturally late calcite (Fig. 3j). However, lawsonite in RR-1 occurs as a major matrix

phase and coexists with other U, Th, L-MREE-bearing minerals (e.g. titanite, apatite; Table 1). This requires alternative explanations, such as the following: (1) lawsonite mode was underestimated; (2) some domains of matrix lawsonite equilibrated with a bulk-rock composition that was richer in L-MREE, U, and Th than the measured bulk composition; and/or (3) the measured lawsonite composition may not represent the average composition.

Matrix garnet from the Sivrihisar blueschist (SV01-75A) and eclogite (SV08-283C) is a major host mineral of HREE (Fig. 11b), explaining the HREE depletion of coexisting lawsonite (Fig. 13c). Although the bulk-rock-normalized average element concentrations and element budget of matrix lawsonite are useful to assess the influence of factors other than host rock signatures (e.g. element fractionation by coexisting minerals), they might de-emphasize the records of zoning by averaging the compositional variations of individual minerals. Therefore, the following sections investigate the zoning records of individual lawsonite grains.

Compositional variations in lawsonite

The possible mechanism of Ti, Fe, and Cr zoning in lawsonite is discussed in relation to variations in other TEs (e.g. REE, Pb, Sr).

Crystallographic control of Ti/REE in lawsonite: hourglass sector zoning

Sector zoning may reflect coeval differences in element selectivity on different growth planes in response to reaction overstepping and fast growth rate (Watson & Liang, 1995; Stowell *et al.*, 2011). Such differences can be retained in the crystal as a result of slow intracrystalline diffusion rates (Wass, 1973; Watson & Liang, 1995). Ti-hourglass sector zoning was observed in lawsonite from blueschist and vein in the Ring Mt blueschist/eclogite (LVT-1), from the Sivrihisar blueschist (SV01-75A), and from the partially replaced area of omphacite in Corsica Lws-Omp metagabbro (C13).

In all these samples, Ti-rich Zone 2 displays REE patterns similar to Zone 1, but has noticeably lower REE contents than Zone 1 (Fig. 14a–d), possibly as a result of the rapid growth of Zones 1 and 2 (Wass, 1973; Ueno, 1999; Vitale Brovarone *et al.*, 2014). In general, Ti-poor Zone 3 has similar to slightly different REE patterns as compared with the associated Zones 1 and 2, and the overall REE abundances are higher than Zone 2 (Fig. 14a, b, e and f). Zone 3 might have readily incorporated REE relative to the other Zones as suggested by Ueno (1999), who documented REE enrichments in the {100} sector, which grows along [100] and $[i00]$ based on his nomenclature. However, Zone 3 in Corsica lawsonite (C13) shows a selective enrichment in HREE relative to the associated Zone 2 (Fig. 14c). Metasomatic lawsonite documented in the previous study of Corsica metasomatite is characterized by continuously decreasing REE contents and a consistent REE pattern across Zones 1–3, possibly owing to the fast growth of Zones 1–3 (Vitale Brovarone *et al.*, 2014). From this perspective, Zone 3 in lawsonite can have variable REE abundances with a similar to significantly different REE pattern compared with the associated Zones 1 and 2. This suggests that factors other than crystallographic control (e.g. element fractionation between minerals, fluid–rock interactions, fast crystal growth) simultaneously influenced REE partitioning in Zone 3. For instance, the selective HREE enrichment in Zone 3 of Corsica lawsonite may either signal the infiltration of HREE-rich fluids or L-MREE fractionation with coexisting minerals (Fig. 14c). In the case of Sivrihisar lawsonite, Zone 4 shows significant variations in HREE contents compared with Zone 1–3 (Fig. 14g), possibly indicating the intermittent influence of HREE-rich fluids and/or interactions with HREE-rich garnet (Fig. 11b).

All domains, including Zone 4, have TE patterns characterized by strong depletions in HFSE and LILE (Fig. 14b, d, and f). Pb and Sr anomalies are variable within the zones (Supplementary Table S1), but their variations among the zones are largely controlled by the abundance of LREE that have a similar compatibility with Pb and Sr (e.g. Ce, Pr). Pb and Sr anomalies tend to be higher in REE-poor Zone 2 relative to REE-richer Zones 1 and 3 (e.g. Sivrihisar blueschist SV01-75A, vein in the blueschist layers of LVT-1; Fig. 14g) unless the changes in Sr and Pb concentrations across Zones 1–3

counterbalance the effect of variations in LREE contents (e.g. Zone 3 of vein-lawsonite with a higher Sr anomaly in the eclogite layers of LVT-1; Fig. 14g and h). In addition, U–Th fractionation ($0 \leq (U/Th)_{PM} \leq 54.9$) are significantly variable within and among the zones (Fig. 14b, d, and f) mainly due to the low U and Th contents ($\leq 2 \mu\text{g/g}$; Supplementary Table S1). Such heterogeneous lawsonite composition corroborates the simultaneous operation of several factors (e.g. element-fractionations between minerals, fluid–rock interaction) in addition to crystallographic control. Despite the influence of other factors, our compositional analyses highlight the importance of crystallographic control on the Ti and REE contents of lawsonite.

