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### Trace element budgets and (re-)distribution during subduction-zone ultrahigh pressure metamorphism: Evidence from Western Tianshan, China





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#### ABSTRACT

We have conducted an LA-ICP-MS *in situ* trace element study of garnet, epidote group minerals, phengitic muscovite and paragonite in rocks of basaltic and sedimentary protolith from an ultrahigh pressure metamorphic belt along Western Tianshan, China. The data are used to evaluate the capacity of these minerals for hosting incompatible elements in response to subduction-zone metamorphism (SZM). The results confirm existing studies in that the presence and stability of these minerals largely control the geochemical behaviors of elements during SZM.

We found that redistribution of rare earth elements (REEs), Th and U into newly-formed minerals during progressive SZM precludes the release of these elements from the down-going ocean crust, which contradicts the common perception in models of slab-dehydration and flux-melting. This suggests that additional processes, such as the involvement of supercritical fluids or hydrous melts formed at depth are required to supply these elements to the mantle wedge for arc magmatism. In addition, the ready release of large ion lithophile elements (LILEs) by different minerals, and the high immobility of REEs in rocks of basaltic protolith indicate that the contribution of altered ocean crust after SZM may not be responsible for the correlated Sr–Nd (Hf) isotope systematics observed in oceanic basalts. That is, subducted ocean crust that has gone through SZM cannot be the major source material for ocean island basalts.

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#### 1. Introduction

Subduction zones are the most important tectonic environment on Earth in terms of the extent, magnitude, and diversity of mass exchange between the Earth's exterior and the deep mantle. Subduction zones provide a highly selective physical and chemical filter, conceptualized as the "subduction factory" (*e.g.*, Tatsumi and Kogiso, 2003). The main process in the subduction factory is metamorphism. In fact, it is the subduction-zone metamorphism (SZM) that is considered to trigger subduction-zone magmatism (*e.g.*, Tatsumi, 1986; Peacock, 1990; McCulloch and Gamble, 1991), while residual material passing through the SZM contributes to mantle chemical and isotopic heterogeneities (*e.g.*, Hofmann and White, 1982; Zindler and Hart, 1986; Niu and

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O'Hara, 2003). Therefore, it is crucial to understand the genuine behaviors of chemical elements in response to the SZM in order to obtain an improved understanding of subduction-zone magmatism and chemical geodynamics.

Seafloor materials which have experienced hydrothermal alteration and weathering become highly altered (termed "altered oceanic crust" or AOC; Staudigel et al., 1995; Kelley et al., 2003). The AOC undergoes dehydration in subduction zones, releasing fluids which selectively carry fluid-soluble or mobile elements (e.g., large ion lithophile elements, LILEs) into the overlying mantle wedge, while leaving the fluid-insoluble or immobile elements (e.g., high field strength elements, HFSEs) in the "residual" slab. Melting of the so preferentially enriched mantle wedge is then expected to produce basaltic rocks with the characteristic arc signatures (McCulloch and Gamble, 1991). The residual slab retaining the immobile elements is thus transported into the deep mantle. Because of the often different behaviors of radioactive parent and radiogenic daughter elements, the residual slab with altered parent/daughter (P/D) ratios during the SZM will determine the nature and extent of mantle isotopic variability as reflected in oceanic basalts (*e.g.*, enriched mantle I [EMI], enriched mantle II [EMII], and high " $\mu$ "





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[HIMU;  $\mu = {}^{238}$ U/ ${}^{204}$ Pb]; Zindler and Hart, 1986; Weaver, 1991; Hofmann, 1997). While the above analysis is reasonable, Niu and O'Hara (2003) argued that the Sr–Nd–Hf isotopic systematics of global ocean island basalts (OIB) recorded no prior subduction-zone histories of OIB sources.

