

Were deep cratonic mantle roots hydrated in Archean oceans?

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ABSTRACT

Using a compilation of mantle peridotites ($n = 2153$), we statistically evaluate the frequency of Si enrichment in cratonic and other types of mantle lithosphere. We define an empirical parameter ($\Delta\text{Mg}/\text{Si}$) describing the degree of Si enrichment or depletion in mantle lithosphere relative to residue trends defined by high-pressure, high-temperature melting experiments. Silica enrichment (a strong skew to negative $\Delta\text{Mg}/\text{Si}$) is absent in the majority of cratonic xenoliths, and mostly occurs only in those from southern Africa, and in abyssal peridotites. The Si-rich composition of cratonic mantle, where it occurs, can be linked to the hydration of its protoliths on the Archean ocean floor before being subducted or imbricated to form a craton root. Oxygen isotopic shifts that correlate with bulk Mg/Si in mid-Atlantic ridge seafloor rocks parallel those seen in the few such data for cratonic peridotite xenoliths, in support of our hypothesis. Chemical variability in the mantle is canonically viewed to have originated from the bottom up by percolating melts. We turn this idea on its head, and explain how Si enrichment in the cratonic lithosphere could have originated by a top-down chemical exchange during weathering or hydrothermal activity when such peridotites resided on an Archean ocean floor.

INTRODUCTION

Mantle lithosphere is formed as a residue of melt extraction. Cratonic mantle lithosphere is host to diamonds, forms the roots to the earliest continents, and represents the most ancient mantle on Earth (Carlson et al., 2005; Pearson, 1999). Cratonic mantle as sampled by kimberlite-borne xenoliths is defined by a unique composition, being depleted in Fe and enriched in Si over Mg relative to lithosphere from other settings (Boyd, 1989; Griffin et al., 1999b). The origin of the Si-rich nature of cratonic mantle relative to many other kinds of mantle types (xenoliths, abyssal peridotites, ophiolite, orogenic massifs) is the source of debate, and bears on some major geological questions such as the composition of the “primitive mantle” and the formation of Earth’s first continents (Francis, 2003; Griffin et al., 1999a; Herzberg, 2004).

Various proposals for the Si enrichment in cratonic mantle mostly require metasomatism by upward emanation of melts or fluids from slabs or deep-seated magmas (Herzberg, 1999; Kelemen et al., 1998; Lee, 2006). Indeed, the metasomatism of mantle lithosphere is almost universally viewed as operating from the bottom upward. In this paper, we follow Schulze (1986) and test whether profound chemical modifications for mantle lithosphere, such as Si enrichment in cratonic mantle, could occur when mantle lithosphere resided near the surface before its subcretion to the deeper parts of the continents.

THE PERIDOTITE ARRAY

In a plot of Mg/Si versus Al/Si (Fig. 1A), the intersection of the array of mantle perido-

tite compositions with that of meteorite compositions provides an estimate of “primitive” upper mantle (PUM) (Jagoutz et al., 1979). The peridotite array arises as a product of either melt extraction of fertile mantle or re-fertilization of melt-depleted residues toward PUM during their residence in the lithosphere over billions of years.

The extent of Si enrichment in cratonic peridotites can be evaluated with a statistical examination of a large data set of many kinds of mantle lithosphere using the array of Mg/Si–Al/Si in peridotites. To compare all peridotites in this array, we plot the trend in residues from two well-constrained studies at 1 and 1.5 GPa on the experimental partial melting (EPM) of fertile peridotite (Baker and Stolper, 1994; Robinson et al., 1998). The EPM trends are fit to linear equations.

The mantle that now resides below continents has had a billion-year integrated history that has possibly modified its composition by metasomatism. For this reason, we first compare the EPM trend with samples of ophiolite mantle. The latter mantle type has formed from a relatively simple process (seafloor spreading) and is typically well sampled in number, size, and quality of samples taken due to extensive terrestrial exposures. Similar oceanic mantle occurs on the modern seafloor as abyssal peridotites, but we show below that a large portion of abyssal peridotites have compositions that have been compromised by seafloor processes.

THE $\Delta\text{Mg}/\text{Si}$ PARAMETER

We examine the extent of Si enrichment or depletion in ophiolite mantle relative to the baseline of the EPM trend by defining the $\Delta\text{Mg}/\text{Si}$ parameter. A sample that plots above the EPM trend has positive $\Delta\text{Mg}/\text{Si}$, and one below has negative. The value and sign of $\Delta\text{Mg}/\text{Si}$ could of course vary whether the 1.0 or 1.5 GPa EPM trend were used as a baseline (Fig. 1B), but this has no consequence as we are interested only in relative difference and frequency distribution of Si enrichment ($\Delta\text{Mg}/\text{Si}$) between different kinds of mantle samples. In what follows, we use the 1.5 GPa EPM trend as a baseline for comparisons of samples with experiments.

