Subducted Carbonates And Their Role In The Metasomatism Of Mantle Wedges: An Example From California

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ABSTRACT
We investigated calcite veins in two spinel-garnet peridotite xenoliths from the sub-Sierra Nevada mantle. These peridotites represent samples of the lithospheric mantle wedge beneath the Mesozoic California magmatic arc, as indicated by radiogenic isotopic ratios measured on clinopyroxenes separates ($^{87}$Sr/$^{86}$Sr $_{\text{Ma}}= 0.7058 - 0.7061$, $^{143}$Nd/$^{144}$Nd $_{\text{Ma}}= 0.512538 - 0.512601$). Mineral chemistry of the peridotite major phases is typical of a mantle section that was depleted of melt. The $\delta^{18}$O values of olivine and orthopyroxene from the two samples are also typical of mantle rocks ($\delta^{18}$O = 6-6.5 ‰). Calcite veins, in contrast have $\delta^{18}$O of 18-20 ‰ and $\delta^{13}$C of −14 ‰, arguing for a subducted sedimentary origin for these carbonates. Presumably, the carbonates were expelled from the down-going slab and fluxed into the overlying mantle wedge as CO$_2$- or CO$_2$-H$_2$O rich fluids or melts. The trace element patterns of two analyzed calcite veins are typical of the arc signatures (e.g. depletions in Nb, Ta) seen in calc-alkaline magmatic rocks worldwide. However, the cores of peridotite clinopyroxenes do not show that pattern, suggesting that at least in this case, the arc trace element signature was introduced via the recycled carbonate agent. A connection between mantle wedge carbonation and diamond formation in a subduction environment is proposed.
INTRODUCTION

Mantle metasomatism at subduction zones is thought to be one of the key processes that lead to arc magmatism (Davies and Stevenson, 1992). Fluids released by the downgoing slab are thought to flux into the upper plate (the mantle wedge) and depress the peridotite solidus (e.g. Wyllie, 1984). Although these fluids are primarily aqueous, carbonic fluids can also play an important role in mantle metasomatism and melting (Wood et al., 1996). In fact, CO₂ fluxes in arc volcanoes are larger than those at mid-ocean ridges (Sano and Williams, 1996; Marty and Tolstikhin, 1998) and thus the largest deep reservoir releasing CO₂ into the atmosphere. Carbon isotopic ratios are used to estimate that about 80% of the CO₂ inventory in arc volcanoes is derived from recycled sediments, i.e. organic sediment and marine limestones (Plank and Langmuir, 1998), the remaining being primary mantle carbon. These observations indicate that CO₂ fluxing in the mantle wedge may be an important ingredient in generating arc magmas. However, available experimental (e.g. Molina and Poli, 2000) and thermodynamic (e.g. Kerrick and Connolly, 1998, 2001) data indicate that decarbonation takes places at higher temperatures than dehydration in the slab and that carbonate rocks may persist in the slab at depths greater than those sampled by arc volcanoes. Overall, quantifying carbonate recycling at subduction zones is a particularly important challenge given the need to understand the role of fluids in arc magmatism as well as the atmospheric carbon cycle over geologic timescales. It is also plausible that much of the recycled CO₂ in arc volcanoes may be derived from sedimentary sections that were emplaced in the lower crust beneath arcs via shortening and not from the subducted slab; this hypothesis has yet to be tested (Stern, 2002). The issue of how much CO₂ is released from the slab into the upper mantle and ultimately in arc volcanoes is very much unresolved and currently suffers from little direct (observational) evidence.

Rare direct observations on upper mantle xenoliths from arc environments confirm the presence of carbonic fluids in the mantle. In this study we present new trace element and isotopic analyses from carbonated peridotite xenoliths from the central Sierra Nevada, California (Dodge et al., 1988). This is the first report of calcite-bearing peridotites from these localities. Crustal and mantle xenoliths from Miocene pipes in the central Sierra Nevada represent fragments of the Mesozoic subarc lithosphere of this
classic Cordilleran subduction zone (Ducea and Saleeby, 1998). We use isotopic tracers to show that the carbonates are recycled via the subducted Farralon slab, and that they imprint an arc-like trace element signature on peridotite clinopyroxenes, a signature that the pyroxenes did not possess originally. Carbonate fluxing may also be responsible for generation of sub-arc diamonds.