The recent recognition of ultrahigh pressure (UHP) hydrous minerals exceeding the amphibole stability field (*e.g.*, lawsonite, phengite; Poli and Schmidt, 1995; Schmidt and Poli, 1998) indicates a geochemically more complex SZM process than previously thought, involving not only discontinuous dehydration but also continuous reactions (Poli and Schmidt, 1995). Furthermore, recent experimental studies and studies on natural rocks have revealed the strong capacity of these UHP minerals for hosting petrologically important trace elements (*e.g.*, Hermann, 2002; Feineman et al., 2007; El Korh et al., 2009). Hence, the retention of these elements may result in a complex element transfer process during the SZM.

With varied geothermal conditions in different subduction zones, the stability of minerals, fluid compositions (aqueous fluids, hydrous melts or supercritical fluids; Kessel et al., 2005; Hermann et al., 2006; Mibe et al., 2011) and reaction kinetics (Becker et al., 2000; Carlson, 2002; John et al., 2004; Beinlich et al., 2010) may all affect elemental behaviors during the SZM (e.g., Spandler et al., 2003). In addition, the quantity of fluids and the nature of fluid flow (e.g., channels/veins, high permeable mélange zones; Zack and John, 2007; John et al., 2008) may also exert important controls on the interactions between fluids and metamorphic rocks during the SZM. The higher the fluid/ wall rock ratio is, the more likely the elements can migrate (Bebout, 2007) and the mobile elements could be homogenized. Therefore, studies of geochemical behaviors of specific elements in response to the SZM and their controls are essential, especially for natural rocks that have experienced the SZM in an open system. Microanalysis techniques (e.g., Laser Ablation-Inductively Coupled Plasma Mass Spectrometry, LA-ICP-MS) allow in situ trace element analysis of minerals with high precision and accuracy, which should improve our understanding of elemental behaviors and the mineralogical controls during the SZM (e.g., El Korh et al., 2009; Schmidt et al., 2009).

In this study, we focus on the distributions of LILEs and rare earth elements (REEs) in different mineral assemblages of blueschistto eclogite-facies metamorphic rocks of basaltic and sedimentary protoliths from the UHP metamorphic belt of Western Tianshan, China, in order to understand trace element behaviors in response to the mineral stability at different metamorphic stages along a specific P-T path. Evidenced by the occurrence of coesite, this metamorphic belt of Western Tianshan in recent studies is considered to have experienced >2.5 GPa subduction-zone metamorphism (Lü et al., 2008, 2009; Lü and Zhang, 2012). Considering that our eclogitic samples are from this UHP metamorphic belt, they can be used to evaluate the geochemical consequences of SZM at depths greater than 75 km, even though coesite has not been found in the samples we studied (Xiao et al., 2012).

## 2. Field geology, petrography and bulk-rock geochemical studies: a review

The Western Tianshan Metamorphic Belt (WTMB) in northwest China has experienced Carboniferous UHP metamorphism (Lü et al., 2008, 2009; Lü and Zhang, 2012). This belt is located along the South Central Tianshan Suture, which defines the south boundary of the Yili-Central Tianshan Plate (Fig. 1; *e.g.*, Zhang et al., 2001; Gao and Klemd, 2003), and is thought to continue westwards into Kazakhstan (Volkova and Budanov, 1999). The WTMB marks the convergent margin associated with successive northward subduction of the South Tianshan Silurian-Devonian-seafloor and the Tarim Plate (*e.g.*, Gao et al., 1999; Gao and Klemd, 2003).

We have studied rock samples of both basaltic and sedimentary protoliths metamorphosed to blueschist and eclogite facies, in which retrograde alteration is common to varying extents (sample locations are given in Fig. 1b and Table DR1). Metamorphic rocks of sedimentary protoliths contain higher modal abundances of quartz and white micas (including phengitic muscovite and paragonite) and display stronger foliations. The eclogite *sensu stricto* (where garnet + omphacite > 70 vol.% of the bulk rock; *e.g.*, TS02-3B, TS02-15A, TS02-17b and TS02-32A) is mainly composed of garnet and omphacite, with varying amounts of glaucophane, white micas, epidote group minerals, quartz, carbonate and rutile. The blueschist contains more glaucophane, while epidote amphibolite contains more retrograde amphibole with (sodic–)calcic component and epidote group minerals. No obvious HP-UHP dehydration/transport veins have been found in these studied rocks. Only some small veins cutting through all the early-formed metamorphic minerals, likely produced after the SZM, are observed on a thin section scale.

