Multidisciplinary constraints on the abundance of diamond and eclogite in the cratonic lithosphere

Joshua M. Garber[1,*], Satish Maurya[2,3], Jean-Alexis Hernandez[4], Megan S. Duncan[5,†], Li Zeng[6], Hongluo L. Zhang[7], Ulrich Faul[8], Catherine McCammon[9], Jean-Paul Montagner[2], Louis Moresi[10], Barbara A. Romanowicz[2,3], Roberta L. Rudnick[1], Lars Stixrude[11]

[1]University of California, Santa Barbara, CA, USA

[2]Institut de Physique du Globe de Paris, Paris, France

[3]University of California, Berkeley, CA, USA

[4]Laboratoire LULI, Ecole Polytechnique, Palaiseau, France

[5]Carnegie Institution of Washington, Geophysical Laboratory, Washington, DC, USA

[6]Harvard University, Cambridge, MA, USA

[7]University of Science and Technology of China, Hefei, China

[8]Massachusetts Institute of Technology, Cambridge, MA, USA

[9]Bayerisches Geoinstitut, University of Bayreuth, Bayreuth, Germany

[10]University of Melbourne, Parkville, Australia

[11]University College London, London, United Kingdom

*current address: Pennsylvania State University, University Park, PA

†current address: University of California, Davis, CA, USA

corresponding author: Joshua M. Garber (jxg1395@psu.edu)

Key Points:

- We used forward modeling to explain high shear-wave velocities in the cratonic lithosphere observed in seismic tomography models.
- Our best estimate for the cause of high shear-wave velocities in the cratonic lithosphere is ≤20 vol.% eclogite and ~2 vol.% diamond.
- Our diamond estimate comports with global carbon mass balance constraints, and could have been implanted over reasonable timescales.

ABSTRACT

Some seismic models derived from tomographic studies indicate elevated shear-wave velocities (≥4.7 km/s) around 120-150 km depth in cratonic lithospheric mantle. These velocities are higher than those of cratonic peridotites, even assuming a cold cratonic geotherm (i.e., 35 mW/m^2 surface heat flux) and accounting for compositional heterogeneity in cratonic peridotite xenoliths and the effects of anelasticity. We reviewed various geophysical and petrologic constraints on the nature of cratonic roots (seismic velocities, lithology/mineralogy, electrical conductivity, and gravity) and explored a range of permissible rock and mineral assemblages that can explain the high seismic velocities. These constraints suggest that diamond and eclogite are the most likely high-V_s candidates to explain the observed velocities, but matching the high shear-wave velocities requires either a large proportion of eclogite (>50 vol.%) or the presence of up to 3 vol.% diamond, with the exact values depending on peridotite and eclogite compositions and the geotherm. Both of these estimates are higher than predicted by observations made on natural samples from kimberlites. However, a combination of ≤20 vol.% eclogite and ~2 vol.% diamond may account for high shear-wave velocities, in proportions consistent with multiple geophysical observables, data from natural samples, and within mass balance constraints for global carbon. Our results further show that cratonic thermal structure need not be significantly cooler than determined from xenolith thermobarometry.

1 INTRODUCTION

Cratons are distinct continental provinces that have been stable since the Archean (e.g., Pearson, 1999; Griffin et al., 2003). They are characterized by thick (≥200 km) lithosphere, as determined from seismic velocities (Jordan, 1975, 1978), surface heat flow (Morgan, 1984; Nyblade & Pollack, 1993; Rudnick et al., 1998; Jaupart & Mareschal, 1999), electrical conductivity (Fullea et al., 2011), and xenolith thermobarometry (Boyd, 1973; Michaut et al., 2007; Michaut et al., 2009; Mather et al., 2011). These "cratonic keels" lack a distinct gravity anomaly and thus appear to be in isostatic equilibrium (Shapiro et al., 1999; Perry et al., 2003), and geochemical evidence from peridotitic xenoliths indicates significant chemical depletion by melt extraction (Lee, 2003; Carlson et al., 2005; Lee et al., 2011) that has had a pronounced effect on their density (Schutt & Lesher, 2006). Such observations have led to the concept of a "tectosphere" (Jordan, 1975, 1978), consisting of thick, neutrally buoyant lithosphere that is chemically distinct from the surrounding asthenospheric mantle. Peridotites in these cratonic keels may even be positively buoyant (Poudjom Djomani et al., 2001), in which case their relatively low densities may be balanced by the presence of denser rocks such as eclogite (Kelly et al., 2003). In addition to isostatic contributions to their long-term stability, cratonic keels are likely drier and thus orders of magnitude more viscous than asthenospheric or sub-oceanic mantle (Pollack, 1986; Hirth et al., 2000; Peslier et al., 2010; Katayama & Korenaga, 2011).

However, cratonic keels are not uniform, as seismic studies have provided evidence for layering and compositional heterogeneity within the cratonic lithosphere. For example, the mid-lithospheric discontinuity [MLD] marks the top of a mid-lithospheric low velocity zone (e.g., Thybo & Perchuć, 1997; Rader et al., 2015) and is also associated with a change in the direction of the fast axis of anisotropy (Yuan & Romanowicz, 2010). Combined with evidence for differences in chemical depletion between the shallower and deeper parts of the lithosphere (e.g., Chesley et al., 1999; Griffin et al., 2003), these data suggest that the cratonic lithosphere consists of several layers that may have been formed or modified by different processes and/or at different times.

These stable cratonic keels exhibit some intriguing seismic properties that are difficult to reconcile with petrologic and geochemical constraints. Notably, as illustrated here, most global-scale and some continental-scale seismic tomographic models show shear-wave velocities (V_s) in excess of 4.7 km/s at depths around 120–170 km in some parts of most cratons at the global scale (Lekic & Romanowicz, 2011; Schaeffer & Lebedev, 2013; Auer et al., 2014; Moulik and Ekström, 2014; French & Romanowicz, 2014; Chang et al., 2015; Debayle et al., 2016) and regional scale (Fichtner et al., 2010; Zhu et al., 2012; Yoshizawa, 2014; Nita et al., 2016). As also illustrated here, these shear-wave velocities are faster than those calculated for any known cratonic peridotite composition, even for the coldest possible cratonic geotherms (Lee, 2003; James et al., 2004) and after accounting for the effect of attenuation on the shear-wave velocities (Bao et al., 2016) (Figure 1; Section 2, below). Explaining these velocity excesses thus requires additional mineral or rock constituents with high shear moduli (G_s) in cratonic keels. Though there is considerable lithologic heterogeneity observed in xenolith suites (Griffin et al., 2002), only a few potential additions exhibit significantly faster shear-wave velocities than cratonic peridotite, including eclogite (predominantly due to garnet) and diamond (cf. compilation in Rader et al., 2015).

Here, we review evidence for high shear-wave velocities in the cratonic mantle lithosphere by comparing results from different seismic tomography studies of different cratons, and confirm the robustness of the high shear-wave velocities with forward modeling of waveforms. We then argue that eclogite and diamond are the most viable candidates that can be added to average peridotite compositions to produce the high shear-wave velocities, and assess heat flow, buoyancy, and electrical conductivity data in concert with phase equilibrium modeling to explore the proportions of eclogite and/or diamond required to produce the high velocities in cratonic keels.

2 STATEMENT OF THE PROBLEM

Many studies have modeled geophysical observations of cratons to understand their compositional and thermal structure (e.g. Afonso et al., 2008; Hieronymus and Goes, 2010; Hirsch et al., 2015; Dalton et al., 2017; Jones et al., 2017; Eeken et al., 2018). These studies reveal disagreement as to how fast cratonic shear-wave velocities are in the depth range ~100-170 km, and whether they can be matched by known cratonic peridotite compositions.

For example, as we will describe further below, many studies that have successfully matched seismologically observed cratonic velocities are based on Rayleigh wave dispersion data, and do not take into account the presence of significant (2-5%) radial anisotropy in the lithosphere. This approach will underestimate the isotropic shear velocity V_{siso}. On the other hand, studies that take into account radial anisotropy, and base their modeling on profiles of V_{siso}, have emphasized that the high shear velocity structure beneath cratons cannot be matched solely by peridotite in the depth range ~100-170 km (e.g. Hirsch et al., 2015). Meanwhile, in some cratons (e.g., South Africa: Jones et al., 2017; India: Maurya et al., 2016), V_s is known to be comparatively low, but these cratons are small – such that “pure path” estimations of velocities (source-station paths contained entirely within the craton region) are more difficult to obtain, especially at the long periods sensitive to the deeper parts of the lithosphere.