SAMPLES AND TECHNIQUES

The Sierra Nevada batholith is a typical Mesozoic Cordilleran magmatic arc, composed primarily of tonalitic and granodioritic plutons. Granitoid rocks are known from present-day surface exposures to extend to a depth of at least 30 km in the Cretaceous crustal column (Saleeby, 1990; Ducea, 2001). Samples of more deeply seated rocks representing the lower crust and the upper mantle beneath the arc are entrained as xenoliths in volcanic rocks of Miocene age from the central Sierra Nevada (Dodge et al., 1988; Mukhopadhyay and Manton, 1994; Ducea and Saleeby, 1998). They include granulite and eclogite facies rocks of the arc root as well as spinel-garnet and garnet-bearing peridotites, which represent samples of the mantle wedge beneath the arc ((Ducea and Saleeby, 1998).

Two large (5-8 cm in diameter) calcite-bearing spinel-garnet peridotites (BC115 and BC125) from the Miocene (8.3 Ma) Big Creek trachyandesitic pipe (Dodge et al., 1988; Mukhopadhyay and Manton, 1994; Ducea and Saleeby, 1996, 1998; Lee et al., 2000) in the Huntington Lake area, central Sierra Nevada, were selected for analyses. The samples are typical for the peridotite suite of the central Sierra Nevada and contain spinels that are rimmed by garnets. Calcite veins make up about 3-5% of the analyzed samples. Calcite crystals are large (up to 0.4 cm), which indicates high temperature crystallization of these veins.

Electron microprobe analyses on minerals were carried out at the University of Arizona using the Cameca SX50 microprobe equipped with 5 LiF, PET and TAP spectrometers. Counting times for each element were 30 seconds at an accelerating potential of 15 kV and a beam current of 10 nA. Measurements with oxide totals outside of the range 100 ± 1% were discarded from the analyzed silicates.
Trace element analyses were performed on carbonates mechanically separated from the peridotites, as well as clinopyroxenes separates (about 80 mg). The following acid leaching routine obtained a rim fraction of clinopyroxenes: unbroken clinopyroxene grains were first leached in mild, cold 2.5M HCl for two hours. This step was performed in order to eliminate any additional carbonate that may reside at clinopyroxenes grain boundaries; then the leachate was discarded, the grains were dried out and inspected optically. The clean grains without any grain boundary staining were used for the next step. A second leaching step was carried out in a hot (150°C) mixture of concentrated HF and HNO₃; after two weeks of dissolution, about 1/3 of the clinopyroxenes mass was dissolved – the leachate was used for the “clinopyroxenes rim” fraction. The undissolved clinopyroxenes grains were then dried out, weighed, crushed into a powder and dissolved in the same manner – the dissolved samples were analyzed as the “clinopyroxenes core” fractions. About 10% of the dissolved core and rim samples were used for trace element determinations in an ICP-MS instrument, the remainder being eluted in standard chromatographic columns for TIMS analyses. For both fractions, the elution procedures for radiogenic isotopic analyses are as in Ducea and Saleeby (1998).

Trace element determinations were performed at Caltech on an Elan Ar source quadrupole ICP-MS. USGS standard BVHO-1 was used for calibrating the ICP-MS instrument. Sr and Nd isotopes were measured using the techniques described in Ducea and Saleeby (1998). Isotopic analyses were performed at Caltech on a VG Sector TIMS instrument (Ducea and Saleeby, 1998) and replicated at the University of Arizona on a Sector 54 VG instrument (Ducea et al., 2002).

