Earth, Planets and Space
a SpringerOpen Journal

LETTER Open Access

Fluid-metapelite interaction in an ultramafic mélange: implications for mass transfer along the slab-mantle interface in subduction zones

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Abstract

The slab-mantle interface in subduction zones is a site of tectonic mixing of crustal and mantle rocks. It is the interface for fluid flow of slab-derived components into the mantle wedge. To assess the fluid-rock interaction along the slab-mantle interface, we studied the bleaching of pelitic schist in an ultramafic mélange. The Nishisonogi metamorphic rocks in Kyushu, Japan, comprise ultramafic mélanges intercalated with epidote-blueschist facies schists. The ultramafic mélange consists of tectonic blocks of various lithologies and a matrix of chlorite-actinolite schist and serpentinite. Along the contact with the mélange matrix, pelitic schist blocks are bleached mainly due to the modal increase of albite and the consumption of carbonaceous material. The bleaching is probably attributed to infiltration of Na-rich external fluid from the mélange matrix. Mass balance analysis indicates losses of C, Rb, K_2O, Ba, Pb, and SiO_2 from the bleached pelitic schist, although Al_2O_3, TiO_2, Sc, Y, Zr, Nb, La, Ce, and Nd remain immobile. This suggests fractionation of large-ion lithophile elements (LILE) and Pb from the high-field-strength elements and rare earth elements during the bleaching. If this ultramafic mélange is analogous to the slab-mantle interface, similar infiltration metasomatism will promote liberation of C, Si, LILE, and Pb from subducting metapelites and enhance metasomatism of the mantle wedge.

Keywords: Element fractionation; Fluid-rock interaction; Metapelite; Subduction zone mélange; Nishisonogi metamorphic rocks

Findings

Introduction

Fluid flow is an agent of large-scale mass transfer in subduction zones and plays a key role in mantle metasomatism and arc magmatism (Peacock 1990; Bebout 1996, 2007a, 2007b, 2013; Manning 2004; Klemd 2013; Spandler and Pirard 2013). At great depths in subduction zones (15 to 100 km), fluids (aqueous fluids in shallower levels and possibly silicate melts in deeper levels; *cf.* Bebout 2007a, b) derived from subducting slabs can transport various components into the mantle wedge, causing hydration and metasomatism of the mantle (Maury et al. 1992; Ryan et al. 1996; Morris and Ryan 2003; Schmidt and Poli 2003; Rüpke et al. 2004). It is generally believed that such fluid flow is essential to generate arc magmas with elevated concentrations of fluid-mobile components such as large-ion lithophile elements (LILE; e.g., Ba, K, Rb, Cs), B, U, and Pb (Morris et al. 1990; Hawkesworth et al. 1993; Tatsumi and Eggins 1995; Davidson 1996; Elliott 2003).

Recent studies of subduction zone mélanges have revealed that significant fluid flow and fluid-rock interaction took place along the slab-mantle interface (Bebout and Barton 1989, 1993, 2002; Sorensen and Grossman 1989, 1993; King et al. 2003, 2006, 2007; Breeding et al. 2004; Marschall et al. 2006; Spandler et al. 2008; Miller et al. 2009; Penniston-Dorland et al. 2010, 2012). The mélanges represent tectonic and metasomatic mixing of sedimentary, mafic, and ultramafic rocks along the interface region at the contact between the subducting slab and the mantle wedge. The mixing may involve devolatilization reactions that would not occur during simple (essentially closed-system) metamorphism of each lithology. Moreover, fluid-rock interactions or exchanges between the sedimentary and ultramafic rocks will produce new hybrid rocks. The

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hybrid rocks have the potential to take up and release components distinct from the protoliths. Thus, fluid-rock interaction in mélanges probably affects the mass transfer of slab-derived components into the wedge mantle.

This paper describes an example of fluid-metapelite interaction in a subduction zone mélange intercalated with epidote-blueschist facies schists. Metapelites are the most important reservoirs of LILE in subducting slabs. Mass balance analysis provides evidence of the liberation of C, Si, LILE, and Pb from metapelites during infiltration metasomatism.