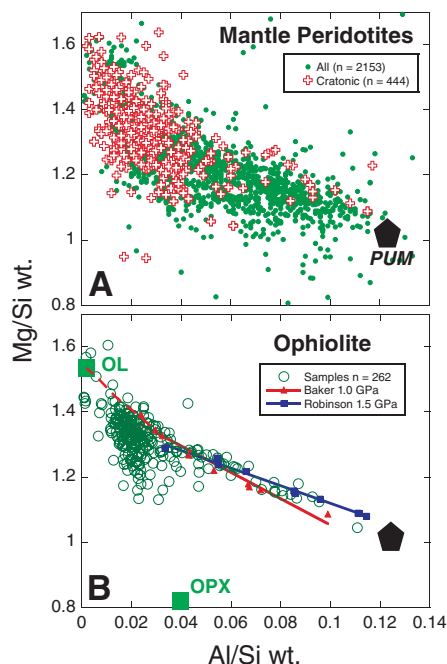


Figure 1. A: Covariation of Mg/Si with Al/Si (by weight) in world mantle peridotites (bulk-rock analyses of peridotites were compiled by Canil [2004], with additional data provided in the GSA Data Repository¹). Filled polygon is primitive upper mantle (PUM). **B:** Mg/Si versus Al/Si in ophiolite peridotites compared with residues derived from mass balance of experimental partial melting (EPM) data on peridotite at 1.0 GPa (triangles—Baker and Stolper, 1994) and 1.5 GPa (squares—Robinson et al., 1998). Note the extension of the EPM trend at 1.0 GPa to olivine (dashed line). The EPM residue data are fit by linear regression: At 1.0 GPa, $\text{Mg}/\text{Si} = -(4.05 \pm 0.29)(\text{Al}/\text{Si}) + (1.45 \pm 0.01)$, and $R = 0.98$; at 1.5 GPa, $\text{Mg}/\text{Si} = -(2.70 \pm 0.08)(\text{Al}/\text{Si}) + (1.39 \pm 0.01)$, and $R = 0.99$.

¹GSA Data Repository item 2009152, whole rock analyses of peridotites, is available online at www.geosociety.org/pubs/ft2009.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

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When compared with the 1.5 GPa EPM trend, we see that ophiolite mantle shows a frequency distribution of $\Delta\text{Mg}/\text{Si}$ that is normally distributed about zero (Fig. 2A). The small number of samples that range to extreme values in $\Delta\text{Mg}/\text{Si}$ of 0.2 to -0.2 is a heterogeneity in sampling. Some ophiolite peridotites (having $\text{Al}/\text{Si} < 0.04$) plot along a vector between olivine ($\text{Mg}/\text{Si} \approx 1.6$) and orthopyroxene ($\text{Mg}/\text{Si} \approx 0.75$, $\text{Al}/\text{Si} \approx 0.04$), attributed to sampling of mineralogically heterogeneous residues that vary from replacive dunites (Kelemen and Dick, 1995) to more orthopyroxene-rich rocks (Dick and Sinton, 1979).

Major elements determined in three different analytical labs vary less than 2% relative (e.g., Canil et al., 2006), equivalent to an uncertainty of less than 0.02 $\Delta\text{Mg}/\text{Si}$ units, which we choose as the bin size in Figure 2. The normal distribution about zero $\Delta\text{Mg}/\text{Si}$ implies that ophiolite mantle shows neither Si enrichment nor depletion.

SILICA ENRICHMENT IN CRATONIC MANTLE?

Comparisons of cratonic mantle to the EPM trend show that as a population they too have a mean $\Delta\text{Mg}/\text{Si}$ of zero, but unlike ophiolite mantle (Fig. 2A) are more broadly distributed (higher kurtosis). Closer scrutiny shows structure in the frequency distribution of cratonic mantle samples, with two distinct maxima above and below zero $\Delta\text{Mg}/\text{Si}$. This distribution with two modes does not depend on the facies (depth) from which the sample was derived (spinel versus garnet) (Fig. 2C). Instead, it is recognized that “Si-rich” mantle, having a negative $\Delta\text{Mg}/\text{Si}$, occurs in many cratons, but as an exception: a heterogeneous feature (Canil, 2008) that is only most frequently sampled in southern Africa in the Kaapvaal craton (Fig. 2D). The assertion that cratonic mantle is universally Si-enriched is not true on a statistical basis.