Olivine mineral separates (1-2 mg) were analyzed for oxygen isotopic ratios at Caltech, following the techniques of Holt and Taylor (1998). The oxygen was extracted using a laser fluorination line, followed by analysis of isotopic ratios of major minerals, expressed as δ¹⁸O in per mil relative to SMOW. The extracted oxygen was converted to CO₂ using a heated carbon rod. This CO₂ was analyzed with a Finnigan MAT252 mass spectrometer. Precision is better than 0.2 per mil (1σ) for the olivine separates analyzed in this study. Stable isotopes were measured at University of Southern California using the techniques outlined in Woods et al. (1999).
RESULTS

Peridotites. The two analyzed peridotites have mineral chemistries typical for the sub-Sierra Nevada mantle, as known from previously reported data (Mukhopadhyay, 1989; Ducea and Saleeby, 1996; 1998). Olivines are Fo$_{91}$ in BC115 and Fo$_{92}$ in BC125, which is close to the average of Sierran peridotites in the central Sierra Nevada Miocene suites. These rocks are thought to be melt-depleted, possibly during several episodes of melting, but certainly during the formation of the overlying Sierra Nevada magmatic arc (Lee et al., 2000). The samples yielded equilibration temperatures of 950-1000$^0$C (using Brey and Kohler, 1990), which are typical for mantle peridotites from Big Creek (Dodge et al., 1988, Mukhopadhyay and Manton, 1994; Ducea and Saleeby, 1996). The equilibration pressure determined on BC115 is 26±2 kbar, and was determined using the Al-in orthopyroxene barometer (Harley, 1984). The high pressure measured in BC115, is consistent with the fact that the spinel-garnet transition can be as deep as 100 km beneath active continental margins (O’Neill, 1981; Robinson and Wood, 1998). BC125 did not yield a reliable pressure, possibly because the garnet, which is rimming spinel, is not in equilibrium with the orthopyroxene; however, the similarity in the overall composition of BC115 and BC125 makes it likely that BC125 has been sampled from a similar depth.

Radiogenic isotopic ratios on separated clinopyroxenes – age corrected to 8 Ma- are: $^{87}\text{Sr}/^{86}\text{Sr}= 0.7058-0.7061$, $^{143}\text{Nd}/^{144}\text{Nd}= 0.51254-0.51260$. These numbers are consistent with previously measured ratios on sub-Sierra Nevada peridotites (e.g. Dodge et al., 1988; Mukhopadhyay and Manton, 1994; Ducea and Saleeby, 1998) and are indicative of a lithospheric mantle origin for the peridotites. The $\delta^{18}$O measured on olivine from the two peridotites (6.4 and 6.5 ‰) are within the range, although on the high side of oxygen isotopic ratios measured worldwide on olivines from the continental mantle (Rosenbaum et al., 1994).

Chondrite-normalized trace element patterns (Table 2) measured on clinopyroxene cores are similar to other measurements on peridotite clinopyroxenes from the Sierra Nevada (Domenick et al., 1983; Ducea, 1998), and are slightly enriched in incompatible elements (Figure 2). Clinopyroxene rims have significantly higher (up to five times) concentrations of incompatible trace elements relative to their cores (Figure
2), and display negative anomalies of the high field strength elements (Nb, Ta, Zr and Ti). The rims of the clinopyroxenes have been chemically modified by metasomatism.

**Carbonates.** The analyzed carbonate vein material in both samples is calcite, with small, <0.7 % MgO. The Mg# of the calcite is ~90, which shows that the carbonate veins have equilibrated – in this respect - with the olivine-rich, peridotite matrix. Sr and Nd isotopic ratios ($^{87}$Sr/$^{86}$Sr= 0.7081-0.7083, $^{143}$Nd/$^{144}$Nd= 0.51221-0.51223) are distinct from the values measured in clinopyroxenes separates and are within the range of Cenozoic Pacific waters (Faure, 1986). The $\delta^{18}$O values are very high (18.6-20.5 ‰), and clearly indicative of a sedimentary origin for the carbonate. Moreover, the negative values of $\delta^{13}$C (-14.1 and -14.3 ‰) strongly suggest an organic origin for the CO$_2$.