Geological background

The Nishisonogi metamorphic rocks in Kyushu, Japan, represent a Late Cretaceous subduction complex that consists of epidote-blueschist facies schists with minor serpentinite (Figure 1). The schists include mainly pelitic, psammitic, and mafic lithologies. Phengitic muscovite in the pelitic and psammitic schists yields K-Ar and $^{40}Ar/^{39}Ar$ plateau ages of 85 to 60 Ma (Hattori and Shibata 1982; Faure et al. 1988). The matrix mineral assemblages of the pelitic and psammitic schists are carbonaceous material + garnet + chlorite + muscovite + albite + quartz ± epidote and albite + muscovite + quartz ± chlorite, respectively. The psammitic schist locally contains glaucophane + epidote/piemontite + hematite + rutile ± garnet in the matrix. The mafic schist occurs as conformable layers (<100 m in thickness) within the pelitic and psammitic schists. The matrix mineral assemblages of the mafic schist are actinolite/barroisite + epidote + chlorite + albite ± quartz and glaucophane + epidote + chlorite + albite + hematite ± quartz, with the former assemblage being dominant. The serpentinite, which is in tectonic contact with the schists, occurs as pod-like bodies (<150 m in diameter) and ultramafic mélanges (<350 m in thickness). The serpentinite pods typically have metasomatic rinds (Uchida and Muta 1957, 1958; Mori et al. 2007). The matrix mineral assemblage of the serpentinite is antigorite + magnetite ± dolomite ± magnesite ± talc; chrysotile and lizardite are rare. The ultramafic mélanges contain tectonic blocks of metagabbro, mafic schist, pelitic schist, and albitite in a matrix of chlorite-actinolite schist, talc schist, and schistose serpentinite (Nishiyama 1989, 1990). Small blocks (<5 m in diameter) of jadeitite, omphacitite, rodingite, and zoisitite are found locally (Nishiyama 1978; Shigeno et al. 2005, 2012a, b; Mori et al. 2011).

One of the largest ultramafic mélanges in the study area is exposed at Nishikashiyama district in Nagasaki, Japan (Figure 1). A wide variety of rocks such as epidote-barroisite schist, epidote-glaucophane schist, pelitic schist, and albitite appear as discrete tabular blocks less than 10 m in thickness (Figure 2a). These blocks are mutually isolated by a mélange matrix (<5 m in thickness) of chlorite-actinolite schist. The mélange matrix locally contains thin lenses (<2 m in thickness) of serpentinite. In this mélange, all pelitic schist blocks show bleaching along the contact with the mélange matrix (Figure 2b,c). The pelitic schist is normally black but becomes white when within several centimeters of the mélange matrix. The boundary between the unaltered part and bleached part of the pelitic schist is parallel to the contact.

Sampling and analytical methods

To study the bleaching of pelitic schist blocks, samples were collected from two outcrops in Nishikashiyama (Figure 2b, c). From each outcrop, one sample of bleached pelitic schist (BPS), three samples of neighboring unaltered pelitic schist (UPS), and one sample of chlorite-actinolite schist (CAS) were taken. The BPS and UPS

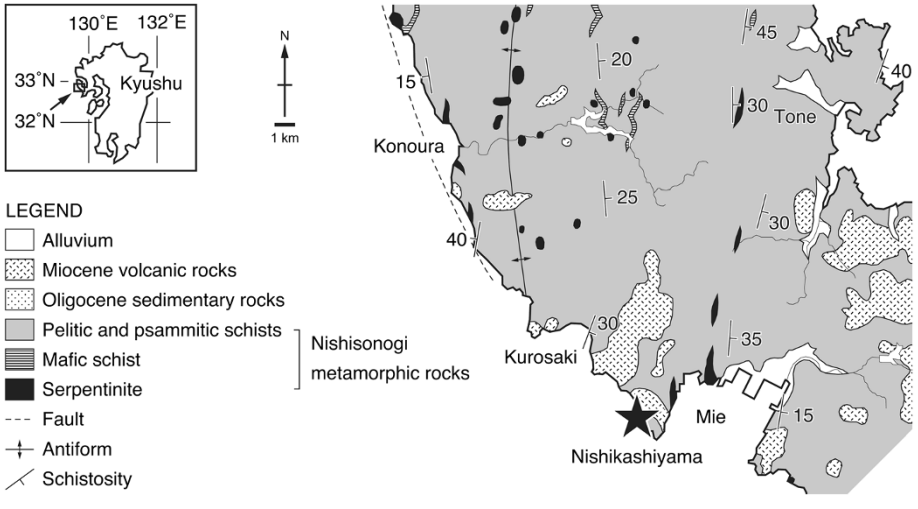

Figure 1 Geologic map of the southern part of Nishisonogi Peninsula, Kyushu, Japan. Star indicates sampling locality.