Evidence for mineral–fluid interactions and element fractionation between minerals Fe/REE zoning

Lawsonite grains that exhibit core-to-rim zoning in Fe but not hourglass zoning in Ti in some cases show covariations between Fe and REE. Some matrix grains in the N Berkeley Hills blueschist (EC-1B) are characterized by a significant MREE depletion compared with LREE and HREE depletions at Fe-rich rims, which are illustrated as high $(La/Dy)_N$ and low $(Dy/Yb)_N$ (Fig. 15a and b). This indicates the concurrent growth of lawsonite with MREE-rich apatite (Fig. 11d) and/or titanite, both of which occur in the matrix of sample EC-1B (Table 1). In contrast, vein-lawsonite records high $(Dy/Yb)_N$ and $(La/Yb)_N$ at the Fe-depleted rims, reflecting an HREE depletion (Fig. 15b and c). The formation of HREE-rich pumpellyite that replaced vein-lawsonite (Fig. 3f) could explain the fractionation of Fe and HREE.

Matrix grains in the Corsica Lws-Omp metagabbro (C13) are nearly constant in $(La/Dy)_N$, $(La/Yb)_N$, and $(Dy/Yb)_N$ regardless of Fe content (Fig. 15a–c; Table 3). Together with the constant REE patterns, Corsica matrix grains record a core-to-rim decrease in Fe and REE concentrations (Fig. 9b). This suggests REE and Fe depletions during continuous growth of lawsonite grains.

The Fe content of lawsonite in calcite-rich veins from the Ring Mt blueschist (RR-1) increases toward the rims and is positively correlated with $(La/Dy)_N$ and $(La/Yb)_N$ (Fig. 15a and c). One possibility to explain this relative LREE enrichment is that lawsonite in the matrix was influenced by LREE fractionation during its earlier growth, later incorporated into the vein, and subsequently influenced by LREE-rich vein-forming fluids. However, vein-associated lawsonite shows changes in REE patterns that become comparable with matrix lawsonite toward the Fe-rich rims, with REE abundances approaching the bulk concentrations of the matrix (~ 1 ; Fig. 15d). Therefore, vein-lawsonite might have co-crystallized with LREE-hosting aragonite during earlier growth, and gradually became equilibrated with the matrix while vein-aragonite was converted to calcite during later exhumation.