In addition, differences in thermodynamic databases, averaging schemes, bulk compositions, steady-state conductive geotherms, and anelastic corrections propagate to significant differences in forward-modeled shear moduli (G), density (ρ), and V_s for cratonic lithologies. The thermodynamic datasets used for some calculations include data and solution models calibrated for crustal conditions (e.g., Holland & Powell, 1998); though the calculated mineral assemblages are similar, equilibrium mineral modes and compositions using such databases may deviate from models calibrated for mantle conditions (e.g., Stixrude & Lithgow-Bertelloni, 2005, 2011). For example, a recent study using the Holland and Powell (1998) thermodynamic dataset yields cratonic peridotite V_s ~0.05 km/s faster than the Stixrude and Lithgow-Bertelloni (2005) thermodynamic dataset, even though the same shear moduli and averaging schemes (Abers & Hacker, 2016) were used for both calculated mineral assemblages (cf. Eeken et al., 2018, their Fig. S2). Further, treating solid-solution endmembers as separable phases (Hacker et al., 2003; Hacker & Abers, 2004; Abers & Hacker, 2016) or using Voigt averages for solution-phase shear moduli, as is done in the thermodynamic modeling software Perple_X (Connolly & Kerrick, 2002; Afonso et al., 2008), yield bulk peridotite V_s that can be an additional ~0.05 km/s too fast relative to thermodynamically justified Reuss averages (Stixrude & Lithgow-Bertelloni, 2005; see Text S1). Finally, corrections for anelastic behavior yield significant differences in forward-modeled V_s for a given bulk composition. These corrections are often opaquely described, are based on outdated parameters, or do not match the frequencies for the seismic models to which the velocities are compared.

Figure 1 shows the cratonic average V_{siso} vs. depth (and 1σ range) from a recent global tomographic model (French & Romanowicz, 2014) and calculated shear-wave velocities for fertile, average, and depleted peridotites (Table S1) along a set of steady-state conductive geotherms that bracket global cratonic peridotite xenolith P-T conditions (Text S1; Fig. 7). The forward-modeled peridotite V_s was calculated using Perple_X free energy minimization software (Connolly, 2009), thermodynamic data, solution models, and shear moduli from Stixrude and Lithgow-Bertelloni (2005, 2011), and temperature, frequency (1 Hz), and grain-size (1 cm) sensitive attenuation corrections from Jackson and Faul (2010). (Calculation details are discussed in Sections 3–4 below and in the Supplementary Information.) Figure 1 illustrates the problem addressed by this study: using state-of-the-art seismological and forward-modeling parameters, even the most depleted peridotites along the coolest possible steady-state conductive geotherms matching xenolith P-T data cannot explain the observed global cratonic average V_s in this tomographic model. Since this tomographic model is arguably on the fast side of the ensemble of available shear velocity models, in the next sections, we consider different seismological models to evaluate a range of representative cratonic V_s profiles, and perform robust forward modeling of different lithologies to try and explain the consistently fast V_s found in some parts of cratons in the depth range 100–170 km.

3 SEISMOLOGICAL CONSTRAINTS

There is significant variability in shear-wave velocity versus depth profiles among different seismic tomographic models, which may be due to a combination of factors: differences in (1) the theoretical assumptions on seismic wave propagation in a 3D earth, (2) accounting (or not) for seismic anisotropy, in particular radial anisotropy, which is known to be prevalent in the upper mantle and in particular in continents (e.g. Dziewonski and Anderson, 1981; Nataf et al., 1984; Montagner and Tanimoto, 1991; Babuska et al., 1999; Gung et al., 2003; Lebedev et al., 2009), (3) how crustal structure is accounted for in the tomographic inversion, and/or (4) how regularization and smoothing affects the resulting velocity-depth profiles. Notably, most previous studies dedicated to explaining seismic observations in terms of mineralogy and associated geotherms have relied either on fitting observed phase velocity dispersion curves for Rayleigh waves (e.g., Darbyshire and Eaton, 2010; Jones et al., 2017; Eeken et al., 2018), or on shear velocity models that were derived from Rayleigh wave dispersion observations (e.g., Bruneton et al., 2008). However, Rayleigh waves are polarized in the vertical plane, thus they are sensitive to V_{sv} rather than V_{siso}. In the lithosphere, the velocity of shear waves polarized horizontally (V_{sh}) is a few percent larger than V_{sv}, which is diagnostic of radial anisotropy and is captured by $\xi>1$, where $\xi=(V_{\mathrm{sh}}/V_{\mathrm{sv}})^2$, the anisotropic parameter to which surface waves are most sensitive. Thus, models based exclusively on Rayleigh waves may underestimate the isotropic shear velocities. For lithospheric studies constrained by surface wave and overtone observations – and in order to access the Voigt average isotropic shear velocity, which can be approximated by $V_{\mathrm{siso}}^2\sim(V_{\mathrm{sh}}^2+2V_{\mathrm{sv}}^2)/3$ – it is necessary to include observations on the transverse component of motion, which contain horizontally polarized Love waves and their overtones. Notably, most studies based on surface wave dispersion data apply approximate crustal corrections, which may introduce biases in the estimation of radial anisotropy in the uppermost mantle (Ferreira et al., 2010; Lekic et al., 2010), and therefore the estimation of isotropic shear velocities. Finally, most models constrained by surface wave data, whether based on dispersion data or seismic waveforms, are based on the "path average approximation", which averages the structure between the source and the receiver in a way that is powerful but not rigorously correct, especially for Love waves (e.g., Megnin and Romanowicz, 1999), and may not allow the accurate resolution of V_{siso} amplitude in regions of small lateral extent, such as the deep roots of some cratons.

Thus, it is necessary to evaluate and quantify the variability across models and determine which type of model provides better fits to observed seismic waveforms, which represent the “raw” seismic data, before any inversion process. For the synthetic calculation of the predicted seismic wavefield in any given model, we take advantage of the spectral element method (SEM), which involves a purely numerical integration of the equations of motion (e.g. Komatitsch and Vilotte, 1998; Komatitsch and Tromp, 1999), makes no theoretical simplifying assumptions, and has been shown to provide accurate predictions of the seismic wavefield in arbitrary 3D earth structure. To assess model fits, we have computed synthetic seismograms in several tomographic models that exhibit large differences in their shear velocity profiles, and compare them to observed 3-component waveforms. The chosen paths, as we will describe below, can be considered as "pure paths", that is, contained entirely within a cratonic region.

To extract shear velocity profiles that are representative of cratonic areas with deep lithospheric roots (> 150 km), one can proceed in several ways. One is based on the geological information on the age of the crust. However, there is not a one to one correspondence between the age of crust and thickness of the lithosphere, with some cratons having clearly lost their deep roots, for example the east China craton (e.g., Chen et al., 2009), or the eastern part of the Superior Craton in north America (Darbyshire et al., 2013; Clouzet et al., 2018). A more objective classification of lithospheric provinces can be done through cluster analysis of upper mantle shear velocity models (Lekic and Romanowicz, 2011). In such an analysis, V_{siso} profiles as a function of depth are first extracted from a given model on a 2° x 2° grid on the earth's surface, for the depth range 50–300 km, and then these profiles are classified into N families of statistically similar velocity profiles using k-means cluster analysis (Macqueen, 1967). The distance between two V_{siso} vectors (i.e. V_{siso} sampled as a function of depth beneath a particular location on the earth's surface) is quantified using the standard L2-norm. As shown in Lekic and Romanowicz (2011) at the global scale, the signature of cratons is clearly distinct from that of other regions for N≥6, and is independent of any geological bias based on crustal ages.