Samples of cratonic mantle from southern Africa are largely from the collections of F.R. Boyd, with major elements (Mg, Al, Si, ...) determined in one X-ray fluorescence (XRF) lab (Boyd and Mertzman, 1987). We evaluated whether Si enrichment is biased to this XRF laboratory, and no bias exists. The shift to negative $\Delta\text{Mg}/\text{Si}$ in southern African samples is not an analytical artifact. We find no correlation of $\Delta\text{Mg}/\text{Si}$ with any alteration parameters (e.g., loss on ignition).

Interestingly, of all other kinds of mantle, only abyssal peridotites exposed on the seafloor show the same distribution of $\Delta\text{Mg}/\text{Si}$ as southern Africa. Whole-rock analyses of abyssal peridotites show a large peak at $\Delta\text{Mg}/\text{Si}$ of zero, similar to ophiolite mantle formed in spreading centers, but a strongly skewed distribution to the most negative $\Delta\text{Mg}/\text{Si}$ of any other mantle type (Fig. 2E). Further scrutiny of abyssal peridotites shows that most samples on the mid-Atlantic ridge (MAR) from drill cores are normally distributed about zero, whereas those dredged from the East Pacific rise and Southwest Indian ridge (EPR and SWIR) show consistently negative $\Delta\text{Mg}/\text{Si}$ (Fig. 2F).

The skewed distribution to negative $\Delta\text{Mg}/\text{Si}$ in cratonic peridotites requires processes that remove Mg from Si, or add Si relative to Mg, from an original residue of partial melting. Both of these chemical shifts are well known in peridotites from modern ocean basins as products of marine weathering and hydrothermal alteration, respectively. Snow and Dick (1995) showed that marine weathering of abyssal peridotites in dredge samples from the seafloor leaches Mg relative to Si. Silica metasomatism of peridotites or serpentinites to produce talc-bearing rocks (low Mg/Si) is well documented at the MAR, where Si leached from higher-temperature alteration of gabbro is redeposited in shear zones as talc in serpentinites (Bach et al., 2004; Boschi et al., 2008). Metasomatism to produce low Mg/Si (or negative $\Delta\text{Mg}/\text{Si}$) in these modern cases has occurred at or near the surface, and is heterogeneous, just as it appears statistically in the cratonic peridotite data set.

CHEMICAL EFFECTS OF INTERACTION WITH THE HYDROSPHERE

Water-rock interaction on the seafloor shifts oxygen isotope ($\delta^{18}\text{O}$) compositions of peridotites from mantle values ($\sim 5.2\text{‰}$) to those of seawater (assumed zero) or to higher values ($>6\text{‰}$) depending on temperature of alteration (Gregory and Taylor, 1981). The shift to low $\delta^{18}\text{O}$ is well observed in the modern ocean at the Atlantis massif and correlates with changing Mg/Si of the bulk rocks during higher-temperature hydrothermal alteration to talc schists (Fig. 3A). The few data for cratonic peridotite xenoliths in which $\delta^{18}\text{O}$ is measured by high-