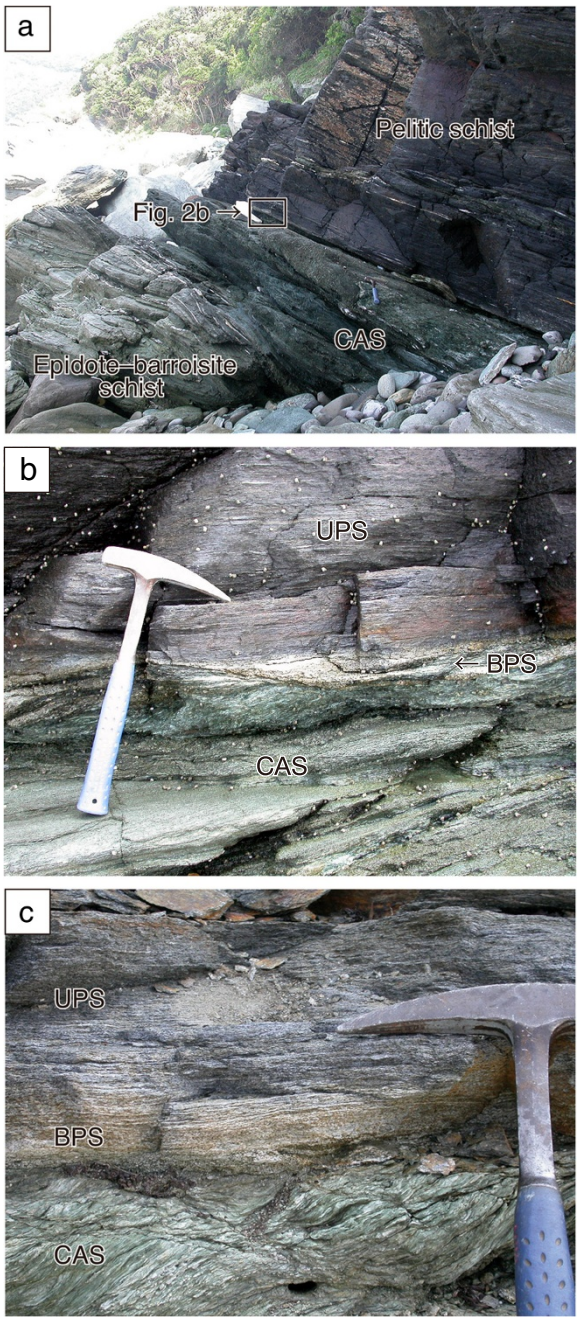

Figure 2 Photographs of the Nishikashiyama mélange. (a) Tectonic blocks of epidote-barroisite schist and pelitic schist in a mélange matrix of chlorite-actinolite schist, outcrop 1. **(b, c)** Bleaching of pelitic schist in outcrops 1 and 2, respectively. BPS, bleached pelitic schist; CAS, chlorite-actinolite schist; UPS, unaltered pelitic schist.

represent different stratigraphic layers of pelitic schist; sampling along a single layer was not possible.

The minerals in the samples were examined with a polarizing microscope under transmitted and reflected light. The modal compositions were measured by counting 2,000 points with a grid interval of 0.1 mm for each sample. Fluid inclusions and opaque minerals were analyzed with a Horiba Jobin Yvon LabRAM HR800 Raman spectrometer (HORIBA Jobin Yvon Inc., Edison, NJ, USA) equipped with a 514-nm argon ion laser housed at Kumamoto University, Japan.

The whole-rock compositions of the samples were determined with a PANalytical MagiX Pro X-ray fluorescence (XRF) spectrometer (PANalytical B.V., Almelo, The Netherlands) and a PerkinElmer 2400 II CHN element analyzer (PerkinElmer, Waltham, MA, USA) housed at the Kitakyushu Museum of Natural History and Human History, Japan. About 300 g of each sample was powdered and homogenized. About 4 g of the sample powder was heated to 950°C for 3 h to measure loss on ignition (LOI). Then, 1 g of the heated powder was prepared as a 1:5 dilution glass bead for XRF. The details of the XRF analysis followed the method of Mori and Mashima (2005). About 0.2 g of the unheated powder was used for CHN (H_2O and C) analysis. Finally, the XRF, LOI, and CHN data were combined as whole-rock compositions.

Petrography

Photomicrographs of the samples are shown in Figure 3. Modal compositions are shown in Figure 4.

Unaltered pelitic schist

The mineral assemblage of UPS is carbonaceous material + chlorite + muscovite + albite + quartz + titanite ± garnet ± calcite ± epidote (Figure 3a). Ore minerals are rare. A minor amount of accessory tourmaline is present in some samples. Carbonaceous material is 5 to 9 vol% in the mode and occurs either as a matrix mineral or as inclusions in albite. It contains graphite and disordered carbons and is considered metamorphosed organic matter. Albite (15 to 37 vol%) typically occurs as porphyroblasts of 1 to 2 mm in diameter. Chlorite (7 to 15 vol%) and muscovite (14 to 33 vol%) define the schistosity of the UPS. Quartz is 19 to 38 vol% in the mode. Titanite is present in all the UPS samples.