Figure 2 shows the results of such a cluster analysis of the upper mantle isotropic velocity structure (V_{siso}) in four recent global radially anisotropic shear-wave velocity models, developed using different methodologies and datasets. While model SEMUCB_WM1 (French and Romanowicz, 2014) exhibits the fastest velocities, in all these models, the average shear-wave velocity in the craton cluster reaches or exceeds 4.7 km at some depths between 100 and 170 km. The 1σ and 2σ standard deviation bands show that in some areas, V_{siso} even exceeds 4.8 km/s. Two other recent global V_{sv} models (Fig. S1) also show high V_{sv} between 120-160 km, providing lower bounds for V_{siso}. An exception is model ND08 (Nettles and Dziewonski, 2008), for which the cratonic V_{siso} profile shows the highest velocities at shallow depth. We note however, that there are also large differences between the ξ profiles in the cratonic regions of the seven models analyzed (Fig. S2)

We further explored the variability of the shear-wave velocity vs. depth profiles in several cratons by comparing profiles from cluster analyses of global tomographic models with those obtained from continental-scale regional models (Figs. S3–S6). In each case, we used k-means cluster analyses to extract the craton regional boundaries, and determined the average shear-wave velocity within each craton. For the regional models, we used N=4, as this choice of N provides robust regional boundaries and consistent average V_s profiles. The choice of N for the regional models is smaller than for the global models because the regional models lack oceanic regions included in the global models. This analysis shows that there are large differences (up to ±5%) between models in the depth range of interest (100–170 km) in all the cratons shown, both in V_{sv} (for models constructed using only vertical component data) and V_{siso} (for models constructed using three-component data and including radial anisotropy) – likely due to a combination of methodology and datasets considered. While the average cratonic profiles in SEMUCB_WM1 (French and Romanowicz, 2014) are consistently on the fast side, other regional models also exhibit average V_{siso} faster than 4.7 km/s in the relevant depth range (e.g. North America, Scandinavian Shield or Australia).

To further assess the robustness of the fast velocities within some well-studied cratons, Figure 3 shows the geographical distribution of the V_{siso} profiles that are 1-2σ faster than the cratonic average V_{siso} determined by cluster analysis of the corresponding model. These velocity deviations were calculated for the depth range 100-150 km for several models of North America and Australia. Interestingly, the distributions are not random, but delineate contiguous high-V_s regions that are increasingly smaller in size and centered towards the interior of the cratons, depending on lateral resolution of the model. This indicates consistency among some models, in which the fastest velocities correspond to a geographically limited area within the cratons. As shown in Fig. S7 for North America, not all models show such a coherent pattern, and some are clearly smoother, but all except ND08 exhibit extended regions within the craton with velocities exceeding 4.7 km/s in the depth range 100-150 km.

The differences between models may be due to the level of regularization applied in the inversion process, or the theory used for 3D seismic wavefield computations: path-average approximation using normal mode summation in most cases versus more accurate spectral element methods in the case of SEMUCB_WM1 (French & Romanowicz, 2014) and EU30 (Zhu et al., 2012), which show similarly fast V_{siso} in the Scandinavian shield (Fig. S4). The differences may also be due to the way crustal structure is accounted for, which can have an influence on the retrieved mantle structure (Ferreira et al., 2010; Lekić et al., 2010); some groups apply crustal corrections based on existing crustal models (ED16, SL13, ND08), others model crustal effects using spectral element methods on an existing crustal model (AuSREM, EU30), and others fit short-period dispersion data (SEMUCB, EU15). Though determining the cause of the discrepancies is beyond the scope of this study, we note that profiles obtained by simultaneous trans-dimensional Markov chain Monte Carlo modeling of fundamental mode Rayleigh wave dispersion and converted P-to-S phases – i.e., studies in which the crustal and lithospheric structures are simultaneous modeled – also obtain ≥4.7 km/s V_{sv} values at ~150 km depth beneath stations located in the North American craton (e.g., Bodin et al., 2016; Calò et al., 2016).

To determine which models best fit the observed seismograms, we considered the case of the North American craton, which is well sampled by seismic paths and for which it is possible to consider source-station paths that are contained within the cratonic region (i.e., "pure paths"). We compared the predictions of three radially anisotropic models with contrasting properties on two such pure paths (Figure 4): i) a model showing particularly slow velocities in the depth range 100–200 km (ND08: Nettles & Dziewonski, 2008), developed using asymptotic normal mode perturbation theory (the "path-average approximation"); ii) a model showing particularly fast velocities in that same depth range (SEMUCB_WM1: French & Romanowicz, 2014), developed using the spectral element method for 3D wavefield computations; and iii) a model with intermediate V_{siso} values (SGLOBE_rani: Chang et al., 2015), which used the path-average approximation but allowed for crustal thickness perturbations. We chose data from a 2010 earthquake in Baffin Island that was not used in the construction of model SEMUCB_WM1, and two paths that are entirely within the craton (Fig. 4, top) as defined from the cluster analysis for each of the three models. Figure 4 (bottom) shows a comparison of the average and standard deviation of V_{siso} and ξ along each path as a function of depth in the upper mantle, and indicates significant differences between the three models in different depth ranges. ND08 is faster than the other two models down to 90 km depth, and slower in the 120–180 km depth range where SEMUCB_WM1 and SGLOBE_rani are in good agreement. At shallower depths, the differences appear to be compensated by differences in the anisotropic ξ parameter, while in the deeper depth range – of interest in this study – the differences in ξ are less pronounced among the three models.

We further compared the synthetic waveforms predicted for the two modeled paths to observed waveforms on the vertical (Z: sensitive to V_{sv}) and transverse (T: sensitive to V_{sh}) components in two frequency bands (40–80 s and 50–130 s) (Figure 5). The synthetics were computed using RegSEM (Cupillard et al., 2012), which is a continental-scale version of a numerical wavefield simulation code based on the Spectral Element Method (SEM). RegSEM includes the effects of sphericity, radial anisotropy, and attenuation, as well as absorbing lateral boundaries (Perfectly Matched Layers or PMLs) to account for the finite boundaries of the region considered. As Figure 5 illustrates, the synthetic fundamental mode waveforms match the data significantly better for the faster upper mantle model (SEMUCB_WM1) than for the slower one (ND08) in both frequency bands. The vertical component (LHZ) synthetics show that the ND08 V_{sv} model is too slow (by almost a quarter period) at both stations and in both frequency bands, while those for SEMUCB_WM1 match the data significantly better both in phase and amplitude. For the transverse component (LHT) waveforms, the match between observed and synthetics is best for SEMUCB_WM1, although the match in phase is good for ND08 in the early part of the Love wave. SEMUCB_WM1 synthetics match the later parts of the waveforms better than the other models (e.g., after 700 s for station FFC and after 1100 s for station WVT), although they are slightly too slow. The fits for SGLOBE_rani synthetics are better than those for ND08 for the early part of the Rayleigh and Love waveforms, i.e., the longer periods that are sensitive to the depth range of interest here (120–180 km), where SGLOBE and SEMUCB_WM1 agree on the presence of high shear-wave velocities. Still, SGLOBE predicts slightly later arrivals than SEMUCB on the T component in both frequency bands and at both stations.

Because the 3D synthetics computed using SEMUCB_WM1 most accurately predict the seismic wavefield, we infer that – at least in some parts of cratons (see Text S2), and at depths around 150 ± 30 km – V_{siso} is indeed ≥4.65–4.7 km/s. In the following section, we aim to fit these velocity profiles with mineralogy and thermal structure.

4 MINERALOGICAL AND PETROLOGICAL CONSTRAINTS

4.1 Constituents with high shear moduli

To determine the mineral or rock constituents responsible for the observed high shear-wave velocities (V_s), we calculated endmember mineral V_s (Fig. 6) over the pressure-temperature (*P*-*T*) range of interest using the free-energy minimization software Perple_X (Connolly, 2009) with the thermodynamic dataset of Stixrude and Lithgow-Bertelloni (2005, 2011). Figure 6 shows a variety of candidate mineral endmembers that meet or exceed the SEMUCB_WM1 high V_s calculated along cratonic geotherms (Text S1; Fig. 7). Diamond has the fastest V_s (~12 km/s), and other mineral endmembers with high V_s are aluminous orthopyroxene, jadeite, kyanite, corundum, Mg-spinel, and most garnet endmembers – all of which exceed the observed cratonic average V_s. Because garnet, jadeite, and kyanite are all common phases in eclogites, it follows that eclogite may also explain the high observed V_s.

Determining which rocks or minerals are responsible for the high V_s also requires understanding their occurrence in the depth interval of interest. The most direct geochemical knowledge of cratonic mantle lithosphere comes from kimberlite magmas that carry mantle xenoliths and diamonds to Earth's surface. These xenoliths are dominated by peridotites (Nixon et al., 1981; Nixon, 1987; Boyd, 1989; Pearson et al., 2003) that likely originate from the Moho to >200 km depths (Fig. 7). Eclogite xenoliths are typically less abundant than peridotite, but are locally enriched in some kimberlites (e.g., Jericho kimberlite, Slave craton: Kopylova et al., 1999; Roberts Victor kimberlite, Kaapvaal craton: Pearson et al., 2003); the abundance of eclogite xenoliths may not be directly proportional to their actual lithospheric abundances, but could reflect a sampling bias of the particular kimberlite, or preferential preservation of certain lithologies. Using garnet chemistry and abundance from multiple eclogite-rich kimberlite concentrates, Schulze (1989) calculated that eclogites constitute less than 2 vol.% of the cratonic upper mantle; analyses from the Slave craton yielded similar results of <4 vol.% (McLean et al., 2007) and <<10 vol.% eclogite (Griffin et al., 1999a). Other non-peridotitic xenoliths – including pyroxenites and mica- and amphibole-rich rocks – are also observed, but are typically much less abundant than peridotite and eclogite (Boyd & Gurney, 1986; Pearson et al., 2003).