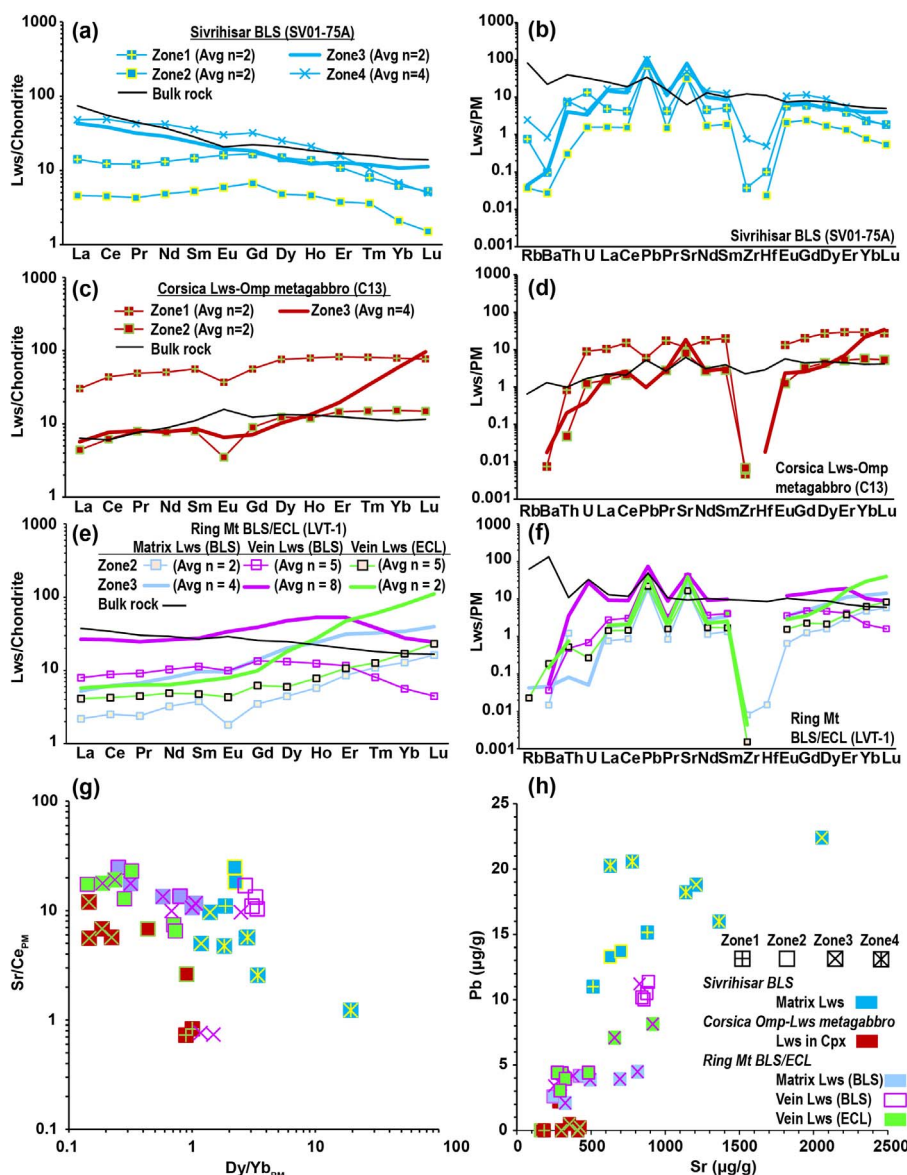


Fig. 14. (a–h) Composition of lawsonite with Ti-hourglass zoning. REE (a, c, e) and TE patterns (b, d, f) of lawsonite from Sivrihisar blueschist (SV01-75A) (a, b), Corsica Lws-Omp metagabbro (C13) (c, d), and Ring Mt blueschist/eclogite (LVT-1) (e, f). (g) $(\text{Sr}/\text{Ce})_{\text{PM}}$ vs. $(\text{Dy}/\text{Yb})_{\text{PM}}$. (h) Pb vs. Sr.

Matrix grains in the Sivrihisar blueschist/eclogite (SV01-50A) have high $(\text{La}/\text{Yb})_{\text{N}}$ and $(\text{Dy}/\text{Yb})_{\text{N}}$ in the Fe-poor core (Fig. 15b and c), which corresponds to low Sr/Pb (17–20) (Fig. 10c; Table 3). This low Sr/Pb is within the range of lawsonite from quartz-bearing metasediment (Fornash *et al.*, 2019; Whitney *et al.*, 2020), indicating possible interactions with siliciclastic metasediment (also high in La/Yb and Dy/Yb) during the growth of core. The subsequent decreases in $(\text{La}/\text{Yb})_{\text{N}}$ and $(\text{Dy}/\text{Yb})_{\text{N}}$ (Fig. 15b and c) may reflect the decreasing influence of siliciclastic metasediment as suggested by a simultaneous increase in Sr/Pb (29–50; Table 3).

The $(\text{La}/\text{Dy})_{\text{N}}$ of some matrix grains in the Pinchi Lake retrogressed eclogite (BLR4) initially increases and then decreases with $(\text{La}/\text{Yb})_{\text{N}}$ toward the Fe-rich rims (Fig. 15a and c). The initial LREE and Fe enrichments might indicate the breakdown of LREE-hosting, Fe-rich epidote (absent in the matrix; Table 1). However, the

Pinchi Lake eclogites evolved within the stability field of lawsonite from prograde to peak conditions (Ghent *et al.*, 1993), thereby requiring a different explanation. Interactions with siliciclastic metasediment can account for the LREE enrichment and a concomitant Sr/Pb decrease (16–18; Table 3), which falls within the range of lawsonite in quartz-rich metasediment (Fornash *et al.*, 2019). The effect of Sr/Pb-fractionation is likely negligible given the absence of other Sr- and Pb-hosting minerals in the matrix and garnet. LREE depletion and Sr/Pb increase (32–101; Table 3) might stem from the decreasing contribution from quartz-rich (meta)sediment and/or the infiltration of LREE-depleted fluids with high Sr/Pb.

Oscillatory zoning

Lawsonite in HP/LT rocks, metasomatite, and eclogite-facies veins in some cases preserves oscillatory zoning, which has been suggested to develop as a result of