Bleached pelitic schist

The mineral assemblage of the BPS is chlorite + muscovite + albite + quartz + titanite ± epidote (Figure 3b). This is essentially the same as the mineral assemblage of the UPS except for the notable absence of carbonaceous material. The BPS lacks carbonaceous material not only in the matrix but also in the albite porphyroblasts. The modal compositions indicate that the BPS is richer in albite (43 and 69 vol% in outcrops 1 and 2, respectively) and is generally poorer in muscovite (18 and 4 vol%) and quartz (29 and 8 vol%) than the UPS. Titanite is present in all the BPS samples.

The albite porphyroblasts have numerous fluid inclusions along clusters of healed microcracks (Figure 3c). This

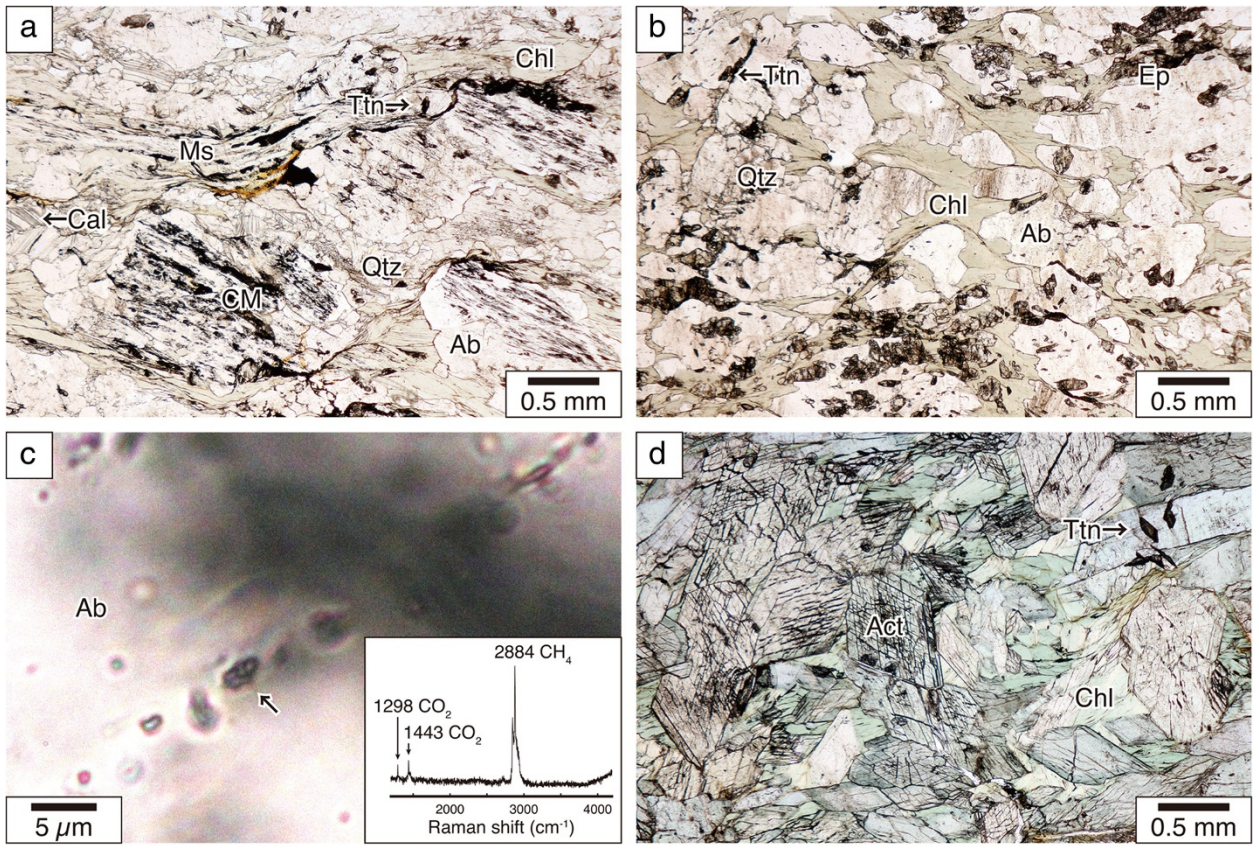

Figure 3 Photomicrographs of selected samples. (a) UPS. **(b)** BPS. **(c)** CH_4-rich (+CO_2) fluid inclusions in BPS and the representative Raman spectrum. **(d)** CAS. Ab, albite; Act, actinolite; Cal, calcite; Chl, chlorite; CM, carbonaceous material; Ep, epidote; Ms, muscovite; Qtz, quartz; Ttn, titanite.