Chondrite-normalized trace element patterns of carbonate veins (Fig 2) show negative anomalies of the high field strength elements such as Nb, Ta, Zr and Ti. The overall concentrations of trace elements in the analyzed calcite veins materials are typical of trace element concentrations in sedimentary carbonates and muds (Sammartano et al., 2002). In contrast, those of mantle-derived, primitive carbonatites tend to be significantly (an order of magnitude) more enriched in incompatible elements (e.g. Bell, 1989).

**INTERPRETATIONS**

**Mantle origin for carbonates.** The low $\delta^{13}$C and high $\delta^{18}$O of the analyzed carbonate material may be suspected to be of surficial origin, i.e. that the veins formed after the xenoliths were brought up to the surface or during volcanic entrainment and ascent. There are three observations that argue for a mantle origin of the calcite vein material: (a) texturally, the calcite crystals are large, up to 4 mm, unlike surficial calcite deposits; (b) the calcite veins are restricted to the xenoliths and do not cut into the host trachyandesite; and (c) calcite formed at low temperature in equilibrium with meteoric waters at the latitude of the Sierra Nevada should have $\delta^{18}$O closer to 0 relative to SMOW, much lower than the measured values (~ +20 ‰). We therefore interpret the calcite veins to have formed while their host peridotites resided in the upper mantle.

**Timing of carbonate metasomatism.** High temperature oxygen isotopic equilibrium between calcite and olivine requires a $\Delta$<3 ‰ at 1000-1200 °C (Clayton et al., 1989), which was not achieved in our samples. The lack of stable isotopic equilibrium
between the calcite and peridotites indicate that fluxing of peridotites with the carbonate fluid took place shortly, < 10^6 Yr before the eruption of the host volcanic plug at 8.3 Ma (Dodge et al., 1988). A similar disequilibrium was used to argue for Cenozoic metasomatism of the Slave province mantle lithosphere with carbonatite melts (van Achterbergh et al., 2002).

**Sedimentary origin of the carbonate precursors.** The stable isotopic ratios of the measured calcite unambiguously demonstrate an organic sedimentary origin for the carbonates. The low δ^{13}C is typical of organic sediments. A low δ^{13}C carbon has been found in many mantle xenoliths (Deines, 2002, for a review), although none of the determinations were made on carbonates- instead, data were obtained on graphite, diamond, CO₂-rich inclusions, or silicate mantle minerals. We interpret the stable isotopic signatures on carbonate veins to indicate that the source of carbon and oxygen is a surficial, organic component that was recycled into the mantle via subduction and then released into the mantle wedge.

The Nd isotopic ratios measured on the two carbonates are consistent with a Pacific ocean origin and a Cenozoic age of the sediment (Shaw and Wasserburg, 1985). Moreover the Sr ratios, if in equilibrium with Pacific water and not modified during metamorphism, subduction and decarbonation, would indicate a 30-35 Ma age of the sediment, using the Sr oceanic curve from Faure (1986); this predates the age of the xenolith entrainment by ~20-25 My.

**Not a typical carbonatite.** The concentrations of incompatible trace elements measured on the calcites in BC115 and BC125 are significantly lower than typical mantle-derived carbonatites (Bell, 1989). Additionally, the anomalous negative spikes of the high field strength elements is opposite from trace element patterns in carbonatites worldwide. Based on these observations, one may argue that these carbonate vein materials were not melts percolating through the upper mantle but CO₂-rich fluids. However, the presence of accompanying silicate glass pockets (Figure 1) in BC115 suggests that a melt phase was in fact present. A water-rich fluid could also be the carrier of dissolved carbonate. Distinguishing upper mantle carbonate “melt” from CO₂-rich or H₂Oand CO₂-rich “fluid” based on textural and chemical criteria may be impossible – the important observation is that these calcite veins are different in trace element
concentration and carbon and oxygen isotopes than most carbonatites worldwide (Wooley, 2001).