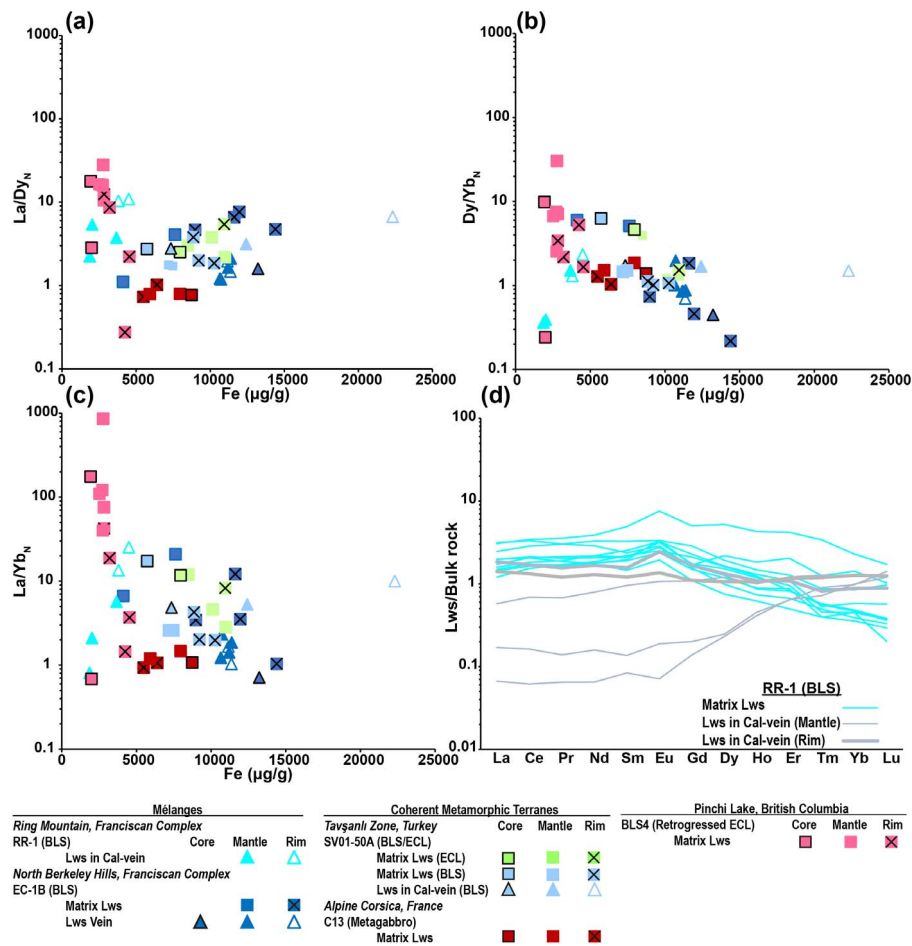


Fig. 15. (a, b, c) Lawsonite with Fe-concentric zoning, showing variations in $(\text{La}/\text{Dy})_N$ (a), $(\text{Dy}/\text{Yb})_N$ (b), and $(\text{La}/\text{Yb})_N$ (c). (d) Bulk-rock normalized REE patterns of lawsonite in calcite vein from Ring Mt blueschist (RR-1).

fluctuating metamorphic P - T - $f\text{O}_2$, and/or of intermittent element supply via fluid-infiltration or mineral breakdown reactions (Sherlock & Okay, 1999; Vitale Brovarone *et al.*, 2014; Fornash *et al.*, 2019; Fornash & Whitney, 2020). In the Ring Mt blueschist RR-1, matrix lawsonite locally shows Fe-oscillatory zoning that negatively correlates with Ti-oscillatory zoning (Fig. 8f). The fluctuating Fe content also covaries with $(\text{La}/\text{Dy})_N$ and $(\text{La}/\text{Yb})_N$ (Fig. 16a) owing to repeated changes in REE contents, of which M-HREE are positively correlated with Fe (Fig. 16a). Mn and Cr contents also covary with HREE content, and they show a positive and negative correlation, respectively (Fig. 16a).

One possible explanation for these element covariations is the destabilization of garnet (absent in the sample; Table 1) during the growth of lawsonite. Garnet that is oscillatory zoned in Mn occurs in zoisite-bearing interlayered eclogite/blueschist and amphibolite in the same region (Viète *et al.*, 2018; Cruz-Urbe *et al.*, 2021). This oscillatory pattern has been interpreted to reflect a periodic dissolution-growth cycle of garnet that developed in response to intermittent pulses of fluid flow (Viète *et al.*, 2018; Cruz-Urbe *et al.*, 2021). Upon the cyclic dissolution to complete breakdown of garnet, Fe, Mn and M-HREE might have been provided intermittently as lawsonite

grew along a retrograde P - T path (Fig. 2). Between the pulses of fluids, matrix lawsonite likely hosted Cr instead of Fe, Mn, and M-HREE (Fig. 16a).

Cr-oscillatory zoning was observed in matrix grains from the Sivrihisar blueschist SV01-75A, which also show Ti-hourglass zoning (Fig. 8e). In Zones 3 and 4, high Cr content coincides with high Fe and low V (Fig. 16b). A positive Cr^{3+} - Fe^{3+} correlation in oscillatory zoning has been documented in Sivrihisar lawsonite and suggested to reflect the fluctuating $f\text{O}_2$ of infiltrating fluids (Fornash *et al.*, 2019). Given that Cr^{3+} - Al^{3+} substitution in lawsonite is facilitated at oxidizing condition (Sherlock & Okay, 1999), high Cr^{3+} and Fe^{3+} and low V may indicate the relatively oxidizing condition of infiltrating fluids. Although Cr is assumed to be relatively immobile in fluids, minerals in HP metasomatic environments (e.g. eclogite-facies veins, margins of HP/LT blocks) preserve evidence supporting enhanced Cr mobility (e.g. Cr-rich domains in oscillatory zoned garnet, lawsonite) (Spanner *et al.*, 2011; Angiboust *et al.*, 2014; Vitale Brovarone *et al.*, 2014; Fornash & Whitney, 2020). In addition, the solubility of Cr can be significantly enhanced in Cl-rich, saline fluids (Huang *et al.*, 2019), the compositions of which resemble serpentinite-derived fluids (Scambelluri *et al.*, 2004; Kendrick *et al.*, 2011; Huang *et al.*, 2017).