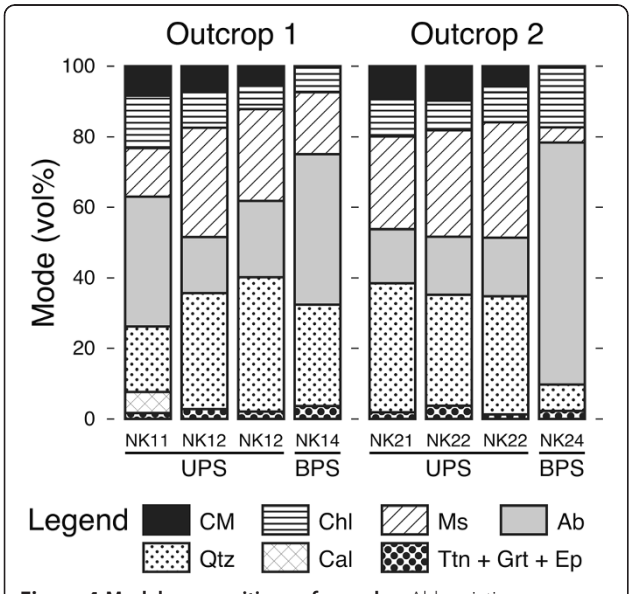

Figure 4 Modal compositions of samples. Abbreviations are shown in Figure 3. The data are listed in Additional file 1: Table S1.

is a feature not found in UPS. The fluid inclusions are typically less than 5 μm in size, have round shapes, and are filled by one phase (H_2O liquid) or two phases (liquid and a small gas bubble) at room temperature. There are two types of the two-phase inclusions: CH_4-rich (±CO_2) inclusions and H_2O inclusions. There is no obvious difference in the mode of occurrence of these two types.

Chlorite-actinolite schist

The mineral assemblage of the CAS is actinolite + chlorite + titanite + albite ± tourmaline (Figure 3d). The actinolite is dominant in the mode (85 and 93 vol% in outcrops 1 and 2, respectively), defines the schistosity, and occurs as euhedral grains of 0.1 to 5 cm long. Chlorite, titanite, and albite occur in the interstices among the actinolite grains.

Whole-rock chemistry

The whole-rock compositions of the samples are listed in Additional file 1: Table S1. As shown in Figure 4, carbonaceous material is the major reservoir of carbon. The sample NK11 contains both carbonaceous material and calcite (Figure 4), but division between C and CO_2 is

difficult. Thus, carbon in all the samples is regarded as C in the mass balance analysis discussed below.

Discussion

Bleaching process of pelitic schist

The bleaching of the pelitic schist involves the changes in mineralogy and whole-rock chemistry. There are two possible mechanisms. As the UPS is a boundary layer, it may represent diffusive exchange of components between the pelitic schist and ultramafic mélange matrix. Boundary flow of external fluids may cause infiltration metasomatism of the pelitic schist.