On the basis of detailed petrography, especially of the mineral assemblage (Table DR1) and the textural relationships (e.g., Fig. 2), together with the estimated metamorphic *P*–*T* path for coesite-bearing eclogite from WTMB by Lü et al. (2009), a schematic P-T path for rocks of basaltic protolith from WTMB is given in Fig. 3. We divided four stages with different representative mineral assemblages, including prograde blueschist facies, UHP eclogite facies, retrograde eclogite facies and epidote amphibolite facies. The box-shaped lawsonite pseudomorphs (replaced by paragonite and clinozoisite, sometimes with albite; Fig. 2a) are common in garnet porphyroblasts, especially in their core-mantle portions. The replacement of dissolved (clino)zoisite by newly-crystallized omphacite and garnet (e.g., Fig. 2b) corresponds to the reaction of 13 Gln + 6 Czs = 9 Prp + 26 Jd + 12 Di + 19  $Coe/Qz + 16 H_2O$  (see the caption to Fig. 2 for mineral abbreviations), forming amoeboid crystal boundaries (John et al., 2008) and a 'sponge' texture. These textural relationships, together with the occurrence of glaucophane, clinozoisite and omphacite inclusions in garnet, point to the prograde blueschist facies, stage 1 in Fig. 3. The characteristic mineral assemblage at this stage is mainly composed of Lws + Czs +Gln + Omp + Grt + Ph + Pg + Rt. Although no coesite has been found in our samples, considering the occurrence of coesite and the absence of diamond in eclogites from Western Tianshan in others' studies (Lü et al., 2008, 2009), the peak metamorphic condition estimated by Lü et al. (2009; 2.4-2.7 GPa and 470-510 °C) is adapted here as stage 2 (Fig. 3), with the model peak mineral assemblage of Grt + Omp +Lws + Ph + Coe + Rt. The presence of large chloritoid poikiloblasts in sample TS02-01, which points to a pressure of ~2.2-2.4 GPa (Pawley and Holloway, 1993; Lü et al., 2009), is also consistent with the UHP at the peak metamorphic condition. On the other hand, the replacement of lawsonite inclusions by paragonite and clinozoisite in garnet indicates the breakdown of lawsonite, which may happen during retrograde eclogite facies (from stage 2 to stage 3 in Fig. 3), although the timing of this reaction remains debatable. For example, Gao et al. (1999), Gao and Klemd (2003) and Spandler et al. (2003) assumed that it happened during prograde metamorphism from lawsonite blueschist facies to eclogite facies. White micas, especially paragonite, may also be re-produced at this stage, and may replace the rim of garnet (Fig. 2a,f), or include the prior garnet (Fig. 2d), epidote and phengitic muscovite (Fig. 2e). The model mineral assemblage at stage 3 is Grt +Omp + Gln + Czs + Ph + Pg. From stage 3 (retrograde eclogite facies) to stage 4 (epidote amphibolite facies, Fig. 3), earlier glaucophane, omphacite, rutile and garnet were replaced by barroisite (Fig. 2g), retrograde epidote group minerals (Fig. 2c), titanite and chlorite (Fig. 2h). Therefore, the characteristic mineral assemblage becomes Czs + Ttn + Brs/Act + Ph + Chl + Ab.

We have previously used the Nb/Y vs. Zr/Ti diagram (Fig. DR1), an immobile trace-element equivalent of the TAS (total alkali vs. silica) diagram, to divide meta-basaltic rocks into three groups in terms of their large range of Nb/Y ratio within a small range of Zr/Ti (also see Table DR1 for grouping; Xiao et al., 2012). These three groups geochemically resemble OIB (Group 1), MORB contaminated by and/or mixing with continental or arc crust materials (Group 2), and depleted MORB

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