DISCUSSION

Release of subducted CO$_2$ in the mantle wedge. Release of volatiles from the subducted slab into the mantle wedge (Tatsumi and Eggins, 1995; Schmidt and Poli, 1998) is a critical process in triggering arc melting (Davies and Stevenson, 1992). Water is clearly the most important volatile and its release by dehydration metamorphic reactions has been extensively studied (Stern, 2002, for a review). The release of CO$_2$ is not well understood, despite many observations suggesting that CO$_2$ fluxes in arc volcanoes are significant (Sano and Williams, 1996; Marty and Tolstikhin, 1998). Some carbonate phases previously observed in peridotite xenoliths, e.g. from beneath the Slave craton (van Achterbergh et al., 2002) and the Alpine peridotite of Finero (Zanetti et al., 1999) could potentially have formed by slab decarbonation reactions at some point in the geologic past, but a direct link could not be established due to the antiquity of the postulated subduction events. To our knowledge, the only other direct observation of carbonated peridotites from beneath a modern subduction zone is from the Tabar-Lihir-Tanga-Feni arc (Papua New Guinea), described by McInnes and Cameron (1994).

Calcite veins described in this paper can be directly linked to the subduction of the Farralon plate beneath western North America and provides some direct qualitative evidence that subducted sedimentary carbonates can be released in the mantle wedge. Subduction was in its waning stages beneath central California (Atwater and Stock, 1998) at the time of the eruption of the Big Creek pipe (~ 8.3 Ma, which must be almost contemporaneous with mantle carbonation in this case). The Sierra Nevada was the main magmatic arc at this latitude for much of the Mesozoic, but it became the forearc as magmatism migrated inland during the Laramide orogeny (Saleeby, 2003). Therefore, these carbonates were released from the downgoing slab into the forearc wedge of this subduction zone.

Subducted carbonates and arc signatures. The analyzed calcite veins have typically higher concentrations of incompatible elements than the peridotite clinopyroxenes as measured in their core fractions. The clinopyroxene rims have
interacted with the metasomatizing carbonate veins, resulting in trace element concentrations intermediate between the calcite and clinopyroxenes cores. One of the most interesting chemical characteristics of the analyzed carbonate veins is that they carry trace element signatures typical for arc volcanoes (Pearce and Peate, 1995) and arc magmas in general. As a metasomatic agent, the carbonate fluid has apparently interacted with the matrix peridotite and imprinted the “arc” chemical signature onto the clinopyroxenes rims. Since the studied peridotites represent fragments of the mantle wedge of a long-lived subduction zone, we speculate that carbonates recycled via subduction may contribute to generating negative HFSE anomalies in mantle wedge regions of subduction zones.

**A connection to sub-arc diamonds?** Several major rivers from the Sierra Nevada contain diamondiferous sediments (Kopf et al., 1990; Heylmun, 1999; Hausel, 1995) that are thought to have a proximal, i.e. Sierran origin based on their crystal size and habit. Recently diamonds have been identified in the trachyandesitic Miocene pipe of Leek Springs from the northern Sierra Nevada (Patrick Hourican, personal communication, 1999), north of, but compositionally similar to the pipe of Big Creek. Recently published experimental data show that percolation with carbonate through a mantle column can lead to diamond formation (Arima et al, 2002). The anomalously low thermal budget of any subduction zone increases the stability field of diamond relative to graphite to shallower depths than in cratonic areas, potentially as shallow as 90-100 km. Big Creek peridotites might be too shallow to preserve diamonds; however future investigations of the C-bearing phases and reactions in Miocene central Sierra Nevada peridotites could provide further insights into the possible link between carbonate metasomatism and diamond formation in a mantle wedge environment.