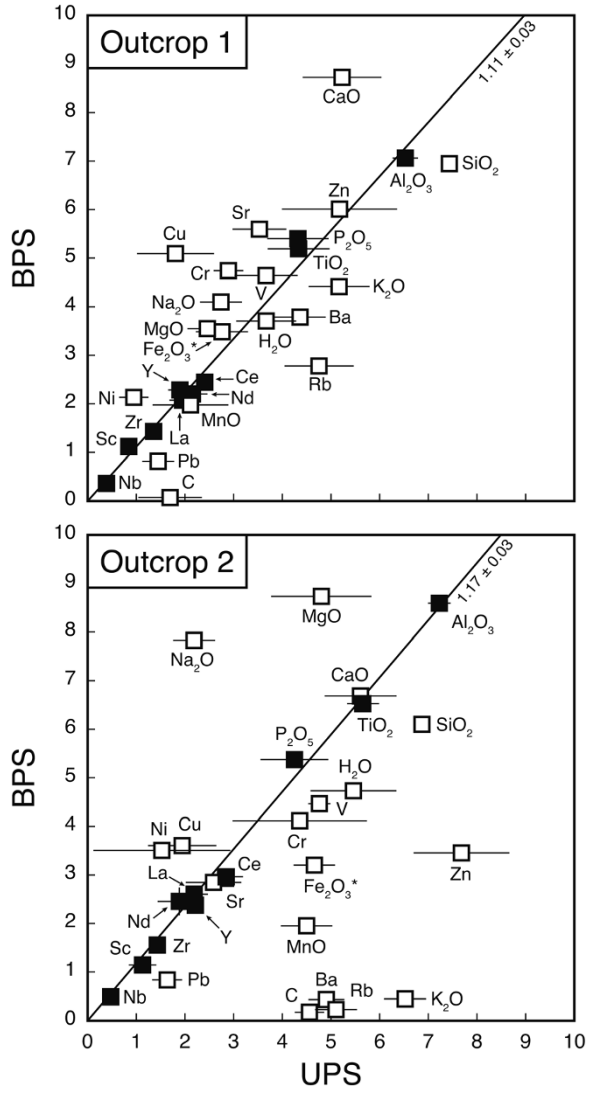

Figure 5 Isocon diagrams for BPS. Concentrations are scaled as follows: ×0.1 for SiO_2; ×0.5 for Al_2O_3; ×1 for total iron (as Fe_2O_3) and Na_2O; ×2 for MgO, K_2O, and H_2O; ×10 for TiO_2, CaO, and C; ×50 for MnO and P_2O_5; ×100 for Zr and Ba; ×500 for V, Cr, Ni, Rb, Sr, Nb, and Ce; and × 1,000 for Sc, Cu, Zn, Y, La, Nd, and Pb. Open and solid squares show mobile and immobile components, respectively. The data are listed in Additional file 1: Table S2.

To examine the bleaching mechanisms, the chemical mass balance between the UPS and BPS was estimated using the isocon method (Grant 1986). As shown in Figure 2, the UPS gradually changes into the BPS upon approaching the margin of the pelitic schist block. There are no structural discontinuities between them, in contrast to the contact between the BPS and CAS. Thus, the UPS is considered the protolith of the BPS. In each outcrop, the average composition of the UPS was used as the unaltered reference frame. We regarded the standard deviation of the UPS compositions as the inherent heterogeneity of the protolith and used it to calculate the uncertainties of the mass balance analysis (Baumgartner and Olsen 1995).

The isocon diagrams for the BPS are shown in Figure 5. The isocon is defined by TiO_2, Al_2O_3, P_2O_5, Sc, Y, Zr, Nb, La, Ce, and Nd. These components are thought to be immobile in this metasomatic system. In outcrop 1, the isocon slope (=1.11 ± 0.03) indicates that the UPS had lost 10% of its bulk mass during the bleaching. The elemental mass changes include losses of 15% SiO_2, 23% K_2O, 95% C, 47% Rb, 21% Ba, and 47% Pb, along with gains of 33% MgO, 34% Na_2O, 109% Ni, and 161% Cu, relative to the protolith values. The isocon diagram for outcrop 2 shows more dispersive distribution of data than that for outcrop 1, suggesting more intense metasomatism. In

Table 1 Element mass changes in BPS

	Outcrop			
	1		2	
	Sample NK14		Sample NK24	
	(g/100 g of protolith)	(% change)	(g/100 g of protolith)	(% change)
SiO_2	−11 ± 2	−15	−16 ± 2	−23
$Fe_2O_3^a$	+0.4 ± 0.5	+14.9	−1.9 ± 0.4	−40.0
MnO	−0.00 ± 0.02	−11.87	−0.06 ± 0.01	−62.60
MgO	+0.4 ± 0.2	+32.9	+1.4 ± 0.5	+56.6
CaO	+0.27 ± 0.08	+51.44	+0.01 ± 0.07	+2.25
Na_2O	+0.1 ± 0.4	+34.4	+4.5 ± 0.4	+203.7
K_2O	−0.6 ± 0.3	−23.2	−3.1 ± 0.2	−93.4
H_2O	−0.16 ± 0.31	18.75	−0.7 ± 0.5	−25.1
C	−0.16 ± 0.07	−94.78	−0.44 ± 0.03	−95.70
	(μg/g of protolith)	(% change)	(μg/g of protolith)	(% change)
V	+11 ± 13	+15	−19 ± 6	−19
Cr	+28 ± 6	+48	−15 ± 28	−18
Ni	+21 ± 6	+109	+30 ± 28	+96
Cu	+29 ± 8	+161	+12 ± 7	+59
Zn	+2 ± 12	+5	−47 ± 10	−61
Rb	−45 ± 14	−47	−98 ± 9	−95
Sr	+32 ± 11	+46	−2 ± 12	−4
Ba	−92 ± 52	−21	−453 ± 36	−91
Pb	−7 ± 3	−7	−9 ± 3	−55

[a]Total iron as Fe_2O_3.

outcrop 2, the isocon slope (=1.17 ± 0.03) indicates that the UPS lost 14% of its bulk mass during the bleaching. The elemental mass changes include losses of 23% SiO_2, 93% K_2O, 96% C, 95% Rb, 91% Ba, and 55% Pb, along with gains of 57% MgO, 204% Na_2O, 96% Ni, and 59% Cu, relative to the protolith values.