Diamonds are also brought to the surface by kimberlite magmas, and are typically found as xenocrysts in the kimberlite matrix or within eclogite xenoliths. Interestingly, diamonds are less commonly found in peridotitic xenoliths (Boyd & Finnerty, 1980; Jaques et al., 1990; Viljoen et al., 1992; Viljoen et al., 2004; Thomassot et al., 2007), potentially due to the breakdown of diamond-bearing peridotite xenoliths during kimberlite infiltration/metasomatism and ascent (Schulze, 1989; Shirey et al., 2013). Graphite pseudomorphs after diamond have also been found in massif peridotites, e.g., in garnet pyroxenite layers in the Beni Bousera peridotite massif in Morocco (Pearson et al., 1989) and in the Ronda peridotite in southern Spain (Davies et al., 1993). As estimated from thermobarometry of their silicate inclusions, most kimberlitic diamonds (~90 vol.%: Stachel & Harris, 2008) formed in the mantle lithosphere between ~4.3–8.3 GPa ($T \approx$ 1153–1673 K); inclusion suites further indicate that ~64% of diamonds are peridotitic (especially harzburgitic) and ~33% are eclogitic in origin (Stachel & Harris, 2008). Diamond concentrations in kimberlite-borne xenoliths are generally low (<0.0001–0.01 vol.%) (Pearson et al., 2003), but some peridotite and eclogite xenoliths contain concentrations up to 0.02–0.5 vol.% and >2 vol.% diamond, respectively, with some xenoliths exhibiting diamond-rich "seams" (Viljoen et al., 1992; Schulze et al., 1996; Anand et al., 2004; Viljoen et al., 2004).

Though we cannot exclude the presence of additional mineralogical or petrological components or phases responsible for the observed high cratonic shear-wave velocities (see Text S3), we note that i) eclogitic minerals (garnet, clinopyroxene, kyanite) and diamond have the highest V_s of commonly observed cratonic mantle constituents in xenoliths, ii) both are key constituents of erupted mantle material from sub-cratonic lithospheric mantle over the depth range of interest in this study, and iii) their bulk abundances in cratonic lithospheric mantle are loosely constrained.

4.2 Thermodynamic and mixing models: methods summary

Complete method details are contained in Text S1. We used Perple_X Gibbs free energy minimization software (Connolly, 2009) to calculate shear-wave velocity profiles through the cratonic lithosphere for peridotite, eclogite, and diamond, using silicate and oxide thermodynamic data and solution models from Stixrude and Lithgow-Bertelloni (2005, 2011), diamond data from Valdez et al. (2012), and graphite data from Holland and Powell (1998, and references therein). Bulk compositional data for cratonic peridotite and eclogite xenoliths in kimberlites were assembled from PetDB (Lehnert et al., 2000), GEOROC [http://georoc.mpch-mainz.gwdg.de], and additional studies; see Figure S8, Text S1, and Data Set S1. We calculated global average "maximum", "minimum", and "average" peridotite and eclogite compositions – defined relative to MgO – to assess the role of compositional heterogeneity in our results (Table S1); mineral modes for each are shown in Figure S9. Steady-state cratonic geotherms for calculation of equilibrium mineral assemblages were modeled with different surface heat flow

(Q_0) values (35, 40, and 45 mW/m^2) that further bracket global kimberlite xenolith thermobarometry data (Fig. 7); geotherms were calculated after Pollack and Chapman (1977), Chapman (1986), and Rudnick et al. (1998). The mantle adiabat (Fig. 7) was constructed with a potential temperature of ~1623 K and a thermal gradient of 0.4 K/km. Adiabatic shear moduli (G_s) for each bulk-rock composition were assembled in two steps: moduli for each solution phase were corrected to a Reuss average of endmember moduli rather than the raw Perple_X output Voigt average (cf. Fig. S10), after which the bulk-rock G_s was calculated as a Voigt-Reuss-Hill average of all solution phases. Anharmonic V_s (i.e., not corrected for anelasticity) calculated for peridotite, eclogite, and diamond using these methods is shown in Figure 8; 1σ velocity uncertainties are 1–2%, but absolute values depend on mineral assemblage (Stixrude and Lithgow-Bertelloni, 2005). Using the endmember lithologic G_s and density data, and applying a correction for temperature, frequency and grain size-dependent anelastic mineral behavior (Jackson and Faul, 2010; Faul and Jackson, 2015), we forward-modeled mechanical mixtures of each lithology that could explain the average cratonic V_s profiles in models SEMUCB_WM1 (French and Romanowicz, 2014) and SGLOBE_rani (Chang et al., 2015) (Figure 9). Models with diamond include graphite at depths shallower than the diamond-graphite transition.

4.3 Thermodynamic and mixing models: Results

The calculated vol.% eclogite (Fig. 9a) and/or diamond (Fig. 9b) required to match the V_s profiles depends primarily upon i) the peridotite and eclogite compositions and ii) the geotherm. Additionally, though velocities shallower than ~100 km are not affected, accounting for anelastic behavior becomes increasingly important with depth and increasing period, and is more significant for hotter geotherms (e.g., compare anharmonic peridotite in Fig. 8a vs. anelastic peridotite in Fig. 9a). For two-component peridotite + eclogite mixtures, >50 vol.% eclogite is required to match the SEMUCB_WM1 V_s profile when surface heat flow Q_0 = 35 mW/m^2, and even 100 vol.% eclogite cannot match this V_s for Q_0 = 40–45 mW/m^2. Lesser but still significant fractions of eclogite are required to match the highest-V_s portions of the SGLOBE_rani profile. However, these combinations yield density increases relative to peridotite (~3–5%) that violate neutral buoyancy constraints (see below). More reasonable fractions of eclogite (~20 vol.%) added to peridotite produce relatively minimal V_s excesses that do not match the observed high velocities (Fig. 9a) even for the fastest peridotite and eclogite compositions.

In contrast, the forward models that include peridotite + diamond mixtures suggest that ~1–3 vol.% diamond in peridotite can match the observed cratonic average V_s profiles for Q_0 = 35–40 mW/m^2, with a negligible associated density increase (≤0.1%). More significant diamond fractions (~4–6 vol.%) are required for the hottest geotherm (Q_0 = 45 mW/m^2). If 20 vol.% eclogite is included in the diamond + peridotite mixtures, the fraction of diamond required to achieve the cratonic average V_s decreases slightly but is still ~1–3 vol.% for the two cooler geotherms (Fig. 9c). Importantly, the addition of diamond is necessarily constrained to depths at which diamond is stable at the expense of graphite (Day, 2012), i.e., >100 km depth for a 35 mW/m^2 geotherm, increasing to >180 km depth for a 45 mW/m^2 geotherm. This consideration is critical because the G_s of graphite is an order of magnitude lower than that of diamond at ambient conditions (Blakslee et al., 1970; Gillis, 1984), and consequently graphite has a significantly lower V_s (~4.0 km/s: Fig. 6) than diamond. Adding graphite to the mixing models introduces a step-wise, geotherm-dependent increase in calculated V_s; this is mostly due to higher diamond V_s relative to peridotite, because the addition of small graphite volume fractions represents a negligible velocity decrease relative to bulk peridotite (Fig. 9b) and the presence of ~20 vol.% eclogite entirely cancels out this decrease (Fig. 9c). It is unlikely that the graphite-diamond transition is as sharp in nature as it is modeled in Figures 9b-c. For example, reactions between the two phases are kinetically inhibited, such that some experiments have produced coexisting diamond and graphite at relevant *P-T* conditions (e.g., Sokol et al., 2001). The phase boundary itself is unlikely to have remained at a stable depth over geologic time, given evidence for fluctuating cratonic geotherms (e.g., Bell et al., 2003). Further, graphite or diamond shielded from the rock matrix as mineral inclusions at depth (e.g., in olivine or garnet) is relatively insensitive to changes in external *P* and *T* (Zhang et al., 1998). We therefore suggest that the modeled step-wise graphite-diamond transition along each geotherm is more likely to be expressed as a broad region of graphite-diamond coexistence, which would not result in a sharp V_s increase observable e.g. by receiver functions. Alternatively, the diamond fraction may gradually increase with depth (see e.g. emplacement model of Smith et al., 2016).