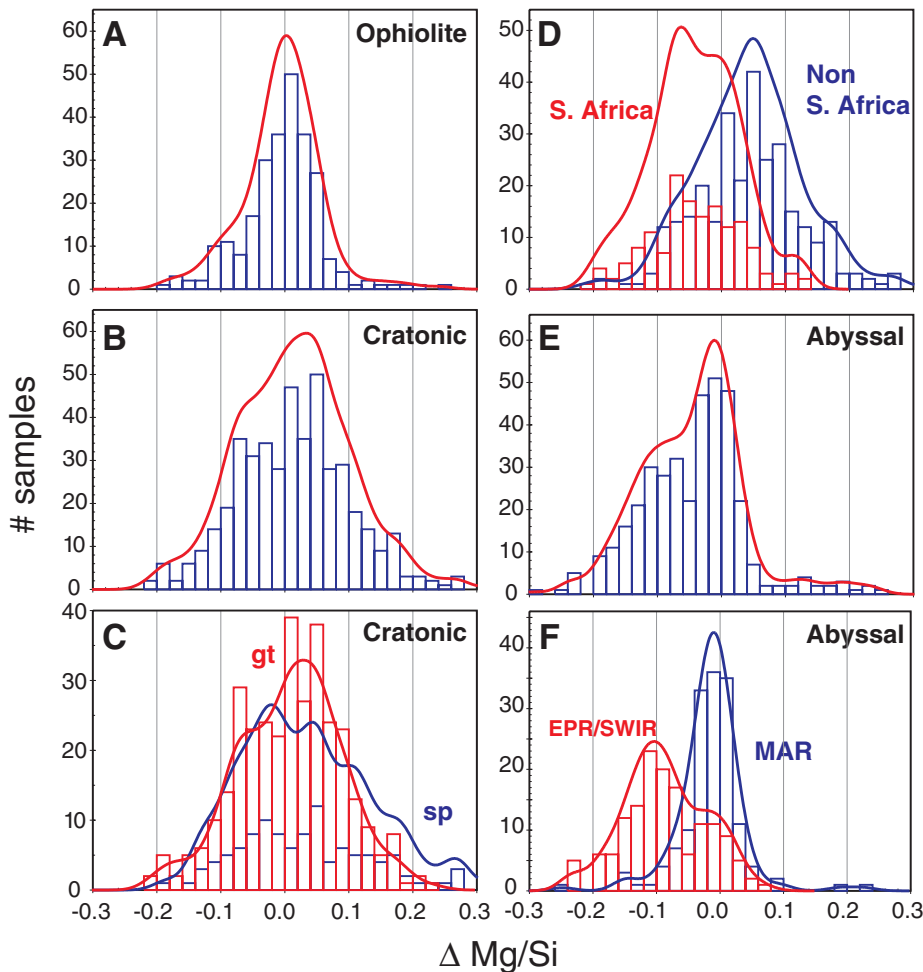


Figure 2. Frequency histograms and relative probability curves for $\Delta\text{Mg}/\text{Si}$ in ophiolite peridotites (A), cratonic peridotites (B), spinel- versus garnet-facies cratonic peridotites (C), world versus southern African cratonic peridotites (D), abyssal peridotites (E), and abyssal peridotites in dredge samples (EPR/SWIR) versus drill cores (MAR) (F). The $\Delta\text{Mg}/\text{Si}$ parameter for each sample is the difference in Mg/Si of the whole rock from that of the 1.5 GPa EPM residue trend at the same degree of depletion (Al/Si) (see Fig. 1B). Only “low-temperature” coarse peridotite xenoliths from kimberlites are considered as “cratonic” peridotites.

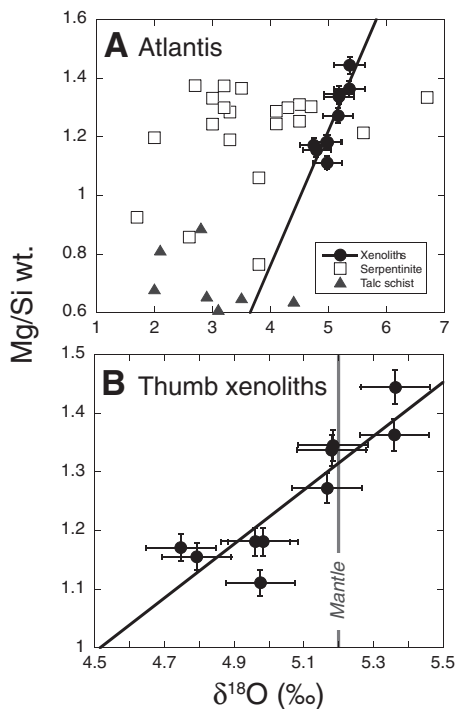


Figure 3. A: Comparison of whole-rock $\delta^{18}\text{O}$ with whole-rock Mg/Si (by weight) for altered peridotites (serpentinites and talc schists) from the Atlantis massif along the mid-Atlantic ridge (Boschi et al., 2008). Solid circles show similar data for xenoliths from The Thumb (Colorado Plateau, USA) using bulk chemical and modal data from Ehrenberg (1982) and $\delta^{18}\text{O}$ of minerals by laser fluorination (Mattey et al., 1994). **B:** As in A, showing only data for the Thumb xenoliths fit by linear regression: $\text{Mg/Si} = (0.46 \pm 0.08)\delta^{18}\text{O} - (1.08 \pm 0.4)$, and $R = 0.89$. Error bars for $\delta^{18}\text{O}$ are as reported in (Mattey et al., 1994) and assuming uncertainty of $\pm 2\%$ in Mg and Si.

precision laser fluorination show an excellent trend of lower $\delta^{18}\text{O}$ with lower Mg/Si, supportive of our hypothesis (Fig. 3B). The trend in cratonic peridotites extrapolates to the same shift in $\delta^{18}\text{O}$ observed with Si metasomatism during high-temperature hydrothermalism at the Atlantis massif (Fig. 3A). On the other hand, cratonic mantle garnets can also have $\delta^{18}\text{O}$ values greater than 6‰, suggestive of low-temperature alteration on the seafloor (Taylor et al., 2005).