The chemical mass balance suggests that the bleaching of the pelitic schist is an open system with respect to some mobile components. For example, the significant gain of Na_2O (Table 1) is probably unable to explain by simple exchange between the pelitic schist and ultramafic mélange matrix. As the Na_2O concentration is typically low in the ultramafic mélange matrix, influx of Na-rich fluid would be necessary. Although the diffusive exchange of components is possible, the fluid flow and infiltration metasomatism are considered more important mechanisms of the bleaching.

Element mobility and mineral stability

The elemental mass changes (Table 1) are coupled to the stability of the key host minerals in the BPS (Figure 4). The loss of SiO_2 and C corresponds to the decrease of quartz and the consumption of carbonaceous material, respectively. The loss of K_2O, Rb, and Ba is correlated with the decrease of muscovite. The gain of Na_2O and MgO reflects the increase of albite and chlorite, respectively. In outcrop 1, the gain of CaO and Sr is comparable with the increase of epidote [see Additional file 1: Table S1]. Some immobile components such as TiO_2, Y, Nb, La, Ce, and Nd are probably held in titanite that remains stable. The mass change of total iron is inconsistent with the modal change. This may be because total iron contains ferric (mainly in epidote and chlorite) and ferrous oxides (mainly in chlorite).

A salient change during the bleaching of the pelitic schist is the consumption of carbonaceous material (Figures 3 and 4). As the BPS and CAS have the low C concentration and do not contain carbonate minerals, we infer that the infiltration of fluid from the mélange matrix converted the carbonaceous material to carbonic fluids. If the infiltrating fluid is reducing relative to the pore fluid in the pelitic schist, the carbonaceous material will be converted to hydrocarbons (CH_4, etc.). Alternatively, if the infiltrating fluid is oxidizing, the carbonaceous material will be converted to CO_2. The CH_4-rich ($\pm CO_2$) fluid inclusions in the BPS may be the traces of such processes (Figure 3c). Unfortunately, there are few records of the redox state of the fluids in the samples. Nishiyama (1990) described CO_2 metasomatism of an exposed metabasite block in the Mie mélange 2.5 km northeast of Nishikashiyama (Figure 1). He speculated that the origin of the CO_2-rich fluid ($X_{CO_2} > 0.785$) was the oxidation of carbonaceous material in pelitic schist blocks. However, such a process may not be a suitable explanation for the case of the Nishikashiyama mélange because the BPS and CAS contain titanite and actinolite (Figure 3b, d). These minerals are destabilized under high- X_{CO_2} conditions (*cf.* Nishiyama 1990).

Implications for mass transfer along the slab-mantle interface

The most striking result of the mass balance analysis is the obvious correlation between the elemental mass changes and the ion radius and charge number. As shown in Figure 6a, a significant mass of LILE (Rb, K, Ba) and Pb