**CONCLUSIONS**

We use carbon and oxygen isotopic ratios to show that calcite veins in peridotites from the sub-Sierra Nevada mantle wedge are derived from materials that were previously at or near the surface of the Earth, probably sedimentary carbonates that were subducted with the Farallon plate. These results underscore the importance of carbonic fluid metasomatism in the mantle wedge beneath arcs. In particular, it is shown here that
the depletions in high field strength elements, which are common in arc volcanic rocks, can be introduced in the mantle by recycled carbonate metasomatism. The trace element composition of the analyzed carbonate vein materials suggests that the CO₂-rich metasomatic fluid, possibly a melt, had a trace element composition very different from most carbonatites. We speculate that the sub-arc Sierran diamonds may have formed during reduction reactions of these carbonate fluids as they were passing through the mantle wedge.

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FIGURE CAPTIONS

**Figure 1**: Microphotographs. A. Cross polarized view of peridotite BC115 showing calcite vein material in the lower right corner of the section. Note the porphyroclastic texture of the peridotite. B. Detailed view of a silicate glass pocket from BC125; the glass, which is isotropic, is intermixed with calcite.

**Figure 2**: Trace element concentrations normalized to chondritic values in calcite veins (diamond symbols), clinopyroxene rims (square symbols) and clinopyroxene cores (circles) from peridotites: (A) BC115, and (B) BC125. Chondrite normalization values are from McDonough (1995).
Table 1. Olivine major element compositions in carbonated peridotites BC115 and BC125. Analytical techniques and errors are described in text.

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<th></th>
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<th>BC115 olivine rim</th>
<th>BC125 olivine core</th>
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Table 2. Trace element concentrations of clinopyroxene cores, rims, and calcite veins (ppm). See text for analytical details and uncertainties.

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<th>BC125</th>
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<td>3.36</td>
<td>1.51</td>
<td>5.34</td>
<td>3.55</td>
<td>2.92</td>
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<td>Hf</td>
<td>0.12</td>
<td>0.11</td>
<td>0.04</td>
<td>0.15</td>
<td>0.12</td>
<td>0.08</td>
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<tr>
<td>Ti</td>
<td>0.03</td>
<td>0.03</td>
<td>0.02</td>
<td>0.05</td>
<td>0.04</td>
<td>0.03</td>
</tr>
<tr>
<td>Yb</td>
<td>0.16</td>
<td>0.17</td>
<td>0.47</td>
<td>0.24</td>
<td>0.23</td>
<td>1.22</td>
</tr>
</tbody>
</table>
Table 3. Isotopic compositions.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$^{87}\text{Sr}/^{86}\text{Sr}$</th>
<th>$^{143}\text{Nd}/^{144}\text{Nd}$</th>
<th>$\delta^{13}\text{C}$</th>
<th>$\delta^{18}\text{O}$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>BC115</strong></td>
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<td></td>
<td></td>
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</tr>
<tr>
<td>Olivine</td>
<td>6.4</td>
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<tr>
<td>Clinopyroxene</td>
<td>0.705821</td>
<td>0.512601</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Calcite vein</td>
<td>0.708112</td>
<td>0.512231</td>
<td>-14.3</td>
<td>18.6</td>
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<tr>
<td><strong>BC125</strong></td>
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<tr>
<td>Olivine</td>
<td>6.5</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Clinopyroxene</td>
<td>0.706105</td>
<td>0.512538</td>
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<tr>
<td>Calcite vein</td>
<td>0.708323</td>
<td>0.512212</td>
<td>-14.1</td>
<td>20.5</td>
</tr>
</tbody>
</table>

1- Strontium and Neodymium isotopic data are age corrected to 8 Ma, the age of the eruption of the host lava. The Sr isotopic ratios of standards and samples were normalized to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$, whereas the Nd isotopic ratios were normalized to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$. Sr and Nd standards are identical to values previously measured on the same batches of standards. The estimated analytical ± 2σ uncertainties for samples analyzed in this study are $^{87}\text{Sr}/^{86}\text{Sr} = 0.002\%$ and $^{143}\text{Nd}/^{144}\text{Nd} = 0.002–0.003\%$. 
Figure 1, Ducea et al.
Figure 2, Ducea et al.