A final consideration is that of the lithosphere-asthenosphere boundary (LAB), which we define here as the depth at which the conductive geotherm intersects the 1350 °C mantle adiabat (Figs. 7–9). For the coolest geotherm modeled in this study (35 mW/m^2), the LAB is >300 km, which is deeper than the LAB beneath Archean cratons inferred from seismic studies (e.g., Yuan & Romanowicz, 2010); for the hotter geotherms, the LAB decreases to ~230 km (40 mW/m^2) and ~170 km (45 mW/m^2). In the case of the 45 mW/m^2 geotherm, the diamond/graphite boundary is deeper than the LAB; this implies that if diamond is responsible for the observed high V_s, it would be present at significant concentrations in asthenospheric mantle rather than the lithosphere for the hottest cratons.

5 BUOYANCY CONSTRAINTS

An upper limit on the fraction of eclogite and diamond in the cratonic lithosphere arises from the absence of gravity anomalies beneath cratons (Shapiro et al., 1999; Kelly et al., 2003; Perry et al., 2003; Eaton & Claire Perry, 2013), consistent with the "isopycnic" hypothesis (Jordan, 1978) that depletion and melt extraction from the cratonic mantle lithosphere has yielded neutrally buoyant, stable cratonic roots. Other studies have shown that cratonic peridotite xenoliths can be either neutrally or positively buoyant at their calculated equilibration depths (Poudjom Djomani et al., 2001; Kelly et al., 2003; Lee, 2003; James et al., 2004). If the cratonic lithosphere is neutrally buoyant, we can establish the maximum fraction of eclogite that can be hosted by peridotite using density constraints.

Using the Perple_X-calculated densities of each peridotite and eclogite composition along each geotherm (Data Set S3), we calculated the density difference between each lithology and the asthenospheric mantle; for the latter we assumed a pyrolite mantle composition (Workman & Hart, 2005) and used Perple_X to calculate the density along a 1623 K mantle adiabat (Fig. 10a). Over the same depths as the observed high V_s, peridotite ranges from <5% negatively buoyant to <5% positively buoyant, whereas eclogite is negatively buoyant over the entire range; for both lithologies, buoyancy relative to asthenosphere increases with depth. The peridotite transition from negative to positive buoyancy is strongly compositional and temperature dependent, and ranges from ~3–6 GPa (similar to the results of Kelly et al., 2003). We also calculated mixtures of eclogite and peridotite such that the density difference between adiabatic mantle pyrolite and the cratonic peridotite-eclogite mixture at the same depth is zero (i.e., isopycnicity: Fig. 10b). Using this approach, the maximum permitted vol.% eclogite increases with depth from zero to ~20 vol.% (35 mW/m^2), ~40 vol.% (40 mW/m^2), and ~60 vol.% (45 mW/m^2). However, these values assume isopycnicity between eclogite and peridotite at each specific depth interval; if deviations from neutral buoyancy are integrated over the entire lithosphere for each geotherm, we calculate that a maximum ~5–10 vol. % eclogite is permitted from 1 GPa to the lithosphere-asthenosphere boundary. Because we are interested in velocity anomalies that span ~50–75% of cratonic lithospheric thickness, we estimate a maximum permissible eclogite volume fraction of ~20% for our depth interval of interest.

Like eclogite, diamond is denser than peridotite in the depth range 100–200 km, but the small modal diamond fractions calculated in our models (~1–3 vol.%) yield a negligible density increase relative to peridotite alone. Further, calculated diamond densities from 2–8 GPa are equivalent to or lower than eclogite, in which case the maximum calculated eclogite fraction is also an upper bound on the diamond fraction.

6 ELECTRICAL CONDUCTIVITY CONSTRAINTS

Electrical conductivity provides an additional observable that can be tested against the mineralogical models. Variations in natural geomagnetic and geoelectric fields induce subsurface electric currents that can be probed by magnetotelluric sounding, where data can be either forward modeled or inverted to yield electrical conductivity profiles as a function of depth. These techniques have evolved in recent years to the extent that 2D and 3D models can be constructed, providing a more detailed picture of how electrical conductivity varies within cratons (Fig. S12). Cratonic roots are generally more resistive than the surrounding mantle, although some more conductive regions have been identified. For example, high conductivities in the North American Slave and Superior Provinces have been attributed to metasomatism (Chen et al., 2009). For this study, we compared conductivities under cratons at depths corresponding to the high shear-wave velocities (V_s, black outlined box in Fig. S12), neglecting the higher conductivities thought to arise from secondary processes.

The electrical conductivity of a rock assemblage can be calculated based on the results of measurements carried out in the laboratory. We tested viable mineralogical combinations that can explain the fast cratonic V_s – presented in the previous section – by comparing their calculated electrical conductivities with existing electrical conductivity profiles for the cratonic lithosphere obtained from magnetotelluric studies. In order to carry out the calculations, we assumed dry mineral assemblages (based on observations that cratonic lithosphere >150 km depth is dry: e.g., Karato, 1990; Hirth et al., 2000); hydrated conditions would increase electrical conductivity, driving the model in the direction opposite to observations.

For olivine, pyroxenes, and garnet, we employed conductivity laws from Jones et al. (2013) (mainly based on Fullea et al. (2011) for dry conditions) that account for the pressure and temperature dependence of conduction mechanisms, although only small polaron conduction (related to the iron content) is expected at the conditions of the cratonic lithosphere. In some of our mineralogical models, the calculated equilibrium eclogite mineral assemblage includes coesite (cf. Fig. S9). Like diamond, coesite is a wide electronic band-gap insulator, and the electrical conductivity of both minerals is very low and relies on the presence of impurities in their structures. Further, there are no published coesite or diamond conductivity laws that account for the relevant *P-T* ranges. We therefore fixed the electrical conductivity of coesite to zero in our calculations, whereas for diamond we used an Arrhenius model based on conductivity measurements of natural type IIa diamonds at ambient pressure between 673 and 1523 K (Vandersande & Zoltan, 1991). At room temperature, the conductivity of high-purity type IIa diamonds is approximately four orders of magnitude lower than the conductivity of type I diamonds (Vandersande & Zoltan, 1991). Therefore, Type IIa diamond conductivity is a lower bound for natural diamonds in the cratonic lithosphere.

We estimated the electrical conductivities of the average peridotite and average eclogite compositions for both the cold and average geotherms (Q_0 = 35 mW/m^2 and 40 mW/m^2, respectively). We computed both self-consistent estimates (Bruggeman, 1935; Landauer, 1952) and bounds for the conductivities of peridotite and eclogite using a Hashin-Shtrikman (HS) averaging scheme (Hashin & Shtrikman, 1962; Berryman, 1995). The self-consistent estimate σ_{SC} is obtained by iteratively solving:

$$\sum_{i=}^{N} \boldsymbol{x_i} \frac{\sigma_i - \sigma_{SC}}{\sigma_i + \sigma_{SC}} =$$

where x_i and σ_i are the volume fraction and electrical conductivity of component *i*, and *N* is the number of mixture components. The HS lower ($\boldsymbol{\sigma_{HS^-}}$) and upper ($\boldsymbol{\sigma_{HS^+}}$) bounds are given by the following equations:

$$\boldsymbol{\sigma_{HS^-}} = \left(\sum_{i=}^{N} \frac{x_i}{\sigma_i + min(\sigma)}\right)^{-} - \boldsymbol{min(\sigma)}$$

$$\boldsymbol{\sigma_{HS^+}} = \left(\sum_{i=}^{N} \frac{x_i}{\sigma_i + max(\sigma)}\right)^{-} - \boldsymbol{max(\sigma)}$$

The self-consistent estimate can be considered as the electrical conductivity of an average host media composed of spherical inclusions of different components, whereas the HS bounds assume an isotropic polycrystal (Berryman, 1995).