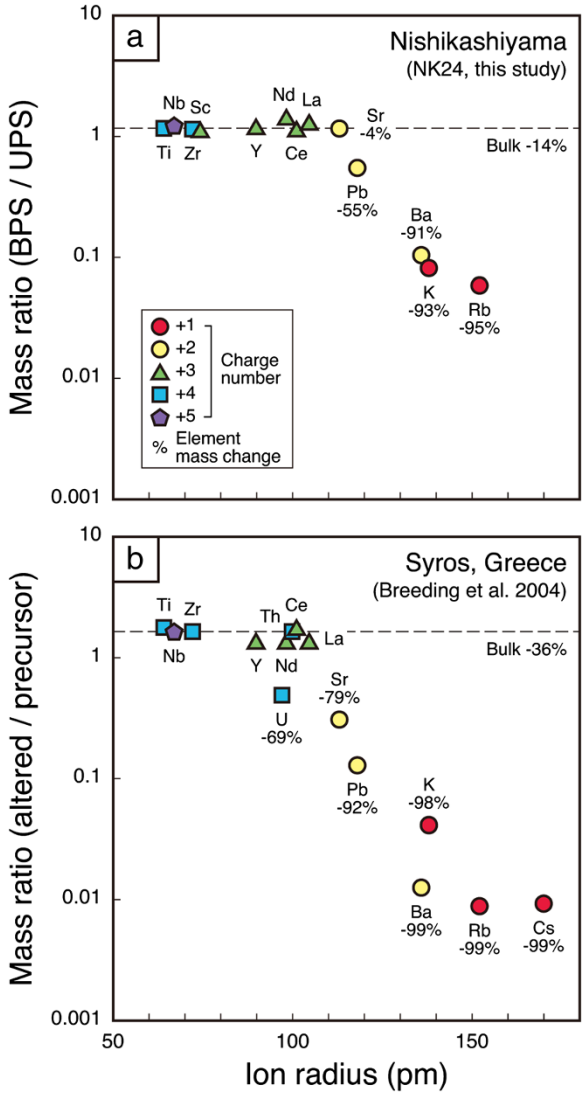

Figure 6 Selected element mobility during fluid-metapelite interaction in subduction zone mélange. Mass ratio is plotted against the ion radius (six-coordinated). Estimated bulk and element mass changes are indicated as percentage relative to the protolith abundance. **(a)** Nishikashiyama, this study. **(b)** Syros, data from Breeding et al. (2004).

with relatively large ion radii and low charge numbers is lost in the BPS. In contrast, the high-field-strength elements (HFSE; e.g., Ti, Nb, and Zr) and rare earth elements (REE; e.g., La, Ce, and Nd) remain immobile. Consequently, the infiltration metasomatism causes fractionation of LILE and Pb from the HFSE and REE along with the bleaching of the pelitic schist. Because the CAS has low concentrations of LILE and Pb [see Additional file 1: Table S2], these components are probably liberated from the pelitic schist blocks and migrate away via the fluid flow.

Breeding et al. (2004) reported similar fluid-metapelite interaction from an eclogite facies mélange exposed in Syros, Greece. Near the meta-ultramafic mélange matrix, the mineral assemblage of the metapelite was altered from phengite + sodic pyroxene + epidote + garnet + glaucophane + quartz + titanite + rutile + zircon to glaucophane + chlorite + albite + magnetite + apatite + titanite + rutile + zircon (Figure 6b). The difference between the mineralogical changes in Syros and Nishikashiyama probably reflects the difference in the *P-T* conditions of the fluid-metapelite interaction. Nevertheless, the elemental mass changes are similar (Figure 6). LILE, Pb, Sr, and U are significantly lost during the alteration, but the HFSE, REE, and Th remain nearly immobile. Such a fluid-metapelite interaction and element fractionation may occur at various depths (eclogite facies and epidote-blueschist facies to lower *P-T* conditions) along the slab-mantle interface in subduction zones.

Conclusions

This paper describes bleaching of pelitic schist blocks in an ultramafic mélange intercalated with epidote-blueschist facies schists of the Nishisonogi metamorphic rocks. The bleaching is attributed to infiltration of Na-rich external fluid and involves liberation of C (as carbonic fluids), Si, Rb, K, Ba, and Pb from the pelitic schist. Titanium, Nb, Zr, La, Ce, and Nd remain immobile, so the LILE and Pb are fractionated from the HFSE and REE. Such a fluid-metapelite interaction may occur at various depths along the slab-mantle interface that is the site of tectonic mixing of sedimentary, mafic, and ultramafic rocks. Therefore, ultramafic mélanges are a potential source of slab-derived components and play an important role in material recycling in subduction zones.

Additional file

Additional file 1: The file contains two tables showing modal compositions of samples (Table S1) and whole-rock compositions of samples (Table S2).

Abbreviations

Ab: albite; Act: actinolite; BPS: bleached pelitic schist; Cal: calcite; CAS: chlorite-actinolite schist; Chl: chlorite; CM: carbonaceous material; Ep: epidote; Ms: muscovite; Qtz: quartz; Ttn: titanite; UPS: unaltered pelitic schist.

Competing interests

The authors declare that they have no competing interests.

Authors' contributions

YM carried out the field investigation and sampling, performed the microscopic observation and analyses (Raman, XRF, CHN, and LOI), and drafted the manuscript. MS participated in the design of the study and helped in the field investigation, sampling, and CHN analysis. TN participated in the design of the study and helped in the field investigation and Raman analysis. All authors read and approved the final manuscript.

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Acknowledgements

This study is financially supported by Grant-in-Aid for Young Scientists B (Nos. 16740303 and 21740385 to YM) from Japan Society for the Promotion of Science and by Grant-in-Aid for Scientific Research A (No. 17204045 to TN) and Grant-in-Aid for Scientific Research C (No. 24540524 to YM) from the Ministry of Education, Culture, Sports, Science and Technology, Japan. We appreciate reviews and comments by T. Morishita and anonymous reviewers.

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Received: 10 December 2013 Accepted: 16 April 2014
Published: 4 June 2014

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doi:10.1186/1880-5981-66-47
Cite this article as: Mori *et al.*: Fluid-metapelite interaction in an ultramafic mélange: implications for mass transfer along the slab-mantle interface in subduction zones. *Earth, Planets and Space* 2014 **66**:47.