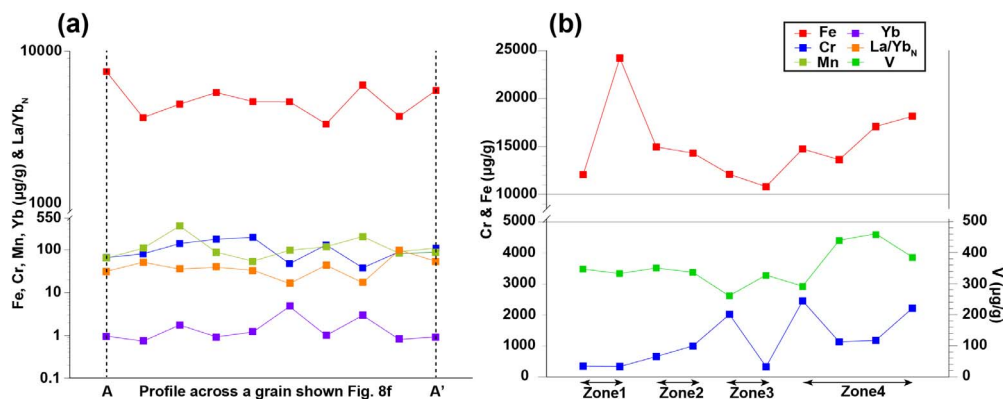


Fig. 16. (a) Matrix lawsonite from Ring Mt blueschist (RR-1), showing oscillatory variations in Fe, Yb, Cr, Mn, and (La/Yb)_N. (b) Matrix lawsonite from Sivrihisar blueschist (SV01-75A), showing oscillatory variations in Cr, Fe, and V across Zones 3–4.

Serpentinite-sourced fluids can also explain the high bulk Cr and Ni contents of the Sivrihisar blueschist (Table 2).

Variations of Sr/Pb and Cr/V

The Sr/Pb ratio in lawsonite varies as a function of rock type; lawsonite in metabasite tends to have higher Sr/Pb than lawsonite in quartz-rich metasediment regardless of the occurrence and variable modes of Sr- and/or Pb-bearing phases (e.g. titanite, apatite, epidote) (Martin et al., 2011; Fornash et al., 2019; Whitney et al., 2020). For instance, matrix grains from a Guatemala eclogite that underwent metasomatic alteration by metasediment-sourced fluids (Hara et al., 2018) record Sr/Pb between those from less-altered eclogite and metachert in the same region (Fig. 10a). Possible Pb-bearing titanite only occurs in the metasomatized eclogite and the metachert (Hara et al., 2018). Our compilation of lawsonite composition suggests that lawsonite in calcschist has Sr/Pb mostly higher than lawsonite in quartz-rich metasediment but within the range of lawsonite in metabasite (Supplementary Table S1).

As with Sr/Pb, the Cr concentration of lawsonite correlates with rock type. In general, lawsonite in quartz-rich metasediment contains lower Cr concentrations than lawsonite in metabasite despite the lower abundance of Cr-bearing competitors (e.g. garnet) in metasediment (Supplementary Table S1). Lawsonite that interacted with serpentinite/metagabbro is enriched in Cr and Ni contents (Angiboust et al., 2014; Vitale Brovarone et al., 2014; Fornash & Whitney, 2020). The V contents of lawsonite in quartz-rich metasediment, calcschist, and metabasite overlap each other (Supplementary Table S1), and they tend to correlate with the Cr contents positively (Fig. 10e).

Consequently, Sr/Pb and Cr/V ratios of lawsonite are useful to evaluate the control of host rock signatures. Sr/Pb_{Bulk} and Cr/V_{Bulk} were additionally calculated relative to bulk-rock ratios of matrix even for vein-associated lawsonite. Significant deviations from the bulk-rock ratios (e.g. Cr/V_{Bulk} > 1) would provide additional information on (1) the timing of specific element

exchanges (e.g. Cr-enrichment), (2) element fractionation with coexisting minerals, and/or (3) changes in mineral modes (excluding lawsonite). The element concentrations of lawsonite that vary at a consistent rate (e.g. constant Sr/Pb, Sr/Pb_{Bulk}) would indicate modal changes in lawsonite because element ratios of lawsonite are not affected by lawsonite mode.