Figure 11a shows endmember peridotite, eclogite, and diamond electrical conductivity profiles; as expected, the endmember conductivity profiles depend strongly on the geotherm considered. Along the average geotherm (Q_0 = 40 mW/m^2), eclogite conductivity increases from 10^{-3} to ~10^{-1} S/m between 4–8 GPa. Over the same pressure range, peridotite and diamond conductivities range between 2x10^{-4}–2x10^{-2} S/m and 10^{-6}–10^{-5} S/m, respectively. The difference between peridotite and eclogite conductivities results from i) differences in iron content and ii) the presence of garnet in the eclogite. For all eclogite, peridotite, and diamond, the cold geotherm (Q_0 = 35 mW/m^2) conductivities are at least one order of magnitude less than those along the average geotherm.

Finally, we calculated the bulk electrical conductivity for the mineralogical mixtures that satisfy the V_s and density constraints (peridotite + 20% eclogite, peridotite + 2% diamond, and peridotite + 20% eclogite + 2% diamond). A further level of averaging was added to the calculation, in that we calculated conductivities with the volume fractions of each eclogite, peridotite, and diamond, and their respective self-consistent conductivity estimates. Figure 11b shows that all of the mineralogical combinations calculated in the previous section to bracket the observed V_s are consistent with electrical conductivity profiles obtained from magnetotelluric measurements of the cratonic lithosphere. All peridotite + 20% eclogite (red curves), peridotite + 2% diamond (blue curves), and peridotite + 20% eclogite + 2% diamond (purple curves) mixtures yield conductivities intermediate between endmember peridotite and eclogite. Even with a difference in electrical conductivity of three to four orders of magnitude between peridotite and diamond, 2 vol.% diamond has a negligible effect on the self-consistent estimates of bulk conductivity for peridotite + diamond mixtures compared to pure peridotite. The most notable effect of adding diamond is the decrease of the low-HS bound by one order of magnitude.

Hence, all of our mineralogical models are compatible with the results of the magnetotelluric measurements. Though the comparison of observed and calculated conductivities does not discriminate between mineralogical models, it provides the important confirmation that the mineralogical models involving eclogite and/or diamond are consistent with geophysical observations. Graphite occurs as isolated grains in peridotite and therefore does not enhance the bulk electrical conductivity (Zhang and Yoshino, 2017). By contrast, the presence of water (hydrogen) enhances electrical conductivity (e.g. Yoshino and Katsura, 2013); this may explain conductivities exceeding the dry conductivities modeled here. Water does not affect seismic velocities (Cline et al., 2018).

7 DISCUSSION

Assuming eclogite and/or diamond are responsible for the high V_s in cratonic roots (Section 4.1 and Text S3), our mineralogical models suggest that ~1–3 vol.% diamond or >>20 vol.% eclogite added to peridotite can independently satisfy the V_s (Fig. 9) and electrical conductivity (Fig. 11) constraints along cold and average cratonic geotherms. The neutral buoyancy constraint additionally suggests that eclogite abundances are unlikely to exceed ~20 vol.% throughout the lithosphere (Fig. 10), requiring instead that peridotite + diamond or peridotite + eclogite + diamond mixtures be invoked to explain the V_s results. Though the electrical conductivity data are compatible with multiple geotherms, the modeled V_s and buoyancy results are highly dependent on the geotherm: cooler geotherms require less eclogite to keep the cratonic root neutrally buoyant, lower estimates of diamond to match the high V_s, a larger diamond stability field, and a deeper lithosphere-asthenosphere boundary (Figs. 7–10). In this study (Fig. 7) and others (Rudnick et al., 1998; Hasterok & Chapman, 2011), cratonic xenolith *P-T* data converge on a global average Q_0 of ~40 mW/m^2 (i.e., a ~200 km thick cratonic lithosphere), suggesting that the intermediate geotherm in our study is the most representative. Considering the V_s-matched mixing models alone, this result requires a *minimum* of ~2 vol.% diamond (Fig. 9b, c). Some studies have found that cratonic shear-wave velocities (V_s) can be matched with peridotite if the cratonic thermal structure is significantly cooler (100–200 K) at any given depth than determined from xenolith thermobarometry (e.g., Eeken et al., 2018). Though there is evidence for non-steady state thermal perturbations in some calculated xenolith *P-T* conditions, it is typically assumed that at least some of the xenolith suite represents steady-state conditions (e.g., Bell et al., 2003). It could be argued that the xenolith-derived geotherms represent past temperatures in the lithospheric roots – which may be cooler at present – but peridotites and eclogite along geotherms significantly cooler than the coldest geotherm calculated in this study (Q_0~35 mW/m^2) may violate electrical conductivity constraints (e.g., Figure 11). Therefore, if the coolest calculated xenolith *P-T* conditions reflect an average steady-state conductive cratonic geotherm, peridotites with 20% eclogite cannot explain the cratonic average V_s, and are compatible with the presence of ~1 vol.% diamond. Kimberlites hosting garnet peridotite xenoliths predominantly occur at the edges of cratons (Figure 2); the resulting average geotherm may therefore not represent temperatures in the seismically fastest portions in the interior.

Our results are also sensitive to composition: the modeled bulk craton V_s increases with more depleted, MgO-rich peridotites and more Al_2O_3-rich, MORB-like eclogites (Fig. 8a, b). More depleted peridotites and more basaltic eclogites would therefore shift the required diamond abundances to slightly lower values, though they still lie between ~1–3 vol.% for the cold and average geotherms. Peridotite and eclogite composition also impacts buoyancy constraints because depleted, MgO-rich peridotites and eclogites are less dense than their fertile, MgO-poor counterparts. Considering the sum of our results, we suggest that a combination of ≤20 vol.% eclogite with ~2 vol.% diamond is the most consistent solution arising from all constraints described here. It has been suggested elsewhere that cratonic V_s may be matched by highly depleted peridotites (e.g., harzburgites or dunites: Afonso et al., 2008; Eeken et al., 2018); these bulk compositions yield faster **V_s than compositionally average cratonic peridotites, but such lithologies do not compose the majority of xenolith compositions sourced from cratonic lithosphere depths (e.g., Griffin et al., 2002).** Even if a forward-modeled V_s for highly depleted peridotites were to match the cratonic average V_s, it is unlikely that cratonic lithosphere is composed solely of depleted peridotites.

In comparing our results to studies of kimberlites and their xenoliths, we note that there are limited constraints on the abundance of eclogite and diamond in cratonic roots (section 3.1). Bulk garnet concentrates from kimberlites suggest a maximum volume fraction of ~2–10 % eclogite in the cratonic mantle lithosphere, even for kimberlites in which the xenolith population is almost entirely eclogite (e.g., Schulze, 1989; Griffin et al., 1999b; McLean et al., 2007). Bulk kimberlite diamond concentrations are typically <0.00002 vol.% (e.g., Bliss, 1992; Pearson et al., 2003) but individual diamond-bearing xenoliths – especially eclogites – may have >2 vol.% diamond (e.g., Viljoen et al., 1992; Anand et al., 2004; Viljoen et al., 2004). An association between diamonds and eclogite is further evident in the abundance of kimberlitic diamonds with eclogitic inclusion suites (~33%), which is higher than eclogite abundance in cratonic lithosphere (~2–20 vol.%: Schulze, 1989; this study). This may be due to diamond formation mechanisms in the lithosphere that are governed by redox interactions between rocks and C-O-H fluids and/or melts: the high redox buffering capacity of Fe-bearing eclogite makes it a particularly fortuitous diamond host (e.g., Luth & Stachel, 2014; Stachel & Luth, 2015). Other diamond formation mechanisms may be favored in harzburgites or dunites, e.g., cooling and decreasing solubility of carbon in a reduced C-O-H fluid (e.g., Luth & Stachel, 2014). Because there are numerous models for the presence of diamond and eclogite in cratonic lithosphere (e.g., Helmstaedt & Schulze, 1989; Shirey & Richardson, 2011; Walter et al., 2011; Palyanov et al., 2013; Weiss et al., 2015), further refinement of seismic models – not only for V_s but also for V_p and density – may help clarify the amount and distribution of eclogite and diamond in cratonic roots, and thus shed light on their origin. Still, the considerations discussed here suggest that the most reasonable solution to the high V_s observation is not endmember eclogite or diamond, but more likely a genetically related and coupled suite of both: the reducing capacity of eclogite in the lithosphere may have produced higher diamond abundances that would not have been present with peridotite alone. Importantly, our proposed cratonic diamond fractions do not constrain the presence of carbon elsewhere in the cratonic lithosphere or in the rest of the mantle. It is possible that deep cratonic lithosphere (i.e., in the diamond stability field) is anomalously carbon-rich compared to shallow cratonic lithosphere due to underplating by subducted MORB (Stachel and Harris 2008; Shirey and Richardson, 2011), or transport from highly reducing deep mantle regions (e.g., Smith et al, 2016). However, given that the diamond fractions proposed here are a small fraction of total mantle carbon (see below), there must be other significant mantle carbon reservoirs, and our study has no implications for their setting or redox state.