Eclogites and diamonds that are commonly associated with cratonic peridotites show many lines of isotopic evidence that unequivocally link their protoliths to the hydrosphere (Farquhar et al., 2002; Jacob, 2004; Jacob et al., 1994; Taylor et al., 2005). It seems natural that the cratonic mantle, host to diamonds and imbricated with eclogite, may too have experienced such prior interaction with the hydrosphere. We suggest that the subset of cratonic mantle with low Mg/Si (negative $\Delta\text{Mg/Si}$) is also a result of interaction with seawater at low and high temperatures, albeit in Archean oceans as, accord-

ing to Os isotopic evidence, most cratonic mantle formed principally in the Archean (Pearson, 1999). Cratonic peridotites, especially in southern Africa, are commonly associated with abundant harzburgite low in Ca, a rock type that has no corollary in any other types of mantle lithosphere (Boyd and Gurney, 1982; Boyd et al., 1993; Schulze, 1986, 1995). Schulze explained the unique composition of low-Ca harzburgites in southern African kimberlites as a result of seafloor alteration of their protoliths, which removes Ca before accretion in the craton root (Helmstaedt and Schulze, 1989).

The hallmark high and uniform Mg# (Mg/(Mg + Fe)) of cratonic peridotite olivines is well explained by depletion under high degrees of melting (Bernstein et al., 2007). The same chemical signature, in our hypothesis, could also be accentuated by hydration on the ocean floor and subsequent subduction and dehydration during accretion in the craton root. For example, serpentinization proceeds by the reaction:



If H_2 is liberated in an open-system process (Evans, 1977), then during subduction and dehydration, the following reaction proceeds:



The Fe^{2+} in olivine that was oxidized to Fe^{3+} in magnetite in Reaction 1 is no longer available for reincorporation into olivine by Reaction 2. The $K_{\text{d, Fe-Mg}} \text{Ol-Srp}$ is such that the new olivine preserves a higher Mg# of the preexisting serpentine of the protolith (Evans and Trommsdorff, 1972; Vance and Dungan, 1977), potentially accentuating the overall higher Mg# of olivine in cratonic peridotites.

Weathering or serpentinization of peridotites on the modern seafloor may alter Mg/Si but leaves Mg# unchanged (Boschi et al., 2008). Interaction of Archean cratonic peridotites with seawater, if it occurred, could also have differed from today because of the changing composition of the oceans with time. The composition of the Archean oceans is conjectural, but there is evidence to suggest they were of high pH and anoxic, containing more dissolved Si and orders of magnitude higher solubility for Fe (as Fe^{2+} complexes) (Holland, 1978). Weathering or hydrothermal interaction of peridotite with Archean seawater is expected to have added more Si or leached more Fe than occurs today.

The question remains, however, why does the process of Si enrichment not occur in geologically younger mantle lithosphere? One reason may be due to the history of crustal growth. Mantle lithosphere is the complement (residue) of melt extraction to form the crust. Most esti-

mates of crustal growth show that ~75% of the crust was produced by 2 Ga ago, and some models even have all crust forming in the Archean (Armstrong and Harmon, 1981; Taylor and MacLennan, 1995). Thus, the majority of mantle lithosphere was made in the Precambrian, and most of that was extracted in the Archean. Like the crust, portions of the mantle lithosphere have been recycled in the past two billion years, but the “seafloor” signature would remain only in those residues that have been isolated from recycling by residence in craton roots for the majority of Earth history since their formation in the Archean. This explains the mean Os isotopic age of mantle rocks and the spectrum of Re-depletion ages in osmium-rich alloys from the mantle (Meibom et al., 2002; Pearson, 1999; Pearson et al., 2003, 2007). Finally, our hypothesis is commensurate with the idea that hydration and serpentinization are necessary ingredients for building cratonic mantle roots by permitting the underthrusting of thick, buoyant oceanic lithosphere (Lee et al., 2008). In our model, the Si enrichment occurring before stacking of mantle lithosphere to build a craton root is not exclusive of any subsequent metamorphism that affects cratonic lithosphere over its later billion-year history (Bell et al., 2005).

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