Lawsonite grains in the Franciscan metabasalts show noticeable differences in Sr/Pb_{Bulk} and/or Cr/V_{Bulk} depending on their textural locations (vein vs. host matrix). In the N Berkeley Hills blueschist (EC-1B) and the Ring Mt blueschist/eclogite (LVT-1), vein-lawsonite is higher in Cr/V_{Bulk} (>1) than matrix lawsonite (≤1), with a lower-to-similar range of Sr/Pb_{Bulk} (Fig. 17a). The higher Cr/V_{Bulk} of vein-lawsonite stems from significantly higher Cr_{Bulk} compared with matrix lawsonite (Fig. 17b). In conjunction with the high Cr_{Bulk} of vein-lawsonite, the vein area of sample EC-1B has bulk Cr content higher than the matrix (Table 2), suggesting the Cr-rich nature of vein-forming fluids. The vein-lawsonite in sample LVT-1 displays a gradual Cr/V_{Bulk} decrease from Zone 2 to Zone 3 due to a significant Cr_{Bulk} decrease (blueschist layer) or increasing V_{Bulk} (eclogite layer) (Fig. 17b). This contrasts with the matrix lawsonite in blueschist layers that shows less Cr_{Bulk} and V_{Bulk} variations (Fig. 17b). These variations in Zones 2 and 3 indicate that Cr- and V-partitioning was not crystallographically controlled. Possible mechanisms for the variable partitioning include compositional changes in infiltrating fluids (e.g. V-poor to -rich vein-forming fluids in eclogite layers) and/or changes in mineral modes and assemblages. The vein-lawsonite with high Cr/V_{Bulk} has Sr/Pb corresponding to the range of lawsonite in metabasite (>30; Fig. 10b–d) and its Sr/Pb is higher than the bulk Sr/Pb of the host matrix (Sr/Pb_{Bulk} > 1; Fig. 17a). The likely source of Sr/Pb- and Cr-rich vein-forming fluids are (meta)ultramafic rocks.

On the other hand, lawsonite grains in the calcite-rich vein of the Ring Mt blueschist (RR-1) have Sr/Pb_{Bulk} lower than that of matrix lawsonite, with Sr/Pb comparable with lawsonite in calcschist-to-metabasite (Figs 10c and 17a). The relatively low Sr/Pb_{Bulk} of vein-lawsonite is attributable to Sr/Pb-fractionation with Sr/Pb-hosting