Using the ~2 vol.% diamond case, we calculated the resulting amount of carbon that would be contained in cratonic roots. Assuming a cratonic root consisting of an inverted cone with a 1000 km base and 50 km height, the mass of carbon (as diamond) in such a root would be ~10^{18} kg today. If 10 such cratonic roots existed globally, the total mass of cratonic lithospheric mantle carbon would be ~10^{19} kg, equivalent to 2.5 ppm C relative to bulk silicate Earth (BSE) (assuming the BSE is two-thirds the mass of the total Earth). This estimate constitutes ~2% of the BSE carbon (120 ppm: McDonough & Sun, 1995) and ~0.8–12.5% of the "modern" mantle reservoir (0.8–12.5x10^{20} kg C: Dasgupta & Hirschmann, 2010). Using this estimate for total lithosphere-hosted diamond (~10^{19} kg C), and recognizing that rising C-O-H fluids may precipitate ~0.5–2 g C per 100 g fluid from ~200–120 km (Luth & Stachel, 2014), a total C-O-H fluid mass of 5x10^{20}–2x10^{21} kg must have flowed through the cratonic lithosphere to implant this diamond. This fluid concentration is almost certainly an overestimate because it only accounts for diamond precipitation due to oversaturation in the fluid, whereas the redox capacity of eclogite could result in further carbon loss from rising fluids than from cooling and decompression alone. Additionally, such fluid flux would likely be punctuated over Earth history, because diamond inclusion dates are not evenly distributed (e.g., Shirey & Richardson, 2011; Stachel & Luth, 2015).

Recognizing that diamonds form from both mantle carbon and subducted organic carbon (e.g., Ickert et al., 2013; Cartigny et al., 2014), we can further compare the amount of sequestered carbon to estimated modern subduction-related carbon fluxes into the deeper mantle beyond arcs, which are on the order of ~0.0001–52 Mt C/yr ≈ 1x10^5–52x10^9 kg C/yr (Dasgupta & Hirschmann, 2010; Kelemen & Manning, 2015). Figure 12 shows the potential timescales of diamond implantation into the cratonic roots given i) our postulate for the total amount of carbon in cratonic lithosphere of 2 vol.% for 10 cratons (~10^{19} kg C), ii) a mantle carbon flux of 5–50 Mt C/yr, and iii) a range in efficiency of carbon extraction from the mantle to the lithosphere (1–100%), i.e., how much of the deeply subducted C is transferred to the cratonic lithosphere. For the parameter space considered here, the time required to emplace 2 vol.% diamond in 10 cratonic roots is >180 Myr. We acknowledge that carbon ingassing via subduction in the early Earth was likely a less efficient process (e.g., Dasgupta & Hirschmann, 2010), therefore requiring longer timescales to reach 2 vol.% diamond in the cratonic roots. Additionally, the flux of mantle carbon (i.e., not subducted organic carbon) into cratonic lithosphere is unknown. Nevertheless, our calculations show that our proposed abundance of diamond in the lithospheric mantle represents a small fraction of the total terrestrial carbon budget, and could have been transferred to cratonic roots over reasonable geologic timescales.

8 CONCLUSIONS

1. Many global- and continental-scale seismic tomography models exhibit a V_s excess in the deep cratonic lithosphere relative to V_s of cratonic peridotites alone. Synthetic seismograms

obtained in these fast-V_s models (using 3D numerical wavefield computations) provide significantly better fits to the observed seismic waveforms than slower-V_s models that are compatible with peridotitic compositions.

2. Using cratonic geotherms that fit cratonic xenolith *P-T* data, mineralogical and petrological mixing models can reproduce the observed V_s with 1–3 vol.% diamond or >>20 vol.% eclogite. These results are inversely related; more eclogite implies less diamond. Other minerals or chemical components may modulate these results but are less likely than eclogite and/or diamond, as they would have to be present in greater abundances to account for their lower V_s relative to diamond.

3. Buoyancy constraints and the absence of a gravity anomaly suggests that no more than ~20 vol.% eclogite is present in the cratonic lithosphere.

4. Electrical conductivity constraints are compatible with all of the mixing model results. Though diamond is significantly less conductive than either peridotite or eclogite, even 6 vol.% diamond added to either lithology is still compatible with observations.

5. Using the most representative cratonic geotherms (35–40 mW/m^2) and considering all constraints, our best estimate for the permissible volume fractions of eclogite and diamond in the cratonic lithosphere is ≤20 vol.% and ~2 vol.%, respectively.

6. Our estimate for the fraction of eclogite in cratonic lithosphere is higher than (but not significantly different from) estimates derived from kimberlite garnet concentrate chemistry. Likewise, the proposed 1 - 3 vol.% diamond is consistent with i) diamond concentrations in individual xenoliths, especially eclogite, ii) estimates of total carbon in the bulk silicate Earth and mantle, and iii) geologically reasonable timescales over which this carbon could have been implanted in cratonic roots.

Acknowledgments and Data

This research project was initiated at the 2016 Cooperative Institute for Dynamic Earth Research (CIDER) summer program at the University of California Santa Barbara. CIDER is funded as a “Synthesis Center” by the Frontiers of Earth Systems Dynamics (FESD) program (https://www.nsf.gov/geo/fesd/) of the National Science Foundation under grant EAR–1135452. BR acknowledges useful discussions with Steve Richardson, Steve Shirey and Pierre Cartigny. We thank Saskia Goes and Derek Schutt for thorough and constructive reviews, and editorial handling and additional input from Claudio Faccenna.

The authors declare no financial conflicts of interest. Data supporting the conclusions in this paper are available in the cited references, supporting information, and supporting datasets (available in the online version of this paper).

FIGURE CAPTIONS

Figure 1. Comparison of forward-modeled cratonic peridotite V_s for three steady-state conductive geotherms that bracket xenolith *P-T* data (cf. Fig. 7); see Sections 3–4 and Text S1 for methodological details. The data emphasize that cratonic lithosphere requires higher-V_s

constituents unless it is composed solely of ~Fo_{90} dunite *and* has a thermal structure ~100–200 K cooler than measured in xenoliths.

Figure 2. Cluster analysis (N=6) of V_{siso} in radially anisotropic global models SEMUCB_WM1 (French & Romanowicz, 2014), S362ANI+M (Moulik & Ekström, 2014), SAVANI (Auer et al., 2014) and SGLOBE_rani (Chang et al., 2015) in the depth range 60–300 km, revealing cratons with faster than average V_s down to at least 180 km depth (dark blue regions and associated colored velocity profiles on the right of each map). Note that Tibet and Altiplano are singled out as regions of lower than average velocities at shallow depth, but similar to cratons below 200 km depth in model SEMUCB_WM1. Diamonds are found primarily on the edges of cratons, and are shown as white dots (from the compilation of Faure, 2010). In all four models, the three oceanic regions show the age progression of the oceanic lithosphere (yellow to brown), and the cratonic regions (dark blue) have comparable geographic extents. There is more variability in the clustering results for the two other continental clusters that come out of the analysis (green and light blue). The grey shaded panels on the right show the average V_{siso} profile in the cratonic region in each model (white line), surrounded by 1σ, 2σ, and 3σ bands (black to light grey). All models show velocities in excess of 4.7 km/s between 100 and 170 km depth in at least some parts of some cratons (see also Fig. 3). The results of a similar analysis for three other global models is shown in Figure S1.

Figure 3. Distributions of V_{siso} as obtained from cluster analysis in north-America (top) and Australia (bottom). The colors are as in Figure 2, with cratonic regions in dark blue. White dots within the cratonic regions indicate locations where the V_{siso} profile is faster than the average for the cratonic region in the depth range 100-150 km, but within 1σ of that average. Green (resp. red) dots indicate locations where those velocities are between 1σ and 2σ of the average (resp. between 2σ and 3σ). Models shown are global models SEMUCB_WM1 (French and Romanowicz, 2014), SAVANI (Auer et al., 2014), and SGLOBE_rani (Chang et al., 2015) for north America, and SEMUCB_WM1, as well as 2 regional models: AMSAN19 (Fichtner et al., 2009) and AUS14 (Yoshizawa, 2014). Compared to global model SEMUCB_WM1, the regional models in Australia provide a refined view of the cratonic structure, with more localized fast velocities, generally consistent with the geological extent of cratons: in particular Slave and Rae cratons in North America, and Pilbara and Yilgarn cratons in Australia (corresponding tectonic maps are not shown but can be found for example in Whitmeyer and Karlstrom (2007) for North America or Yoshizawa (2014) for Australia).