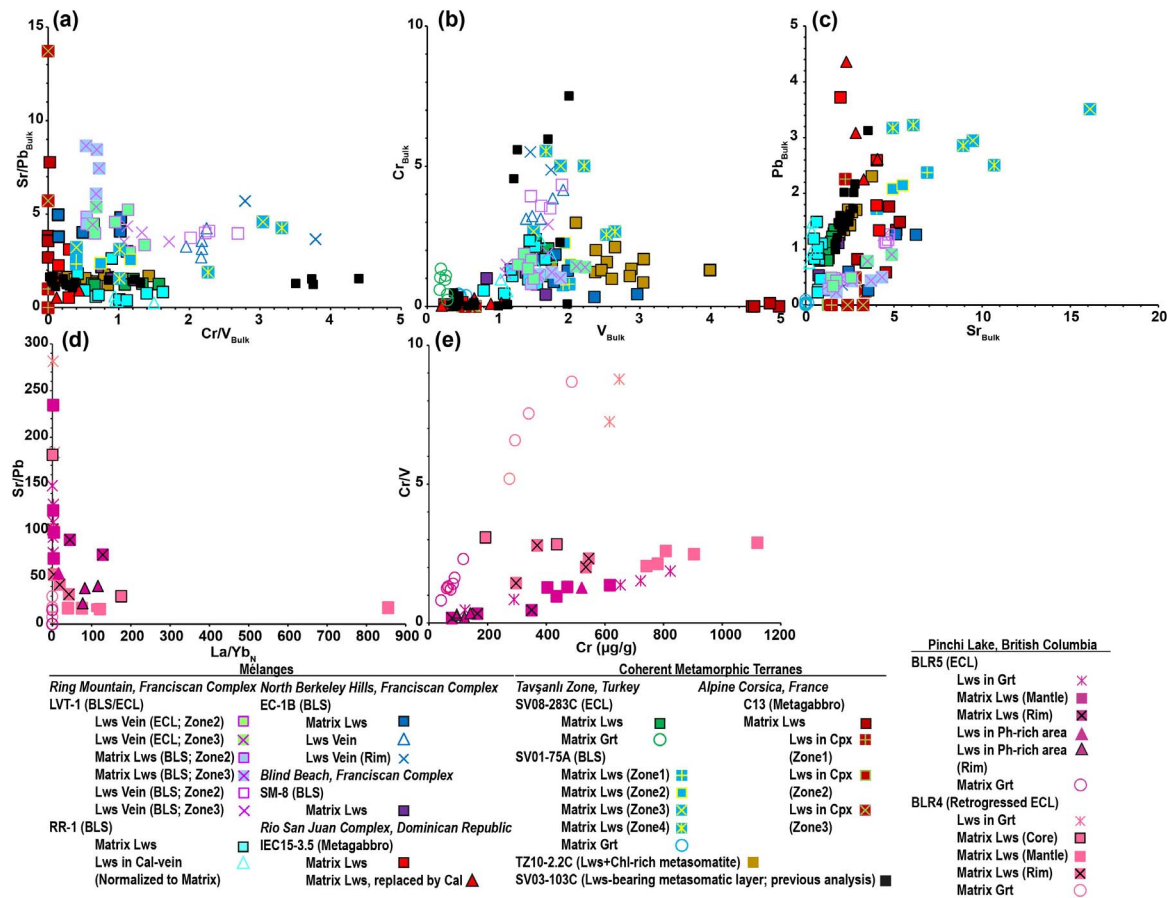


Fig. 17. (a) $\text{Sr/Pb}_{\text{Bulk}}$ vs. $\text{Cr/V}_{\text{Bulk}}$. (b) Cr_{Bulk} vs. V_{Bulk} . (c) Pb_{Bulk} vs. Sr_{Bulk} . (d) Sr/Pb vs. $(\text{La/Yb})_{\text{N}}$ (d) and Cr/V vs. Cr (e) in lawsonite from the Pinchi Lake eclogites (BLR4, 5).

calcite (Table 1). It is also likely that the vein-forming fluids were Sr/Pb-poorer than the host matrix; bulk-rock measurements support this interpretation (vein: 131; host matrix: 151 Sr/Pb). Together with the lower Sr/Pb, the vein has higher bulk LREE and lower Ni and Cr contents than the host matrix (Figs 5 and 6). These compositional characteristics suggest that vein-forming fluids were likely derived from (meta)sediment, such as calcschist.

Matrix grains from the Sivrihisar eclogite (SV08-283C) and the Tavşanlı metasomatite (TZ10-2.2C) are characterized by variable $\text{Cr/V}_{\text{Bulk}}$ and consistent $\text{Sr/Pb}_{\text{Bulk}}$ (Fig. 17a). Only the grains from the Sivrihisar blueschist (SV01-75A) show changes in both $\text{Sr/Pb}_{\text{Bulk}}$ and $\text{Cr/V}_{\text{Bulk}}$ (Fig. 17a). The $\text{Cr/V}_{\text{Bulk}}$ variations could arise from interactions with Cr/V-rich or -poor fluids and/or Cr/V-fractionations with coexisting minerals. Matrix garnet in the Sivrihisar blueschist and eclogite (SV01-75A, SV08-283C) is one of major host minerals of Cr (Fig. 13c), so Cr-fractionation with garnet cannot explain high Cr_{Bulk} and $\text{Cr/V}_{\text{Bulk}}$ (>1) in matrix lawsonite. For instance, Zones 3 and 4 of matrix lawsonite in the blueschist records $\text{Cr/V}_{\text{Bulk}}$ significantly higher than that of associated Zones 1 and 2 (0.4–3.31 vs. 0.40–1.17) and coexisting matrix garnet (0.51–1.05) but comparable with the range of lawsonite in metasomatic layers that developed at the

margins of eclogite pods in the same region (Fig. 17a). Fornash & Whitney *et al.* (2020) proposed that these layers obtained Ni and Cr by the influx of external fluids sourced from (meta)ultramafic rocks. The Sivrihisar blueschist might have been influenced by similar fluids, as evidenced by the high Cr_{Bulk} and $\text{Cr/V}_{\text{Bulk}}$ in matrix lawsonite comparable with metasomatic lawsonite (Fig. 17a and b), Cr-oscillatory zoning in matrix lawsonite (Fig. 8e), and the bulk Ni and Cr enrichment of matrix blueschist (Table 2). From Zones 1 and 2 to Zones 3 and 4, $\text{Sr/Pb}_{\text{Bulk}}$ increases with $\text{Cr/V}_{\text{Bulk}}$ (Fig. 17a) and matrix lawsonite has Sr/Pb corresponding to lawsonite in metabasite (>30) (Fig. 10a). These features further support the influence of (meta)ultramafic rocks. Relative to the Sivrihisar metabasalts, the Tavşanlı metasomatite is higher in bulk V content, but lower in bulk Cr content (Fig. 5). Matrix lawsonite in this metasomatite significantly hosts bulk V (Fig. 13c) shows wider variations in V_{Bulk} than Cr_{Bulk} (Fig. 17b) and has Sr/Pb within the range of metabasite (Fig. 10a). This indicates possible interactions with V-rich (thus Cr/V-poor) fluids potentially sourced from metabasite during metasomatic alteration.