Figure 4. Comparison of predicted velocity and anisotropy profiles for three radially anisotropic shear-wave velocity models, and for two "pure paths" across the North American craton. *Top*: Regionalized maps of North America from cluster analysis with N=4, for models SEMUCB_WM1 (French and Romanowicz, 2014; left), SGLOBE_rani (Chang et al., 2015; middle) and ND08 (Nettles and Dziewonski, 2008; right), showing the two paths considered. *Bottom*: Comparison of average depth profiles of shear-wave velocity V_{siso} (left), V_{sv} (middle) and anisotropic parameter ξ (right) in the three models, along the paths from the event in Baffin Bay to station FFC (continuous lines) and WVT (dashed lines). Red: SEMUCB_WM1; Blue: ND08; orange: SGLOBE_rani. The grey band shows the range of velocities in model SEMUCB_WM1 for the North American craton cluster (dark blue in top panels), while the standard deviation for the craton clusters in the other two models are indicated by horizontal bars.

Figure 5. Comparison of observed and synthetic waveforms at station FFC (left) and WVT (right) in two different period bands: 40–80 s (top) and 50–130 s (bottom). Each panel shows the comparison on the vertical component (LHZ) and the transverse component (LHT). The data are shown in a black dashed line, the predictions for model SEMUCB_WM1 in red, the predictions for model SGLOBE in green, and those for model ND08 in blue. In all cases, the predictions from model SEMUCB_WM1 generally fit the data best both in phase and in amplitude. Notably, the Z component predictions are systematically too slow for model ND08. The quality of fits for model SGLOBE is intermediate.

Figure 6. Calculated V_s profiles for mantle mineral endmembers and pure phases along cratonic geotherms, plotted with the SEMUCB_WM1 (French & Romanowicz, 2014) average cratonic V_s from the cluster analysis in this study (solid black curve bounded by gray shading and outlined with black dashed lines). The profiles are shown over the entire depth range of interest, i.e., without regard for the stability field of each endmember; see text for modeling details. The colored regions span the V_s for each endmember or phase along the three geotherms shown in Figure 7, with the fastest velocities corresponding to the coolest geotherm. Note i) the difference in scale for the corundum and diamond results and ii) that G_s for a solution phase constructed from these endmembers is a Reuss average, not a Voigt average. Mineral abbreviations are as follows: alm = almandine, maj = majorite, prp = pyrope, gr = grossular, jmaj = Na-majorite, fs = ferrosilite, odi = orthodiopside, en = enstatite, ts = NaAl-orthopyroxene, hed = hedenbergite, di = diopside, cen = clinoenstatite, jd = jadeite, fa = fayalite, fo = forsterite, herc = hercynite, sp = spinel, coe = coesite, ky = kyanite, graph = graphite, cor = corundum, dmd = diamond.

Figure 7. Comparison of pressure-temperature conditions estimated from kimberlite-hosted garnet peridotite xenoliths (Data Set S2), with the modeled geotherm range calculated for different surface heat flows labeled in mW/m^2, and the diamond-graphite phase boundary (Day, 2012). See Text S1 for additional details.

Figure 8. V_s profiles for endmember cratonic peridotite, eclogite, and diamond; see also Data Set S3. 1σ velocity uncertainties are 1–2%, but absolute values depend on mineral assembage (Stixrude and Lithgow-Bertelloni, 2005). **a)** Cratonic peridotite shear-wave velocity (V_s) calculated for three different geotherms (green shaded regions), compared to the average cratonic V_s profiles determined using cluster analyses on seismic tomography models SEMUCB_WM1 (darker gray) and SGLOBE_rani (lighter gray). The velocities are not corrected for temperature and grain-size sensitive anelastic behavior (Faul & Jackson, 2005, 2010) and thus are maxima. As in Fig. 1, shaded regions do not represent 2σ error bounds, but rather identify V_s ranges calculated for different peridotite compositions; lines reflect "average" peridotite compositions (Table S1). "LAB" identifies the lithosphere-asthenosphere boundary (defined in the main text) for the two hottest geotherms shown in each figure, whereas the LAB for the coolest geotherm (35 mW/m^2) is deeper than the extent of the figure. **b)** Cratonic eclogite shear wave velocity profiles for the same geotherms as in (a), uncorrected for anelastic behavior. Note that the broad shaded regions are not symmetric about the "average" eclogite V_s because i) cratonic eclogite compositions are more compositionally heterogeneous than peridotite and ii) alternative bulk compositions pass through *P-T* fields with different mineral assemblages, distinct mineral compositions, and thus variable bulk rock shear moduli. **c)** Diamond V_s for the same geotherms as in (a) and (b), uncorrected for anelastic behavior. Each line is restricted to the diamond stability field specific to that geotherm, after Day (2012) (cf. Fig. 7, this paper); at shallower depths, graphite (V_s~4.0 km/s; cf. Figs. 6 and 9, this paper) is stable.

Figure 9. Results of peridotite ± eclogite ± diamond mixing models. See Text S1 for details of the mixing calculations, Figure 8 for individual, anharmonic lithologic V_s profiles, and Data Sets S4–S9 for anharmonic (elastic) velocities. All profiles are corrected for anelasticity with a 1s period and 1 cm grain size (Jackson & Faul, 2010; Data Set S10), and are shown for the compositional "average" peridotite and eclogite (Fig. S8; Table S1). Calculated V_s profiles from two shear-wave tomographic models are shown in each figure as gray shaded fields; note that these models are referred to 1s periods, i.e., identical to the anelasticity correction. In each panel, mixing results for the coolest geotherm that brackets xenolith thermobarometry (cf. Fig. 7) are shown in blue dashed-dotted lines (35 mW/m^2), results for the intermediate geotherm are shown in green solid lines (40 mW/m^2), and results for the hottest geotherm are shown in red dashed lines (45 mW/m^2). **a)** Two-component peridotite + eclogite mixing model results; only 20 vol.% eclogite is shown to account for neutral buoyancy constraints. **b)** Results of two-component peridotite + graphite/diamond mixing models, with the position of the graphite to diamond transition determined by the geotherm (see text) and data from Day (2012). **c)** Results of three-component peridotite + eclogite + diamond mixing models, showing the effect of combining ~20 vol.% eclogite with ~2 vol.% diamond or graphite.

Figure 10. Summary of peridotite and eclogite constraints on lithospheric density/buoyancy. **a)** Calculated peridotite or eclogite density along a cratonic geotherm relative to asthenosphere along an adiabat at the same depths, for the geotherms and 1623 K mantle adiabat in Figure 7 and the peridotite and eclogite compositions in Table S1. Lines and shading are as in Figure 9. **b)** Maximum vol.% eclogite permitted at each depth in the case that eclogite + peridotite is neutrally buoyant relative to asthenosphere, calculated over regions in which peridotite alone is positively buoyant. Each color is for a distinct geotherm, and each line represents a mechanical mixture of either minimum, average, or maximum peridotite and eclogite composition (a total of nine mixtures per geotherm).

Figure 11. Electrical conductivities calculated from the mineralogical models and comparison with electrical conductivities derived from magnetotelluric regional measurements. **a)** Self-consistent estimates of average cratonic peridotite, average cratonic eclogite, and diamond along the average (continuous lines) and cold (dash-dotted lines) cratonic geotherms corresponding to surface fluxes of 40 mW/m^2 and 35 mW/m^2 respectively. **b)** Peridotite + eclogite (red), peridotite + diamond (blue), and peridotite + 20 vol.% eclogite + diamond (purple) assemblages that match V_s along the average (continuous lines) and cold (dash-dotted lines) cratonic geotherms. Colored areas delimit the Hashin-Shtrikman bounds corresponding the self-consistent estimate of the same color. The dashed grey line encloses the range of electrical conductivity derived from magnetotelluric measurements relevant to the high shear-wave velocity region (see black outlined box in Fig. S12).

Figure 12. Timescales for 2 vol.% diamond implantation into 10 cratonic roots. Each curve represents a different potential mantle carbon flux to the lithosphere; varying the efficiency of carbon transfer from rising C-O-H fluids (x-axis) changes the timescale required to implant our proposed diamond abundance (y-axis).

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