Lawsonite in metagabbro (C13, IEC15–3.5) has the lowest range of $\text{Cr/V}_{\text{Bulk}}$ and the widest range of $\text{Sr/Pb}_{\text{Bulk}}$ among the analyzed grains (Fig. 17a). This is mainly

because of the extremely low Cr_{Bulk} and significant variations in Pb_{Bulk} and Sr_{Bulk} (Fig. 17b and c). Amphibole in IEC15-3.5 and clinopyroxene in C13 contain up to 727 and 278 Cr $\mu\text{g/g}$, respectively (Supplementary Table S1), and their modal abundances are significantly high in the host matrix (Table 1). These matrix minerals, thus, might have fractionated Cr from coexisting lawsonite (Spandler *et al.*, 2011; Angiboust *et al.*, 2014). In addition, the low bulk Cr content in C13 could contribute to the low $\text{Cr/V}_{\text{Bulk}}$ (Fig. 5; Table 2). Element fractionation with Sr- and/or Pb-bearing phases (e.g. calcite, epidote; Table 1) likely induced $\text{Sr/Pb}_{\text{Bulk}}$ variations, as evidenced by the lower $\text{Sr/Pb}_{\text{Bulk}}$ of lawsonite partially replaced by calcite (0.53–1.55; Fig. 3j) compared with lawsonite with no textural replacement (0.53–3.11; e.g. IEC15-3.5) (Fig. 17a). This element fractionation might have varied in response to modal changes in Sr- and/or Pb-bearing minerals, in which case, caused additional $\text{Sr/Pb}_{\text{Bulk}}$ variations. Lawsonite in the partially replaced domains of omphacite (C13) records a higher range of $\text{Sr/Pb}_{\text{Bulk}}$ than matrix lawsonite, possibly indicating the Pb-poor (thus Sr/Pb-rich) nature of infiltrating fluids (Fig. 17a and c).

In the Pinchi Lake eclogites, matrix lawsonite in the more retrogressed sample (BLR4) shows a clear correlation between Sr/Pb and LREE contents; LREE contents initially increase and subsequently decrease, whereas Sr/Pb decreases and then increases toward the Fe-rich rims (Fig. 17d). We interpreted that the initial changes in LREE and Sr/Pb likely resulted from interactions with quartz-bearing (meta)sediment. Similar (meta)sedimentary signatures are recorded at the rims of lawsonite grains in the matrix and the phengite-rich area of less retrogressed sample (BLR5) (Fig. 17d). These grains in BLR5 display noticeable Cr depletion and decreasing Cr/V, which are in accordance with the (meta)sedimentary signatures (Fig. 17e). In contrast to the BLR5 grains, initial LREE enrichment is accompanied by Cr enrichment in matrix BLR4 grains (Fig. 17d and e), likely owing to Cr variations induced by lawsonite modal changes given the consistent Cr/V (Fig. 17e). The subsequent changes in LREE and Sr/Pb are only observed in matrix lawsonite from the more retrogressed BLR4 (Fig. 17d). Lawsonite inclusions in garnet are significantly depleted in L-MREE (Fig. 9c and d) and have Cr/V within the range of host garnet (Fig. 17e). In particular, the BLR4 inclusion records noticeably high Cr/V (Fig. 17e), with high Sr/Pb corresponding to lawsonite in metabasite (>30 ; Fig. 17d). These features might have originated from Cr/V and Sr/Pb-rich fluids released during serpentinization, which was interpreted to be contemporaneous with blueschist-facies metamorphism of metabasaltic units (Paterson, 1977).

IMPLICATIONS AND CONCLUSION

This study shows that the composition and zoning of lawsonite is a useful indicator of fluid–rock interaction during subduction metamorphism, although care must

be taken to understand the mechanisms controlling lawsonite composition and its variation. Vein-lawsonite from Franciscan metabasites records low Sr/Pb or high Cr/V, reflecting various sources of vein-forming fluids ((meta)sediment, serpentinite/metagabbro). Lawsonite in metasomatized domains (e.g. replaced domains of omphacite in Corsica metagabbro) differs in Sr/Pb or Cr/V from matrix lawsonite, suggesting the compositional characteristics of infiltrating fluids. The bulk LILE, LREE, and/or Th enrichments of most metabasites (except from Corsica) and the Tavşanlı metasomatite indicate possible interactions with (meta)sediment, although the composition of matrix lawsonite records no such signatures. Instead, matrix lawsonite shows compositional variations in REE, TES, and element ratios (Sr/Pb, Cr/V) that suggest (1) element fractionation between minerals, (2) the influence of crystallographic control (i.e. in Ti-sector zoning), and/or (3) intermittent interactions with fluids sourced from (meta)mafic-to-ultramafic rocks. Only a few grains preserve possible sedimentary signatures in their cores (e.g. low Sr/Pb). This inconsistency might be related to the timing of interaction with (meta)sediments that likely occurred prior to or at the early stages of lawsonite growth. In this case, matrix lawsonite inherited (meta)sediment-influenced signatures from its host rock, and its compositional variations only record the changes that occurred during its growth. This hypothesis could be tested if advances are made in lawsonite geochronology.

Our study shows that lawsonite is an effective recorder of elemental redistributions driven by various stages of mineral reactions and fluid–rock interactions in subduction systems. To track element transfer history during subduction metamorphism, it is important to document lawsonite and its host rock compositions and to evaluate compositional zoning patterns.

DATA AVAILABILITY STATEMENT

The data are available in online supplementary material.

SUPPLEMENTARY DATA

Supplementary data are available at *Journal of Petrology* online.

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